

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

Lin Lin

Department of Mathematics, UC Berkeley;
Lawrence Berkeley National Laboratory

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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.

[Georges and Kotliar, PRB 1992] [Georges et al, Rev. Mod. Phys. 1996]
[Kotliar et al, Rev. Mod. Phys. 2006] [Zgid, Chan, J. Chem. Phys. 2011]
[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

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

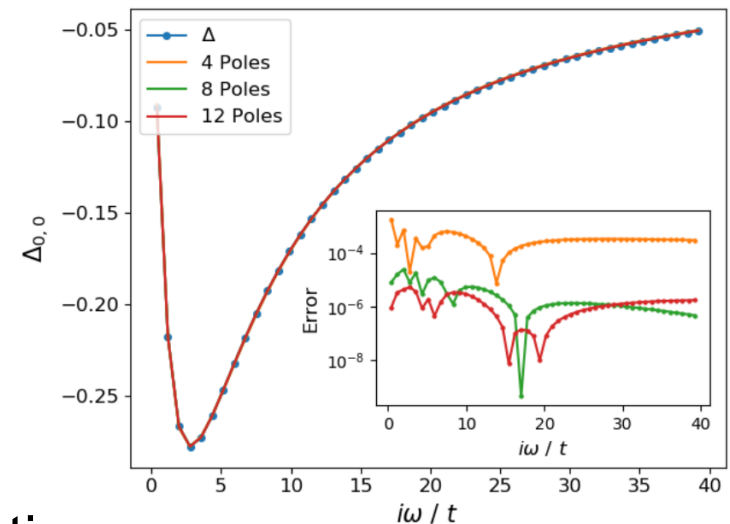
- Replace a large (and possibly infinite) sized bath by a smaller one.
- Closed Hamiltonian system

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

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



(a) Diagonal component of Δ .

Semidefinite relaxation

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

- With a possibly smaller N_p
- Relaxed minimization problem

$$\min_{\lambda_\ell, X_\ell \succeq 0} \mathcal{J}_{SDR}(\{\lambda_\ell, X_\ell\}_{\ell=1}^{N_p})$$

$$\mathcal{J}_{SDR}(\{\lambda_\ell, X_\ell\}_{\ell=1}^{N_p}) = \frac{1}{N_\omega} \sqrt{\sum_{n=1}^{N_\omega} \left\| \Delta(i\omega_n) - \sum_{\ell=1}^{N_p} \frac{X_\ell}{i\omega_n - \lambda_\ell} \right\|_F^2}.$$

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

$$\sum_{\ell=1}^{N_p} \frac{\hat{X}_\ell}{i\omega - \hat{\lambda}_\ell} = \sum_{\ell=1}^{N_p} \sum_{q=1}^{N_c} \frac{U_{\ell,q} U_{\ell,q}^*}{i\omega - \hat{\lambda}_\ell} = \sum_{r=1}^{N_b^{\text{eff}}} \frac{\tilde{V}_r \tilde{V}_r^*}{i\omega - \tilde{\epsilon}_r}$$

Optimization for the poles

- Nested optimization

$$\mathcal{J}_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p}) = \min_{X_\ell \geq 0} \mathcal{J}_{\text{SDR}}(\{\lambda_\ell, X_\ell\}_{\ell=1}^{N_p})$$

- Derivative evaluation

$$\frac{\partial}{\partial \lambda_k} \mathcal{J}_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p}) = \frac{\partial}{\partial \lambda_k} \left(\sqrt{\sum_{j=1}^{N_\omega} \left\| \Delta(i\omega_j) - \sum_{\ell=1}^{N_p} \frac{X_\ell}{i\omega_j - \lambda_\ell} \right\|_F^2} \right) \bigg|_{X_\ell = X'_\ell},$$

$$X'_\ell = \arg \min_{X_\ell \geq 0} \mathcal{J}_{\text{SDR}}(\{\lambda_\ell, X_\ell\}_{\ell=1}^{N_p})$$

Pseudocode

Algorithm 1 Pseudo-code for the nested optimization routine.

Require: Initial guess $\{\lambda_\ell\}_{\ell=1}^{N_p}$

Ensure: $\{\hat{\lambda}_\ell, \hat{X}_\ell\}_{\ell=1}^{N_p}$

while $\|\nabla \mathcal{J}_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p})\| > \epsilon_{\text{tol}}$ **do**

$X'_\ell \leftarrow \arg \min_{X_\ell \geq 0} \mathcal{J}_{\text{SDR}}(\{\hat{\lambda}_\ell, X_\ell\}_{\ell=1}^{N_p})$ by solving the SDR problem

$\lambda_\ell \leftarrow \text{BFGS}(\lambda_\ell, \nabla \mathcal{J}_{\text{pol}}(\{\lambda_\ell\}_{\ell=1}^{N_p}))$

end while

$\hat{\lambda}_\ell \leftarrow \lambda_\ell$

$\hat{X}_\ell \leftarrow \arg \min_{X_\ell \geq 0} \mathcal{J}_{\text{SDR}}(\{\hat{\lambda}_\ell, X_\ell\}_{\ell=1}^{N_p})$

Results

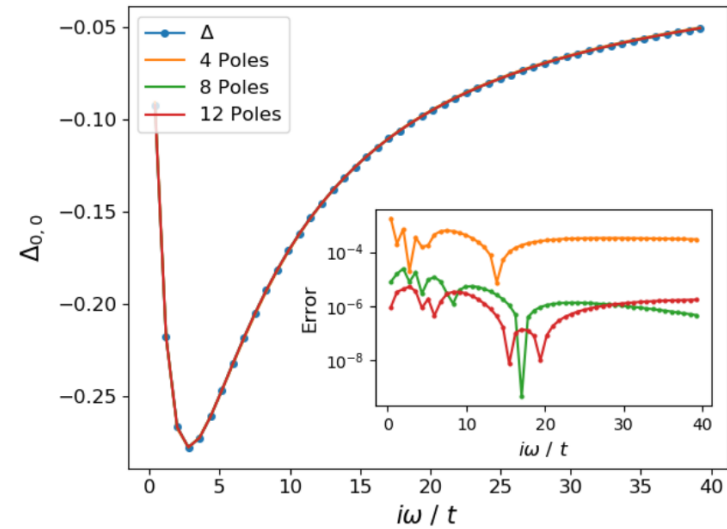
- 2D Hubbard model.
- Impurity size (to be solved by the impurity solver)
- Impurity solver: adaptive sampling configuration algorithm (ASCI)
- Can use other impurity solvers as well, e.g. coordinate descent

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

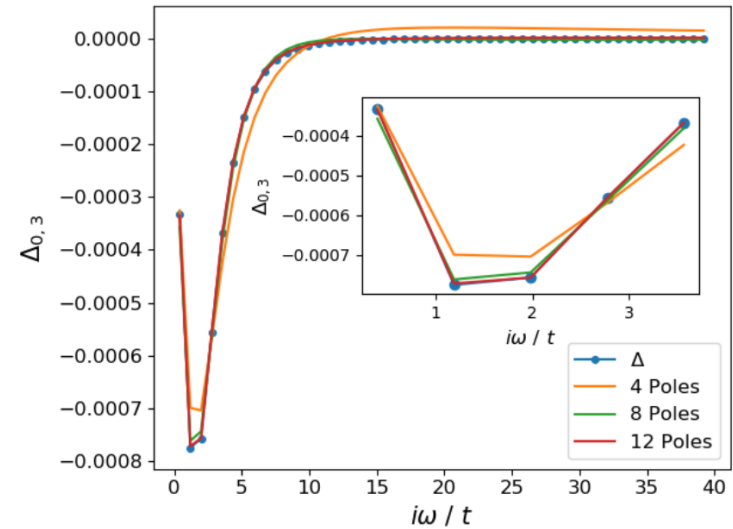
Accuracy

- Fitting error

	(2×2)		(3×3)		(4×4)	
N_p	N_b^{eff}	Error	N_b^{eff}	Error	N_b^{eff}	Error
2	8	2.18e-3	16	2.16e-3	24	1.56e-2
3	12	1.59e-3	24	1.57e-3	36	8.44e-3
4	16	2.88e-4	32	3.97e-4	47	2.09e-3
5	20	2.64e-4	40	2.93e-4	56	1.31e-3
6	24	1.75e-5	48	5.99e-5	63	9.55e-4
7	28	1.14e-5	52	4.04e-5	72	8.37e-4
8	32	2.50e-6	55	5.41e-5	80	7.20e-4
9	35	1.56e-6	62	5.26e-5	86	7.16e-4
10	38	1.50e-6	66	3.08e-5	88	7.07e-4



(a) Diagonal component of Δ .



(b) Off-diagonal component of Δ .

Efficiency: Timings for fitting

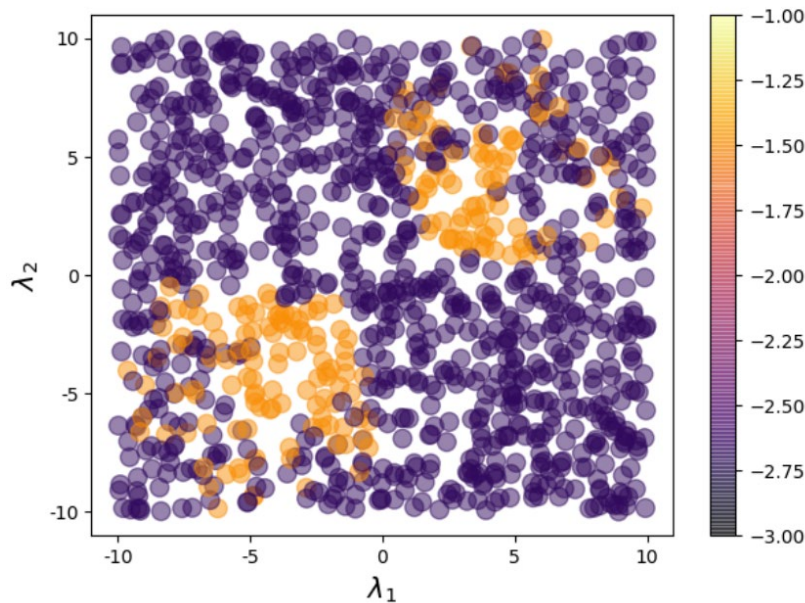
	(2×2)						(3×3)					
	SDR		BFGS		BOBYQA		SDR		BFGS		BOBYQA	
N_p	[s]	Error	[s]	Error	[s]	Error	[s]	Error	[s]	Error	[s]	Error
2	5	2.18e-3	4	1.09e-1	28	2.18e-3	8	2.16e-3	87	9.24e-2	19729	1.17e-2
3	17	1.59e-3	5	9.98e-2	989	1.47e-3	16	1.57e-3	183	2.72e-2	21429	1.15e-2
4	8	2.88e-4	33	4.90e-3	2717	1.29e-4	14	3.97e-4	262	5.10e-3	31889	1.15e-2
5	7	2.64e-4	48	3.30e-3	5535	1.43e-4	24	2.93e-4	363	1.50e-3	40528	1.15e-2
6	27	1.75e-5	56	3.43e-4	1060	1.14e-4	29	5.99e-5	458	1.20e-3	> 24h	—
7	46	1.14e-5	69	2.27e-4	1629	9.64e-5	68	4.04e-5	522	1.10e-3	75922	1.15e-2
8	28	2.50e-6	79	9.92e-5	1790	7.83e-5	102	5.41e-5	617	1.10e-3	> 24h	—
9	96	1.56e-6	87	9.87e-5	2633	5.35e-5	56	5.26e-5	651	1.10e-3	45882	1.15e-2
10	39	1.50e-6	113	6.09e-5	1844	7.41e-5	129	3.08e-5	798	1.10e-3	84600	1.15e-2

BOBYQA (**B**ound **O**ptimization **BY** **Q**uadratic **A**pproximation) [Powell,2009]

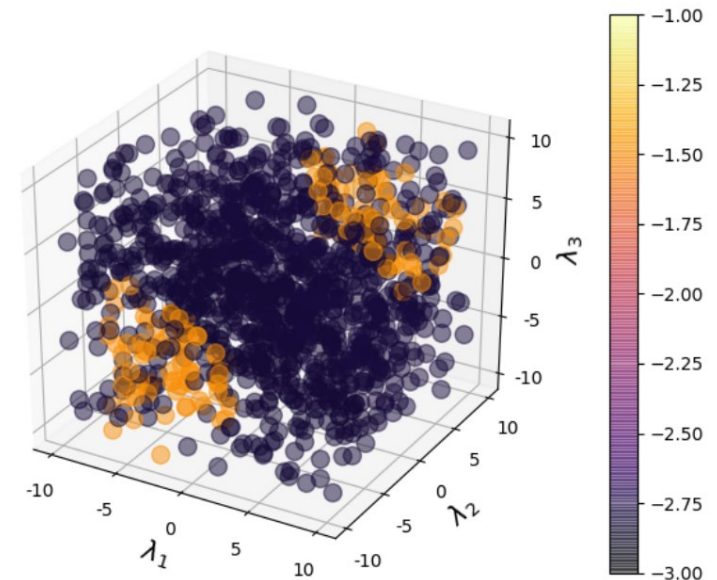
Derivative-free method.

Gradient based method easily trapped in local minima (empirical evidence)

Robustness: fit error vs. initial pole energies



$N_p = 2$



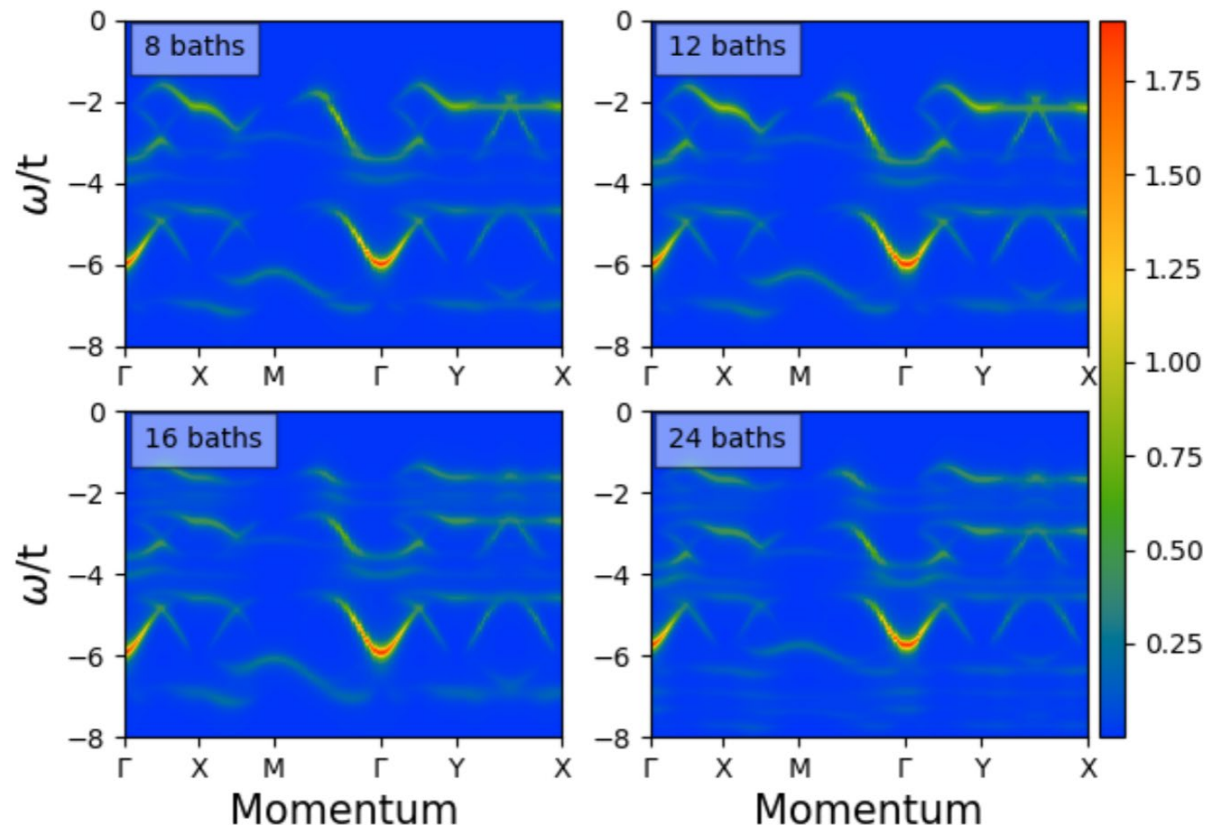
$N_p = 3$

	$N_p = 2$	$N_p = 3$	$N_p = 4$
Nr. of samples	1184	1195	1469
Rate of Success	83%	88% (43% + 45%)	55%
$\max[\sigma(\epsilon_\ell)]$	6e-6	5e-4	2e-4

Initial poles randomly distributed in $[-10, 10]^{\otimes N_p}$.

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 $i(0,2)t$



$$A(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im}(G_{\text{latt}}(\mathbf{k}, \omega))$$

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)



- Hamiltonian

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

- Self energy is a sparse matrix

$$\Sigma(z) = \begin{pmatrix} 0 & & & & \\ & \ddots & & & \\ & & \sigma(z) & & \\ & & & \ddots & \\ & & & & 0 \end{pmatrix}$$

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.

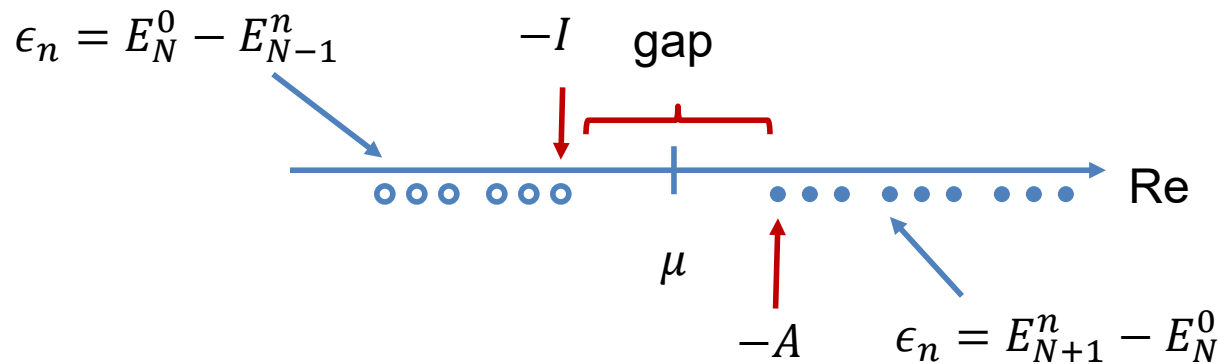
$$\Sigma(z) = \begin{pmatrix} \Sigma_{\text{imp}}(z) & 0 \\ 0 & 0 \end{pmatrix}.$$

- 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)

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



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, Σ^R, Δ^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

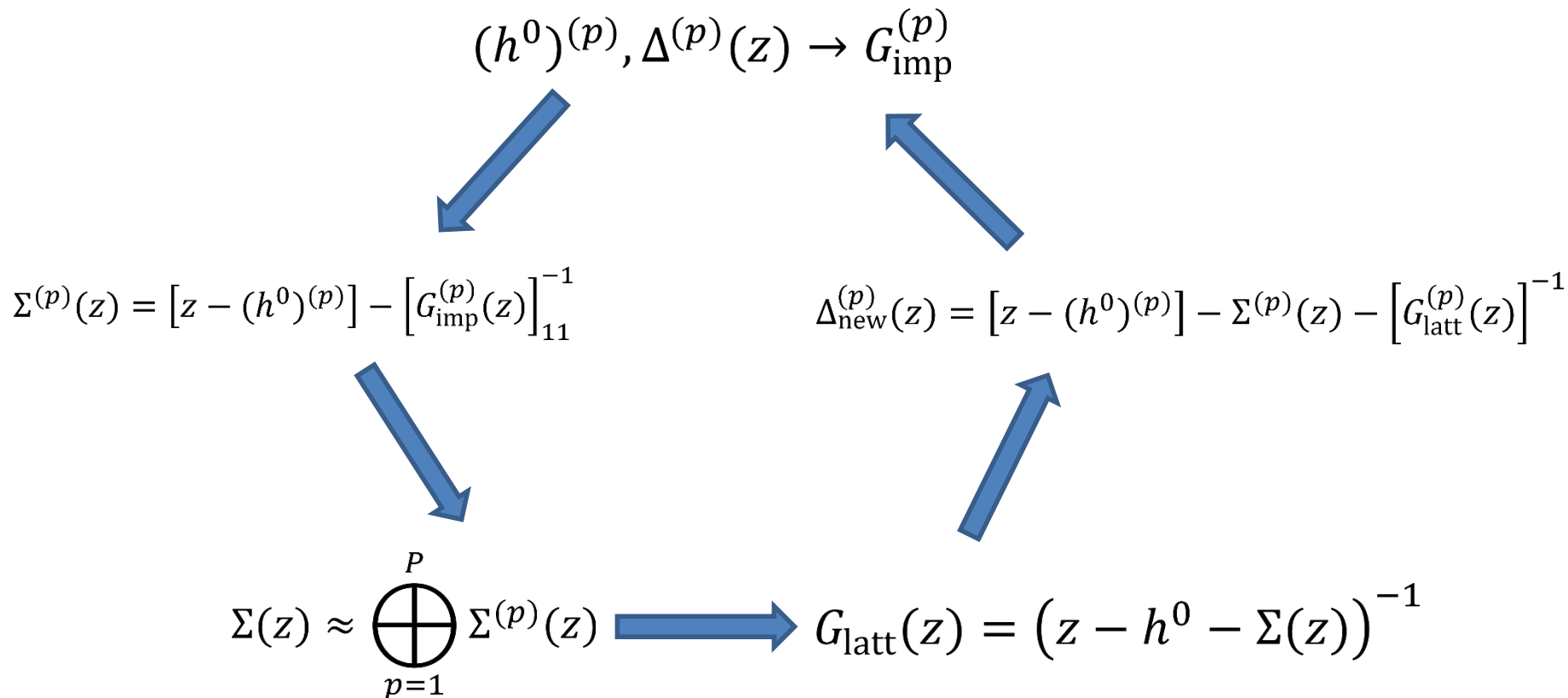
$$\mathcal{S} := \left\{ f : \mathbb{C} \rightarrow \mathbb{C}^{N_b \times N_b} \left| f(z) = \sum_{\ell=1}^K \frac{X_{\ell}}{z - \lambda_{\ell}}, X_{\ell} \succeq 0, \lambda_{\ell} \in \mathbb{R} \right. \right\}$$

- Question: $G(z), \Sigma(z), \Delta(z) \in \mathcal{S}$?

Well-posedness

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

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



Sketch of the proof

- Step 1. Lehmann representation

$$\Delta^{(p)}(z) \in \mathcal{S} \rightarrow G_{\text{imp}}^{(p)} \in \mathcal{S}$$

- Step 2.

$$G_{\text{imp}}^{(p)} \in \mathcal{S} \rightarrow \left(G_{\text{imp}}^{(p)}\right)^{-1}, \Sigma^{(p)} \in ?$$

Proposition

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

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

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

Sketch of the proof

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

- 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 $f \in \mathcal{S}, M \succ 0$.

Suppose that $\bigcap_{k=1}^K \ker(X_k) = \{0\}$. Then $g^{-1} \in \mathcal{S}$

- Hence $\Delta_{\text{new}}^{(p)} \in \mathcal{S}$

Continued fraction representation

- Continued fraction along the iteration

$$\Delta_{\text{new}}^{(p)}(z) \approx D_0 \frac{1}{z - E_0 - D_1 \frac{1}{z - E_1 - D_2 \frac{1}{z - E_2 - \dots} D_2^*}} D_0^*$$

- 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

- Functional perspective, Free energy functional. [Luttinger-Ward 1960]

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

- 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?

PRL **114**, 156402 (2015)

PHYSICAL REVIEW LETTERS

week ending
17 APRIL 2015

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

Evgeny Kozik,^{1,2,*} Michel Ferrero,² and Antoine Georges^{3,2,4}

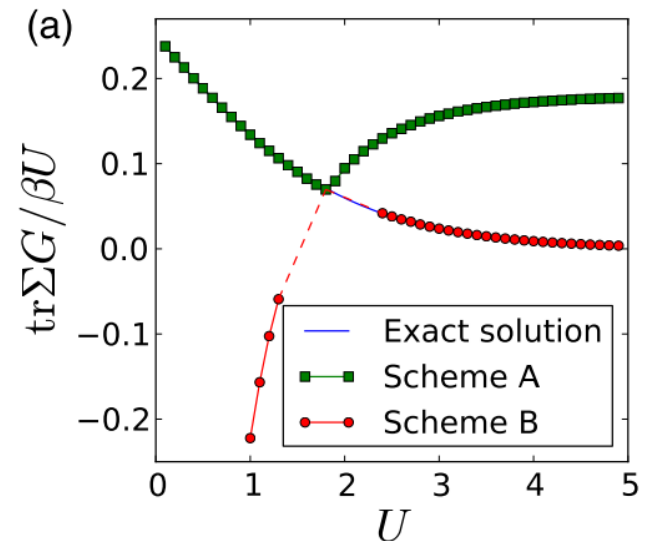
¹Physics Department, King's College London, Strand, London WC2R 2LS, United Kingdom

²Centre de Physique Théorique, Ecole Polytechnique, CNRS, 91128 Palaiseau Cedex, France

³Collège de France, 11 Place Marcelin Berthelot, 75005 Paris, France

⁴DPMC, Université de Genève, 24 Quai Ernest Ansermet, CH-1211 Genève, Suisse

- $\Sigma[G]$ 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

Lin Lin^{a,b,1,2} and Michael Lindsey^{a,1}

^aDepartment of Mathematics, University of California, Berkeley, CA 94720; and ^bComputational Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

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

- Euclidean lattice field model (Gibbs measure)

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

Gibbs measure

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

- Quartic potential (mimicking **Coulomb interaction**)

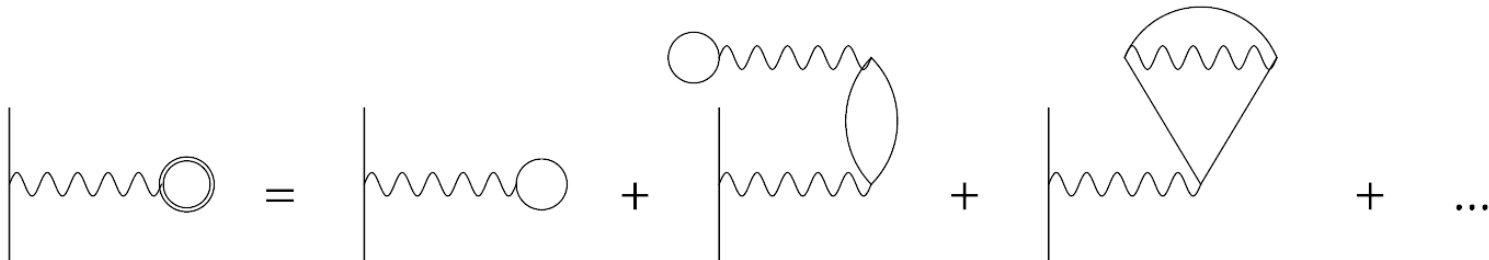
$$U(x) = \frac{1}{8} \sum_{i,j=1}^N v_{ij} x_i^2 x_j^2$$

- Two-point correlator (Green's function)

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

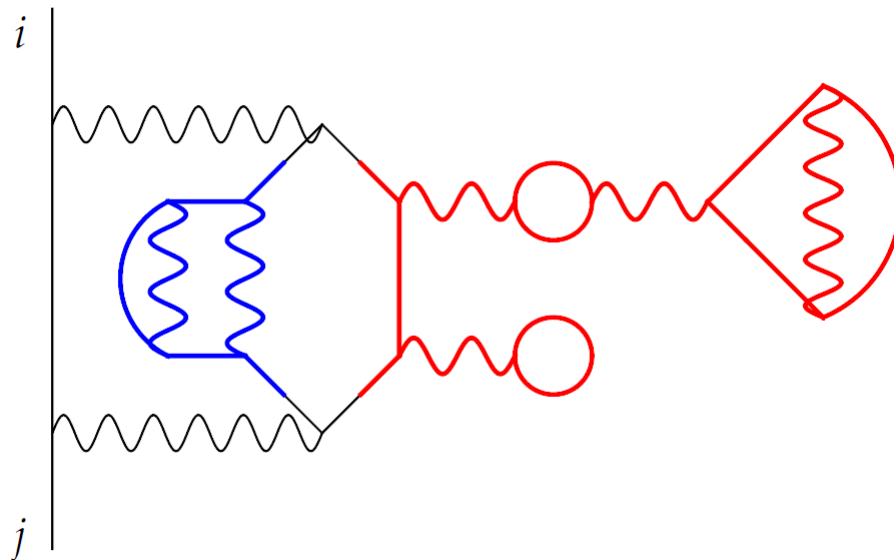
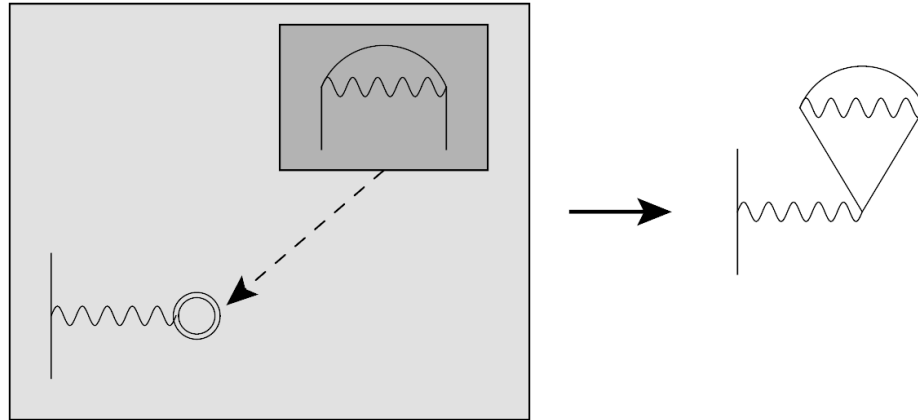
Bold diagrams

- “thin-line” diagram: expand with G_0
- “thick-line” diagram (“bold” diagram): expand with G

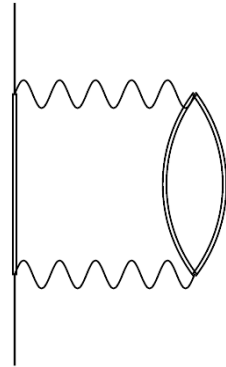


- Partial resummation to **infinite order**

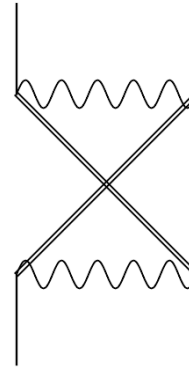
Green's function insertion



Bold diagram simplifies self energy



“ring”



2nd-order EX

- Just 2 terms for 2nd order expansion!
- All bold diagrams are 2-particle irreducible (2PI)
- Luttinger-Ward formalism [Luttinger-Ward 1960]:
perturbative argument

Constrained minimization

Theorem (Variational structure)

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

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)

$\Phi[G]$ extends continuously to ∂S_+^N

Bold diagrams, rigorous statement

Quartic interaction $\varepsilon U(x)$

Theorem (bold diagrammatic expansion)

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

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

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

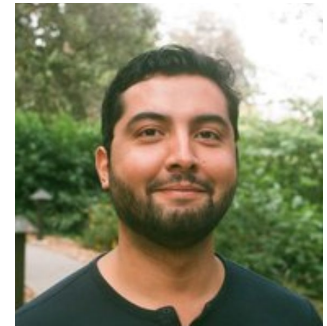
Bold diagram explained!

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] ($d = \infty$): Σ 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

Acknowledgement

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- Reinhold Schneider (TU Berlin)
- Birgitta Whaley (UCB Chem)



DOE Early Career; CAMERA; DOD MURI

Thank you for your attention!