LDA+DMFT
for strongly correlated materials

Alexander Lichtenstein
University of Hamburg

In collaboration with
E. Stepanov, A. Rubtsov, M. Katsnelson

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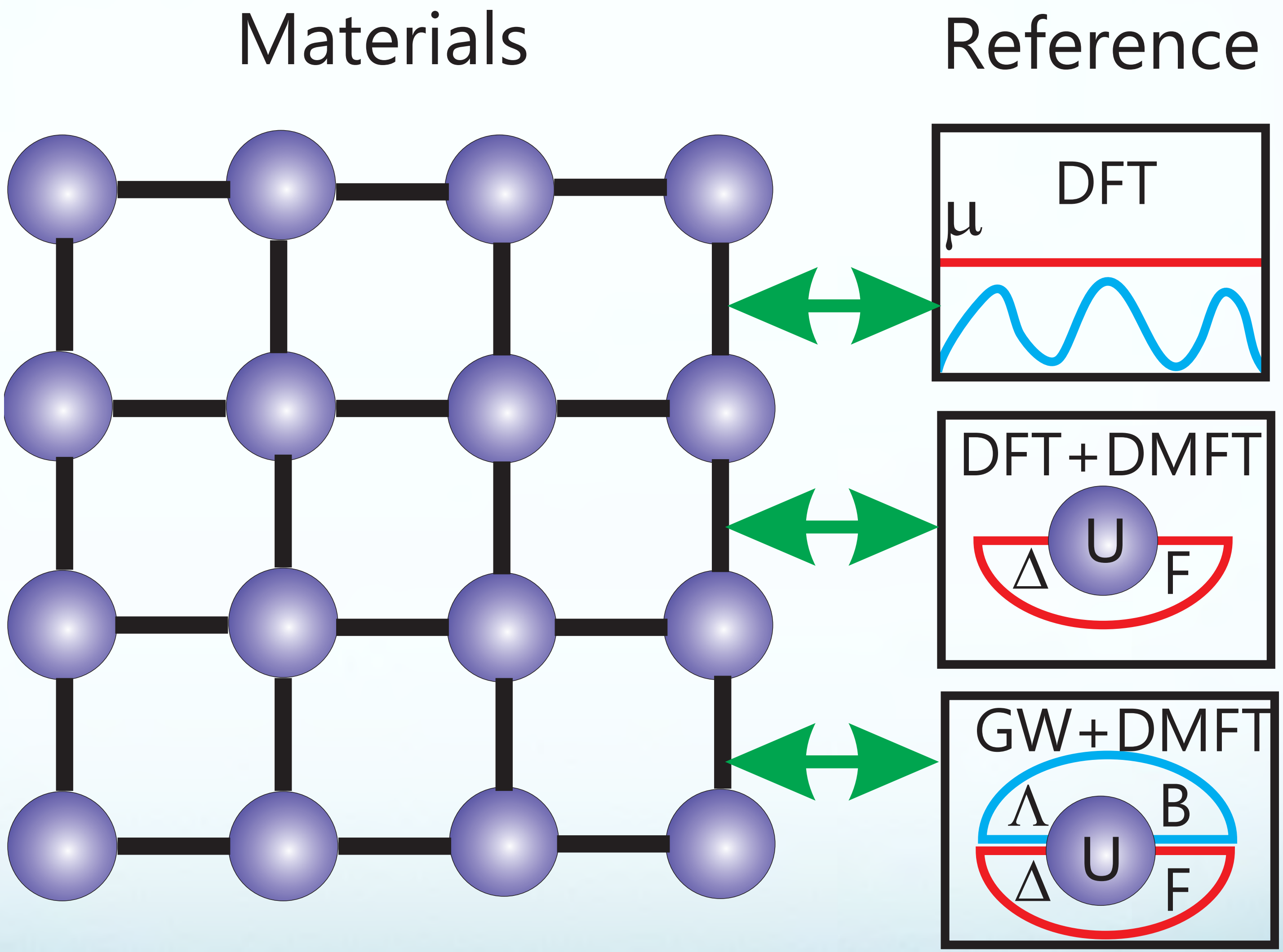
Outline

- Introduction: Reference system
- Path integral for fermions
- Functional approach: BK, DFT
- DF super-perturbation: beyond DMFT
- LDA+DMFT scheme for real materials

Real Materials: Reference Systems



Reference system is important: Archimedes
„Give me the place to stand, and I shall move the earth.“

QM-Alphabet

1-Q

$$\left(-\frac{1}{2}\Delta + V_{eff}(\vec{r})\right)\psi(\vec{r}) = \varepsilon\psi(\vec{r})$$

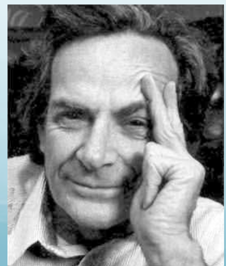
2-Q

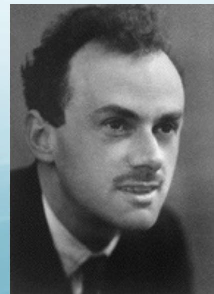
$$\hat{H} = \sum_{ij\sigma} t_{ij}\hat{c}^{+}_{i\sigma}\hat{c}_{j\sigma} + \sum_{i} U\hat{n}_{\uparrow}\hat{n}_{\downarrow}$$

3-PI

$$Z = Sp(e^{-\beta\hat{H}}) = \int D[c^*,c]\, e^{-\int_0^\beta d\tau\left[c^*_\tau \partial_\tau c_\tau + H(c^*_\tau, c_\tau)\right]}$$

Richard Feynman
1948





Paul Dirac
1933

References

- John W. Negele and Henri Orland „Quantum Many-particle Systems“ (Addison Wesley 1988)
- Piers Coleman „Introduction to Many-Body Physics“ (Cambridge Uni Press 2015)
- Eduardo Fradkin „Field Theories of Condensed Matter Physics“ (Cambridge Uni Press 2013)
- Alexander Altland and Ben D. Simons „Condensed Matter Field Theory“ (Cambridge Uni Press 2010)
- Alexey Kamenev „Field Theory of Non-Equilibrium Systems“ (Cambridge Uni Press 2011)

Summary for Fermions $\{\hat{c}_i, \hat{c}_j^{+}\} = \delta_{ij}$

$$\hat{c}_i\,|1\rangle = |0\rangle \qquad \hat{c}_i\,|0\rangle = 0$$

$$\hat{c}_i^{+}\,|0\rangle = |1\rangle \qquad \hat{c}_i^{+}\,|1\rangle = 0$$

Pauli principle

$$\begin{aligned} \hat{c}_i^{+}\hat{c}_i\,|n\rangle &= n_i\,|n\rangle \\ \hat{c}_i^{2} = (\hat{c}_i^{+})^2 &= 0\,. \end{aligned}$$

Fermionic coherent states $|c\rangle$

$$\hat{c}_i\,|c\rangle = c_i\,|c\rangle$$

Left-eigenbasis has only annihilation operator - bounded from the bottom:

$$\hat{c}_i\,|0\rangle = 0\,|0\rangle$$

Grassmann numbers c_i

F. A. Berezin: Method of Second Quantization (Academic Press , New York, 1966)

Eigenvalues of coheren states

$$\begin{aligned} c_i c_j &= -c_j c_i \\ c_i^2 &= 0 \end{aligned}$$

Exact representation

$$|c\rangle = e^{-\sum_i c_i \hat{c}_i^+} |0\rangle$$

Proof for one fermionic states

$$\hat{c}\,|c\rangle = \hat{c}(1 - c\hat{c}^+)\,|0\rangle = \hat{c}\,(|0\rangle - c\,|1\rangle) = -\hat{c}c\,|1\rangle = c\,|0\rangle = c\,|c\rangle$$

Left coherent state ⟨c| :

$$\langle c|\,\hat{c}_i^+ = \langle c|\,c_i^*$$

$$\langle c| = \langle 0|\,e^{-\sum_i \hat{c}_i c_i^*}$$

general function of two Grassmann variables

$$f(c^*, c) = f_{00} + f_{10}c^* + f_{01}c + f_{11}c^*c$$

Grassmann calculus

Formal definition of derivative

$$\frac{\partial c_i}{\partial c_j} = \delta_{ij}$$

Due to anti-commutation rule:

$$\frac{\partial}{\partial c_2} c_1 c_2 = -c_1$$

Example: $f(c^*, c) = f_{00} + f_{10} c^* + f_{01} c + f_{11} c^* c$

$$\frac{\partial}{\partial c^*} \frac{\partial}{\partial c} f(c^*, c) = \frac{\partial}{\partial c^*} (f_{01} - f_{11} c^*) = -f_{11} = -\frac{\partial}{\partial c} \frac{\partial}{\partial c^*} f(c^*, c)$$

Formal definition of integration over Grassmann variables

$$\int ... dc \rightarrow \frac{\partial}{\partial c} ...$$

$$\int 1 dc = 0 \qquad \int c dc = 1$$

Resolution of unity operator

Overlap of any two coherent fermionic states $\langle c|c\rangle = e^{\sum_i c_i^* c_i}$

Proof for single particle

$$\langle c|c\rangle = (\langle 0| - \langle 1|\, c^*)\,(|0\rangle - c\,|1\rangle) = 1 + c^*c = e^{c^*c}$$

Unity operator

$$\int dc^* \int dc\ e^{-\sum_i c_i^* c_i}\,|c\rangle\,\langle c| = \hat{1} = \int\int dc^* dc\,\frac{|c\rangle\,\langle c|}{\langle c|c\rangle}$$

Proof for single particle

$$\int\int dc^* dc\ e^{-c^*c}\,|c\rangle\,\langle c| = \int\int dc^* dc\,(1 - c^*c)\,(|0\rangle - c\,|1\rangle)\,(\langle 0| - \langle 1|\,c^*) =$$

$$-\int\int dc^* dc\ c^*c\,(|0\rangle\,\langle 0| + |1\rangle\,\langle 1|) = \sum_n |n\rangle\,\langle n| = \hat{1}$$

Trace Formula

Matrix elements of normally ordered operators

$$\langle c^*|\,\hat{H}(\hat{c}^+,\hat{c})\,|c\rangle = H(c^*,c)\,\langle c^*|c\rangle = H(c^*,c)\,e^{\sum_i c_i^* c_i}$$

Trace of fermionic operators in normal order

$$\begin{aligned} Tr\left(\widehat{O}\right) &= \sum_{n=0,1} \langle n|\,\widehat{O}\,|n\rangle = \sum_{n=0,1} \int\!\!\int dc^* dc\; e^{-c^* c}\,\langle n|\,c\rangle\,\langle c|\,\widehat{O}\,|n\rangle = \\ &= \int\!\!\int dc^* dc\; e^{-c^* c} \sum_{n=0,1} \langle -c|\,\widehat{O}\,|n\rangle\,\langle n|\,c\rangle = \int\!\!\int dc^* dc\; e^{-c^* c}\,\langle -c|\,\widehat{O}\,|c\rangle \end{aligned}$$

„Minus“ fermionic sign due to commutations:

$$\langle n|c\rangle\,\langle c|n\rangle = \langle -c|n\rangle\,\langle n|c\rangle$$

Mapping: $\left(\hat{c}_i^+,\hat{c}_i\right) \to \left(c_i^*, c_i\right)$

Partition function

Grand-canonical quantum ensemble $H = \widehat{H} - \mu \widehat{N}$

N-slices Trotter decomposition [0,β)

$$\tau_n = n\Delta\tau = n\beta/N \; (n = 1, ..., N). \qquad e^{-\beta H} = \lim_{N\to\infty} (e^{-\Delta\tau H})^N$$

Insert N-times the resolution of unity:

$$\begin{aligned} Z &= Tr\left[e^{-\beta H}\right] = \int\int dc^* dc e^{-c^* c} \langle -c| e^{-\beta H} |c\rangle \\ &= \int \Pi_{n=1}^N dc_n^* dc_n e^{-\sum_n c_n^* c_n} \langle c_N| e^{-\Delta\tau H} |c_{N-1}\rangle \langle c_{N-1}| e^{-\Delta\tau H} |c_{N-2}\rangle \dots \langle c_1| e^{-\Delta\tau H} |c_0\rangle \\ &= \int \Pi_{n=1}^N dc_n^* dc_n e^{-\Delta\tau \sum_{n=1}^N [c_n^*(c_n - c_{n-1})/\Delta\tau + H(c_n^*, c_{n-1})]} \end{aligned}$$

In continuum limit (N → ∞)

$$\Delta\tau \sum_{n=1}^N \dots \mapsto \int_0^\beta d\tau \dots$$

$$\frac{c_n - c_{n-1}}{\Delta\tau} \mapsto \partial_\tau$$

$$\Pi_{n=0}^{N-1} dc_n^* dc_n \mapsto D\left[c^*, c\right]$$

$$Z = \int D\left[c^*, c\right] e^{-\int_0^\beta d\tau [c^*(\tau)\partial_\tau c(\tau) + H(c^*(\tau), c(\tau))]}$$

Antiperiodic boundary condition $c(\beta) = -c(0), \qquad c^*(\beta) = -c^*(0)$

Gaussian path integral

Non-interacting "quadratic" fermionic action

$$Z_0\left[J^*,J\right]=\int D\left[c^*c\right]\,e^{-\sum_{i,j=1}^N c_i^* M_{ij} c_j+\sum_{i=1}^N\left(c_i^* J_i+J_i^* c_i\right)}=\det\left[M\right]e^{-\sum_{i,j=1}^N J_i^*(M^{-1})_{ij}J_j}$$

Hint for proof: $$e^{-\sum_{i,j=1}^N c_i^* M_{ij} c_j}=\frac{1}{N!}\left(-\sum_{i,j=1}^N c_i^* M_{ij} c_j\right)^N$$

Exercise for N=1 and 2: $$\int D\left[c^*c\right]e^{-c_1^* M_{11} c_1}=\int D\left[c^*c\right]\left(-c_1^* M_{11} c_1\right)=M_{11}=\det M$$

$$\int D\left[c^*c\right]e^{-c_1^* M_{11}c_1-c_1^* M_{12}c_1-c_2^* M_{21}c_1-c_2^* M_{22}c_2}=$$

$$\frac{1}{2!}\int D\left[c^*c\right]\left(-c_1^* M_{11}c_1-c_1^* M_{12}c_1-c_2^* M_{21}c_1-c_2^* M_{22}c_2\right)^2=M_{11}M_{22}-M_{12}M_{21}=\det M$$

Shift of Grassmann variable: $$c^*Mc-c^*J-J^*c=\left(c^*-J^*M^{-1}\right)M\left(c-M^{-1}J\right)-J^*M^{-1}J$$

correlation functions for a non- interaction action (Wick-theorem)

$$\begin{aligned}\langle c_i c_j^*\rangle_0 &= -\frac{1}{Z_0}\frac{\delta^2 Z_0\left[J^*,J\right]}{\delta J_i^*\,\delta J_j}\Big|_{J=0}=M_{ij}^{-1}\\ \langle c_i c_j c_k^* c_l^*\rangle_0 &= \frac{1}{Z_0}\frac{\delta^4 Z_0\left[J^*,J\right]}{\delta J_i^*\delta J_j^*\delta J_l\delta J_k}\Big|_{J=0}=M_{il}^{-1}M_{jk}^{-1}-M_{ik}^{-1}M_{jl}^{-1}\end{aligned}$$

Path Integral for Everything

Euclidean action

$$
\begin{aligned}
Z &= \int \mathcal{D}[c^*, c] e^{-S} \\
S &= \sum_{12} c_1^* \left(\partial_\tau + t_{12}\right) c_2 + \frac{1}{4} \sum_{1234} c_1^* c_2^* \, U_{1234} \, c_4 c_3
\end{aligned}
$$

One- and two-electron matrix elements:

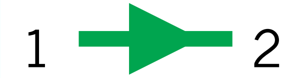
$$
\begin{aligned}
t_{12} &= \int d\mathbf{r}\, \phi_1^*(\mathbf{r}) \left(-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \mu\right) \phi_2(\mathbf{r}) \\
U_{1234} &= \int d\mathbf{r} \int d\mathbf{r}'\, \phi_1^*(\mathbf{r})\phi_2^*(\mathbf{r}')\, U(\mathbf{r}-\mathbf{r}')\, \phi_3(\mathbf{r})\phi_4(\mathbf{r}')
\end{aligned}
$$

Shot notation:

$$
\sum_1 \ldots \equiv \sum_{im} \int d\tau \ldots
$$

One- and Two-particle Green Functions

One-particle Green function



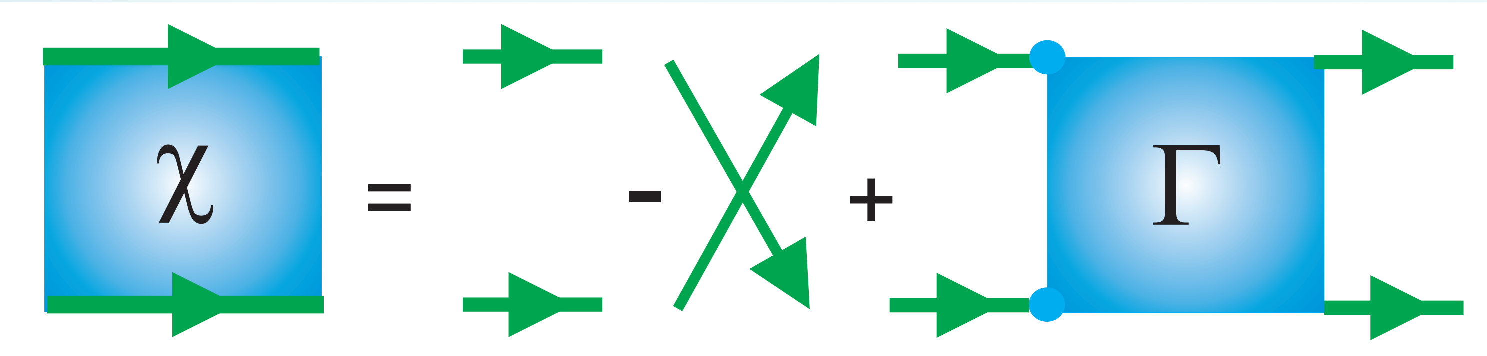
$$G_{12} = -\langle c_1 c_2^* \rangle_S = -\frac{1}{Z} \int \mathcal{D}[c^*, c]\, c_1 c_2^*\, e^{-S}$$

Two-particle Green function (generalized susceptibilities)

$$\chi_{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_S = \frac{1}{Z} \int \mathcal{D}[c^*, c]\, c_1 c_2 c_3^* c_4^*\, e^{-S}$$

Vertex function:

$$X_{1234} = G_{14} G_{23} - G_{13} G_{24} + \sum_{1'2'3'4'} G_{11'} G_{22'} \mathit{\Gamma}_{1'2'3'4'} G_{3'3} G_{4'4}$$



Baym-Kadanoff-Luttinger-Ward functional

Source term

$$S[J] = S + \sum_{ij} c_i^* J_{ij} c_j$$

Partition function and Free-energy:

$$Z[J] = e^{-F[J]} = \int \mathcal{D}[c^*, c]\, e^{-S[J]}$$

Legendre transforming from J to G:

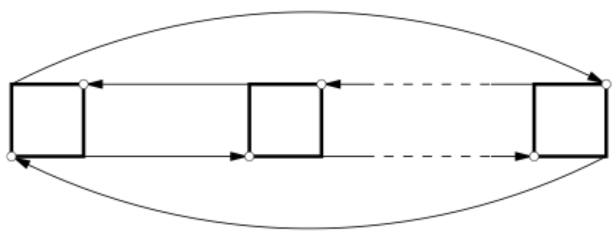
$$F[G] = F[J] - \mathrm{Tr}(JG)$$

$$G_{12} = \frac{1}{Z[J]} \frac{\delta Z[J]}{\delta J_{12}}\bigg|_{J=0} = \frac{\delta F[J]}{\delta J_{12}}\bigg|_{J=0}$$

Decomposition into the single particle part and correlated part

$$F[G] = \mathrm{Tr}\ln G - \mathrm{Tr}\,(\Sigma G) + \Phi[G]$$

$$\Phi[G] = \sum_i$$



Problems with BKLW-functional

Physical and unphysical regimes of self-consistent many-body perturbation theory

K. Van Houcke, E. Kozik, R. Rossi, Y. Deng, F. Werner, arXiv:2102.04508

$$G^{-1} = G_0^{-1} - \Sigma \quad \Longleftrightarrow \quad \Sigma = \Sigma_{\text{bold}}[G] = \sum_{n=1}^{\infty} \Sigma^{(n)}_{\text{bold}}[G]$$

Toy model – Hubbard atom: H= $-\mu \sum_s n_s + U n_\uparrow n_\downarrow$ $\quad s \in \{\uparrow, \downarrow\}$

Partition function, Action and Grteen Function in Grassmann integral over φ_s and $\bar{\varphi}_s$

$$Z = \int \left(\prod_s d\varphi_s d\bar{\varphi}_s\right) e^{-S[\bar{\varphi}_s, \varphi_s]}$$

Exact results for $\mu > 0$ with rescaling $g := G\sqrt{|U|}, \quad \sigma := \Sigma/\sqrt{|U|}.$

$$S[\bar{\varphi}_s, \varphi_s] = -\mu \sum_s \bar{\varphi}_s \varphi_s + U \bar{\varphi}_\uparrow \varphi_\uparrow \bar{\varphi}_\downarrow \varphi_\downarrow \quad \Rightarrow \quad \sigma_{\text{bold}}(g) = \sum_{n=1}^{\infty} \sigma^{(n)}_{\text{bold}}(g)$$

$$\sigma^{(n)}_{\text{bold}}(g) = a_n (-1)^n g^{2n-1}$$

$$a_n = \frac{(-1)^{n-1}(2n-2)!}{n!(n-1)!}$$

$$G = -\frac{1}{Z} \int \left(\prod_s d\varphi_s d\bar{\varphi}_s\right) e^{-S[\bar{\varphi}_s, \varphi_s]} \varphi_s \bar{\varphi}_s$$

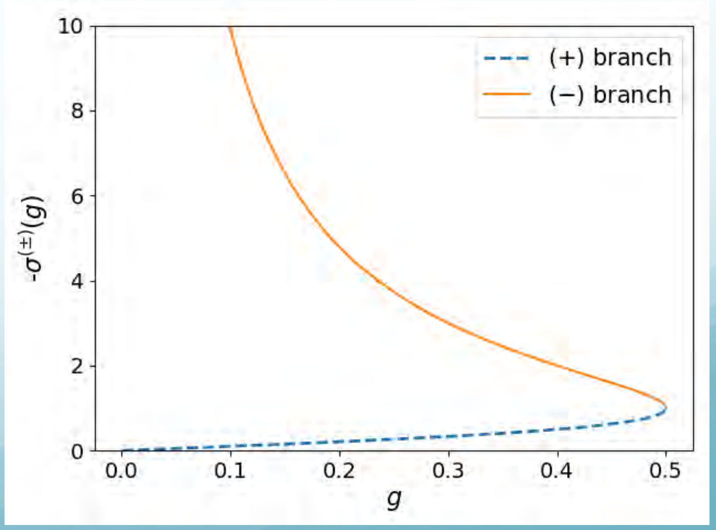
Exact self-energy and propagator

$$\begin{aligned} \sigma_{\text{exact}}(g_0) &= -g_0 \\ g_{\text{exact}}(g_0) &= \frac{g_0}{1+g_0^2} \end{aligned} \quad \Rightarrow \quad \sigma^{(\pm)}(g) = \frac{-1 \pm \sqrt{1-4g^2}}{2g}$$

$$g_0 := \sqrt{|U|}\, G_0 = \sqrt{|U|}/\mu.$$

Physical branch (+) $\quad |U| < \mu^2$

Unphysical branch (-) $\quad |U| > \mu^2$



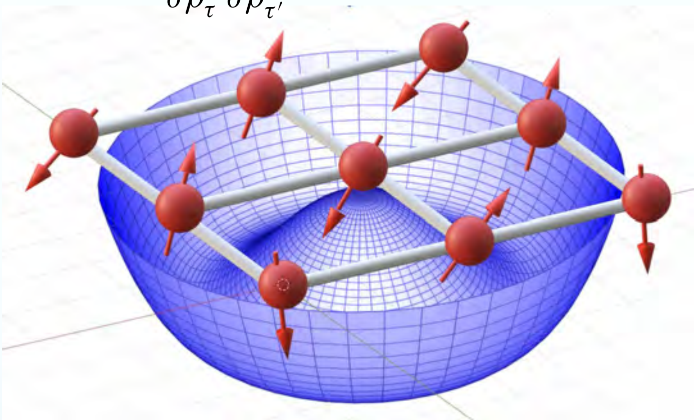
Emergent Magnetic Moment

$$\mathcal{S}_{\rm latt} = \int_0^\beta d\tau \Big\{ - \sum_{ij,\sigma\sigma'} c^*_{i\tau\sigma}\left[\delta_{ij}\delta_{\sigma\sigma'}(-\partial_\tau + \mu) - \varepsilon^{\sigma\sigma'}_{ij}\right] c_{j\tau\sigma'} + \sum_{i,\sigma\sigma'} U n_{i\tau\uparrow} n_{i\tau\downarrow} + \frac{1}{2}\sum_{ij,\varsigma} \rho^{\varsigma}_{i\tau} V^{\varsigma}_{ij} \rho^{\varsigma}_{j\tau} \Big\} \quad (1)$$

Correlated electrons lattice model

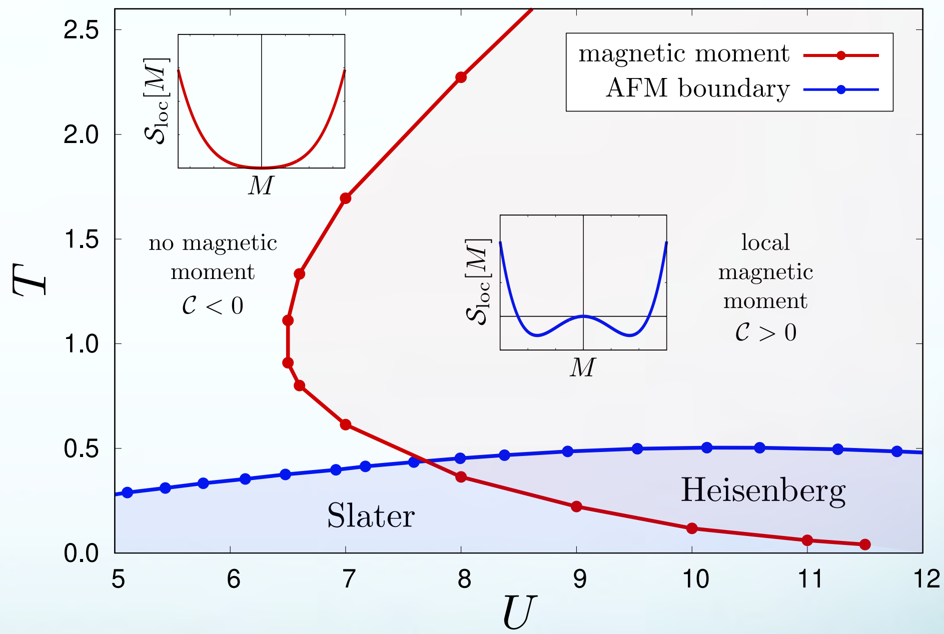
Criterium of Local Moment formation:

$$-\frac{\partial^2 \mathcal{S}_{\rm loc}[\rho^s]}{\partial \rho^s_\tau \, \partial \rho^s_{\tau'}} = [\chi^s]^{-1}_{\tau\tau'} - J^{\rm loc}_{\tau\tau'}$$



Rotated local frame

$$R_{i\tau} = \begin{pmatrix} \cos(\theta_{i\tau}/2) & -e^{-i\varphi_{i\tau}}\sin(\theta_{i\tau}/2) \\ e^{i\varphi_{i\tau}}\sin(\theta_{i\tau}/2) & \cos(\theta_{i\tau}/2) \end{pmatrix}$$



E. Stepanov et al, PRB 105, 155151 (2022)

Effective bosonic action in the adiabatic limit

$$\mathcal{S} \simeq -\frac{1}{4}\int_0^\beta d\tau\, d\tau' \sum_{ij,\varsigma\varsigma'} \rho^{\varsigma}_{i\tau} \mathcal{I}^{\varsigma\varsigma'}_{ij,\tau\tau'} \rho^{\varsigma'}_{j\tau'} + \int_0^\beta d\tau \sum_i A^z_{i\tau} M_{i\tau}$$

kinetic term (spin precession)

$$-\frac{1}{2}\int_0^\beta d\tau\, d\tau' \sum_i \left\{ \rho^c_{i\tau} \chi^{c\,-1}_{\tau\tau'} \rho^c_{i\tau'} + M_{i\tau} \chi^{z\,-1}_{\tau\tau'} M_{i\tau'} \right\}$$

longitudinal (Higgs) fluctuations

DFT functional:

$n(1) = G_{12}\delta_{12} =< c_1^* c_1 >_S$

M. Valiev, G. W. Fernando, Phys. Rev. B 54, 7765 (1996); R. Fukuda et al, Prog. Theor. Phys. 92, 833 (1994)

Hamiltonian with "λ-scaled" interaction part $\hat{H} = \hat{T} + \lambda \hat{U}$

DFT-functional

$$\Gamma[n,\lambda] = F[J,\lambda] - J(1)\,n(1)$$

$$\frac{\delta F[J,\lambda]}{\delta J(1)} = n(1)$$

Inversion method (R. Fukuda):

$$J[n,\lambda] = J_0[n] + \lambda J_1[n] + \lambda^2 J_2[n] + \dots$$
$$F[J,\lambda] = F_0[J] + \lambda F_1[J] + \lambda^2 F_2[J] + \dots,$$
$$\Gamma[n,\lambda] = \Gamma_0[n] + \lambda \Gamma_1[n] + \lambda^2 \Gamma_2[n] + \dots.$$

$$\sum \lambda^i \Gamma_i[n] = \sum \lambda^i F_i\left[\sum \lambda^k J_k[n]\right] - \sum \lambda^i J_i(1)\,n(1)$$

Formal exact expression: $\Gamma_i[n] = F_i[J_0] + \sum_{k=1}^{i} \frac{\delta F_{i-k}[J_0]}{\delta J_0(1)} J_k(1) - J_i(1)\,n(1) + \sum_{m=2}^{i} \frac{1}{m!} \sum_{k_1,\dots,k_m \geq 1}^{k_1+\dots+k_m \leq i} \frac{\delta^m F_{i-(k_1+\dots+k_m)}[J_0]}{\delta J_0(1)\dots\delta J_0(m)} J_{k_1}(1)\cdots J_{k_m}(m)$

DFT (effective single particle) related with zero-order term: $\Gamma_0[n] = F_0[J_0] - J_0(1)\,n(1)$ $\quad n(1) = \frac{\delta F_0[J_0]}{\delta J_0(1)}.$

Kohn-Sham potential: $V_{KS} = V_{ext} + V_H + V_{xc} \Longleftrightarrow J_0$

$$F_{DFT}[n] = T_0[n] + V_{ext}[n] + V_H[n] + V_{xc}[n]$$

$$\begin{aligned} T_0[n] + V_{ext}[n] &= \sum_k \int d\mathbf{r}\, \phi_k^*(\mathbf{r})[-\frac{1}{2}\nabla^2 + V_{ext}(\mathbf{r}) - \mu]\phi_k(\mathbf{r}) \\ V_H[n] &= \frac{1}{2}\int d\mathbf{r}\, n(\mathbf{r}) U(\mathbf{r}-\mathbf{r}') n(\mathbf{r}') \\ V_{xc}[n] &= -\frac{1}{2}\int d\mathbf{r}\, n(\mathbf{r},\mathbf{r}') U(\mathbf{r}-\mathbf{r}') n(\mathbf{r}',\mathbf{r}) + \sum_{i=2}^{\infty} \Gamma_i[n] \end{aligned}$$

$$n(\mathbf{r}) = \sum_k \phi_k^*(\mathbf{r})\phi_k(\mathbf{r})$$

LDA $\quad V_{xc}[n] = \int d\mathbf{r}\, n(\mathbf{r}) \varepsilon_{xc}(n(\mathbf{r}))$

With ε_{xc} from VMC-calculation of D. M. Ceperley and B. J. Alder Phys. Rev. Lett. 45, 566 (1980) – Reference System with fixed n

Why DFT-LDA works?

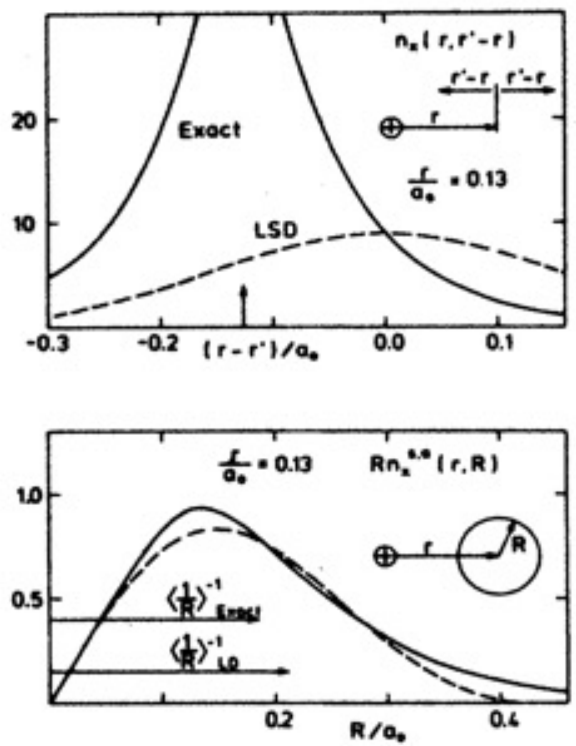
- Errors in the approximation of exchange and correlation cancel

- LDA does fulfill the sum rule for the exchange-correlation hole

$$\int d\mathbf{r}' n_{xc}(\mathbf{r}, \mathbf{r}' - \mathbf{r}) = -1$$

$n_{xc}(\mathbf{r}, \mathbf{r}') = n(\mathbf{r}')[\tilde{g}(\mathbf{r}, \mathbf{r}') - 1]$ = exchange-correlation hole density;

$\tilde{g}(\mathbf{r}, \mathbf{r}')$ = pair correlation function averaged over coupling constant.



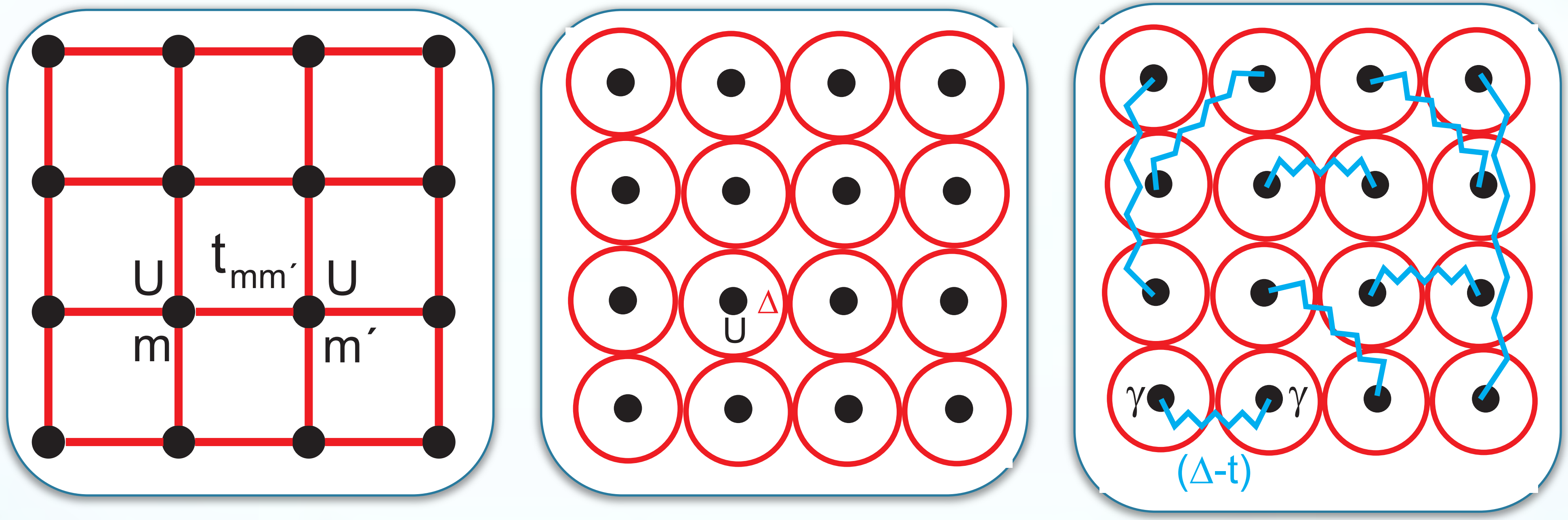
- The exchange-correlation energy depends only on the angle-averaged exchange-correlation hole which is well described in LDA.

$$2E_{xc}[n] = \int \frac{n(\mathbf{r}) n_{xc}(\mathbf{r}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' = \int n(\mathbf{r}) d\mathbf{r} \int \tilde{n}_{xc}(\mathbf{r}, R) dR/R,$$

$$\tilde{n}_{xc}(\mathbf{r}, R) = \int n_{xc}(\mathbf{r}, \mathbf{r} + \mathbf{R}) d\Omega_R / 4\pi.$$

R. O. Jones and O. Gunnarsson, Rev. Mod. Phys. 61, 689 (1989)

How to find “optimal”-functional?



Dual Fermions: Basic

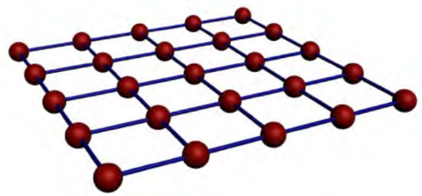
Start from
Correlated Lattice

Find the optimal
Reference System
Bath hybridization

Expand around
DMFT solution

Dual Fermion scheme

General Lattice Action $Z = \int \mathcal{D}[c^*, c] \, \exp\left(-S_L[c^*, c]\right)$

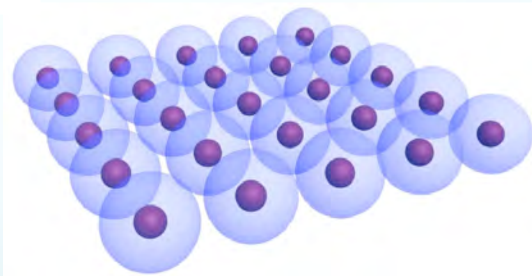


$$S_L[c^*, c] = -\sum_{\mathbf{k}\nu\sigma} c^*_{\mathbf{k}\nu\sigma} \left(i\nu + \mu - t_{\mathbf{k}}\right) c_{\mathbf{k}\nu\sigma} + \sum_i \int_0^\beta d\tau \, U \, n^*_{i\tau\uparrow} n_{i\tau\downarrow}$$

Reference system: Local Action with hybridization Δ_ν

$$S_\Delta[c_i^*, c_i] = -\sum_{\nu,\sigma} c^*_{i\nu\sigma} \left(i\nu + \mu - \Delta_\nu\right) c_{i\nu\sigma} + \sum_\nu U n^*_{i\nu\uparrow} n_{i\nu\downarrow}$$

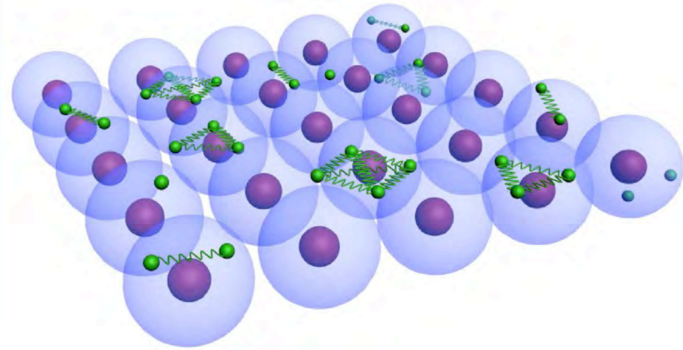
Lattice-Impurity connection:



$$S_L[c^*, c] = \sum_i S_\Delta[c_i^*, c_i] - \sum_{\mathbf{k}\nu\sigma} c^*_{\mathbf{k}\nu\sigma} \left(\Delta_\nu - t_{\mathbf{k}}\right) c_{\mathbf{k}\nu\sigma}$$

A. Rubtsov, et al, PRB **77**, 033101 (2008)

Dual Transformation



Gaussian path-integral

$$e^{c_1^* \,\widetilde{\Delta}_{12}\, c_2} = \det \widetilde{\Delta} \int \mathcal{D}\,[d^*, d]\; e^{-d_1^* \widetilde{\Delta}_{12}^{-1} d_2 - d_1^* c_1 - c_1^* d_1}$$

new Action:

With $\widetilde{\Delta}_{\mathbf{k}\nu} = (\Delta_\nu - t_{\mathbf{k}})$

$$\tilde{S}[d^*, d] = -\sum_{\mathbf{k}\,\nu\sigma} d^*_{\mathbf{k}\nu\sigma}\; \tilde{G}^{-1}_{0\mathbf{k}\nu}\; d_{\mathbf{k}\nu\sigma} + \sum_i V_i[d_i^*, d_i]$$

Diagrammatic:

$\longrightarrow$ $$\tilde{G}^0_{\mathbf{k}\nu} = \left(\left(t_{\mathbf{k}} - \Delta_\nu\right)^{-1} - g_\nu\right)^{-1}$$

$\square$ $$\gamma_{1234} = \chi_{1234} - \chi^0_{1234}$$

$$g_{12} = -\langle c_1 c_2^* \rangle_\Delta$$

$$V[d^*, d] = \frac{1}{4} \sum_{1234} \gamma_{1234}\, d_1^* d_2^* d_4 d_3$$

$$\chi_{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_\Delta$$

g_ω and $\chi_{\nu,\nu',\omega}$ from DMFT impurity solver

Dual Fermion Action: Details

Lattice - dual action $\frac{Z}{Z_d} = \int \mathcal{D}[c^*, c, d^*, d] \exp\big(- S[c^*, c, d^*, d]\big)$ $Z_d = \det \widetilde{\Delta}$

$$S[c^*, c, d^*, d] = \sum_i S^i_\Delta + \sum_{\mathbf{k},\nu,\sigma} d^*_{\mathbf{k}\nu\sigma}\big(\Delta_\nu - t_k\big)^{-1} d_{\mathbf{k}\nu\sigma}$$

$$S^i_\Delta[c^*_i, c_i, d^*_i, d_i] = S_\Delta[c^*_i, c_i] + \sum_{\nu,\sigma}\big(d^*_{i\nu\sigma}\, c_{i\nu\sigma} + c^*_{i\nu\sigma}\, d_{i\nu\sigma}\big)$$

For each site (i) integrate-out original c-Fermions:

$$\frac{1}{Z_\Delta}\int \mathcal{D}[c^*, c] \exp\big(-S^i_\Delta[, c^*_i, c_i, d^*_i d_i]\big) = \exp\Big(-\sum_{\nu\,\sigma} d^*_{i\nu\sigma}\, g_\nu d_{i\nu\sigma} - V_i[d^*_i d_i]\Big)$$

Dual potential: $V[d^*, d] = \frac{1}{4}\sum_{1234} \gamma_{1234}\, d^*_1 d^*_2 d_4 d_3$ +... $\gamma_{1234} = \chi_{1234} - \chi^0_{1234}$

$$\chi_{1234} = \langle c_1 c_2 c^*_3 c^*_4\rangle_\Delta = \frac{1}{Z_\Delta}\int \mathcal{D}[c^*, c]\, c_1 c_2 c^*_3 c^*_4\, e^{-S_\Delta[c^*,c]}$$

$$\chi^0_{1234} = g_{14}g_{23} - g_{13}g_{24}$$

$$g_{12} = -\langle c_1 c^*_2\rangle_\Delta = \frac{1}{Z_\Delta}\int \mathcal{D}[c^*, c]\, c_1 c^*_2\, e^{-S_\Delta[c^*,c]}$$

Dual and Lattice Green's Functions

Two equivalent forms for partition function:

$$e^{F[J^*J,L^*L]} = \mathcal{Z}_d \int \mathcal{D}[c^*c, d^*d]\, e^{-S[c^*c,d^*,d]+J_1^*c_1+c_2^*J_2+L_1^*d_1+d_2^*L_2}$$

$$e^{F[L^*,L]} = \tilde{\mathcal{Z}}_d \int \mathcal{D}[d^*, d]\, e^{-S_{\mathrm{d}}[d^*,fd+L_1^*d_1+d_2^*L_2}$$

$$\tilde{\mathcal{Z}}_d = \mathcal{Z}/\tilde{\mathcal{Z}}$$

Hubbard-Stratanovich transformation:

$$F[J^*J, L^*L] = L_1^*(\varDelta - t)_{12}L_2 + \ln \int \mathcal{D}[c^*, c] \exp\Big(-S[c^*, c] + J_1^*c_1 + c_2^*J_2 + L_1^*(\varDelta - t)_{12}c_2 + c_1^*(\varDelta - t)_{12}L_2\Big)$$

Relation between Green functions:

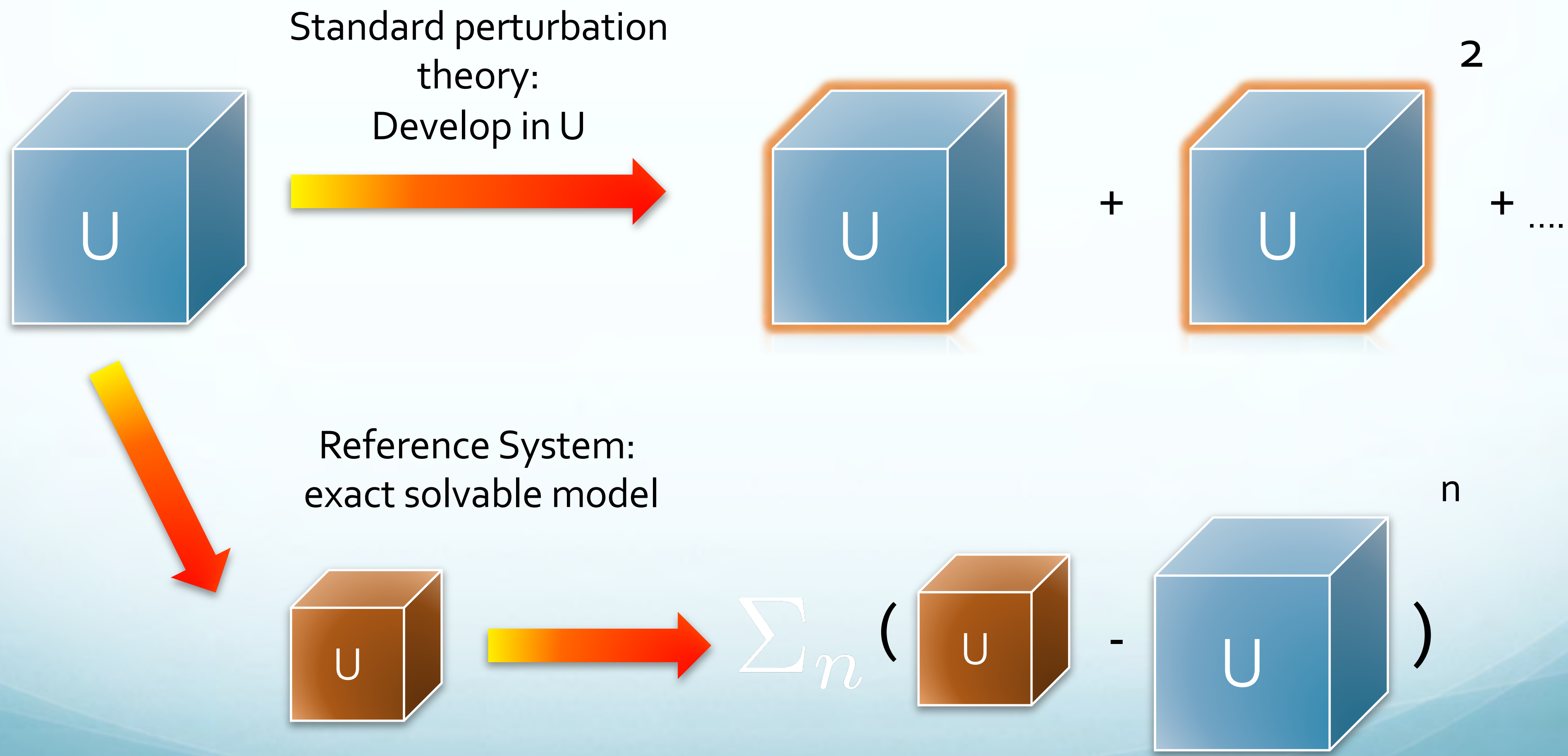
$$\tilde{G}_{12} = -\frac{\delta^2 F}{\delta L_2 \delta L_1^*}\bigg|_{L^*=L=0}$$

$$\tilde{G}_{12} = -(\varDelta - t)_{12} + (\varDelta - t)_{11'}G_{1'2'}(\varDelta - t)_{2'2}$$

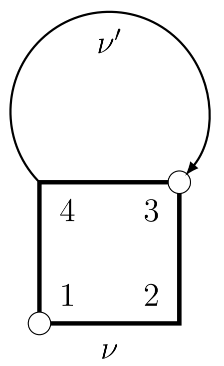
T-matrix like relations via dual self-energy

$$G_{\mathbf{k}\nu} = \left(\left(g_\nu + \widetilde{\Sigma}_{\mathbf{k}\nu}\right)^{-1} - \widetilde{\Delta}_{\mathbf{k}\nu}\right)^{-1}$$

Super-perturbation



1-st order diagram for dual self-energy



$$\widetilde{\Sigma}^{(1)i}_{12}(\nu) = \sum_{\nu',3,4} \gamma^{d}_{1234}(\nu,\nu',0)\, \widetilde{G}^{ii}_{43}(\nu')$$

Density (d) and Magnetic (m) Vertices:

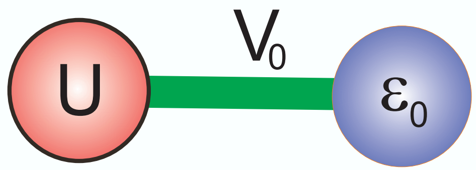
$$\gamma^{d/m}_{1234}(\nu,\nu',\omega) = \gamma^{\uparrow\uparrow}_{1234}(\nu,\nu',\omega) \pm \gamma^{\uparrow\downarrow}_{1234}(\nu,\nu',\omega)$$

Connected 2-particle GF:

$$\gamma^{\sigma\sigma'}_{1234}(\tau_1,\tau_2,\tau_3,\tau_4) = -\left\langle c_{1\sigma} c^{*}_{2\sigma} c_{3\sigma'} c^{*}_{4\sigma'} \right\rangle_{\Delta} + g^{\sigma}_{12} g^{\sigma'}_{34} - g^{\sigma}_{14} g^{\sigma}_{32} \delta_{\sigma\sigma'}$$

Two site test

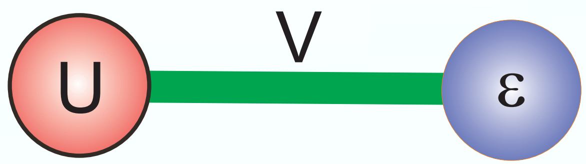
$U\!=\!2,\ \varepsilon_0\!=\!0$ $\quad V_0\!=\!0.5$



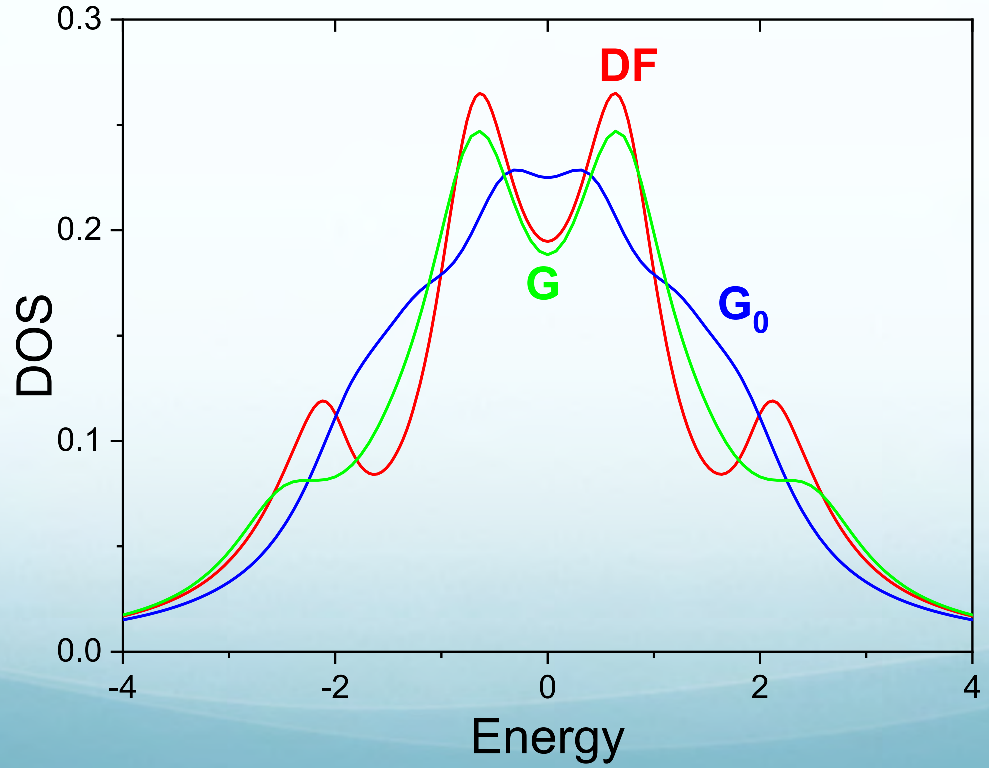
Ref.

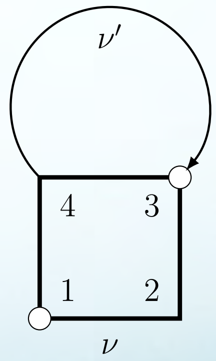
Fixed

$V\!=\!1.5V_0$

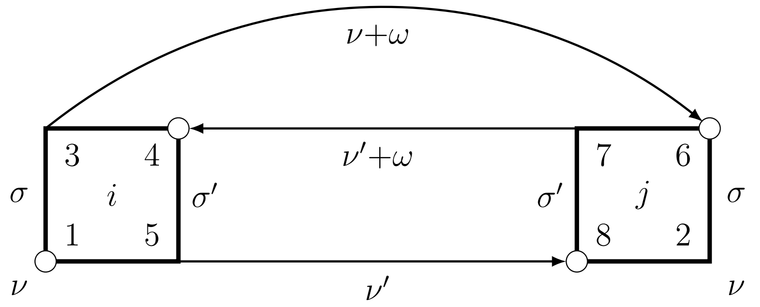


Sys.





2-nd order diagram for dual self-energy



$c_d = -1/4$ and $c_m = -3/4$

$$\widetilde{\Sigma}^{(2)ij}_{12}(\nu) = \sum_{\nu'\omega} \sum_{3\text{-}8} \sum_{\alpha=d,m} c_\alpha \gamma^{\alpha,i}_{1345}(\nu,\nu',\omega)\, \widetilde{G}^{ij}_{36}(\nu+\omega) \widetilde{G}^{ji}_{74}(\nu'+\omega)\, \widetilde{G}^{ij}_{58}(\nu')\, \gamma^{\alpha,j}_{8762}(\nu',\nu,\omega)$$

Lattice Self-Energy:

$$\Sigma_{\mathbf{k}\nu} = \Sigma^0_\nu + \Sigma'_{\mathbf{k}\nu}$$

Non-Local DF-correction:

$$\Sigma'_{\mathbf{k}\nu} = g_\nu^{-1} - \left(g_\nu + \widetilde{\Sigma}_{\mathbf{k}\nu}\right)^{-1}$$

Lattice Green Function:

$$G_{\mathbf{k}\nu} = \left(\left(g_\nu + \widetilde{\Sigma}_{\mathbf{k}\nu}\right)^{-1} - \widetilde{\Delta}_{\mathbf{k}\nu}\right)^{-1}$$

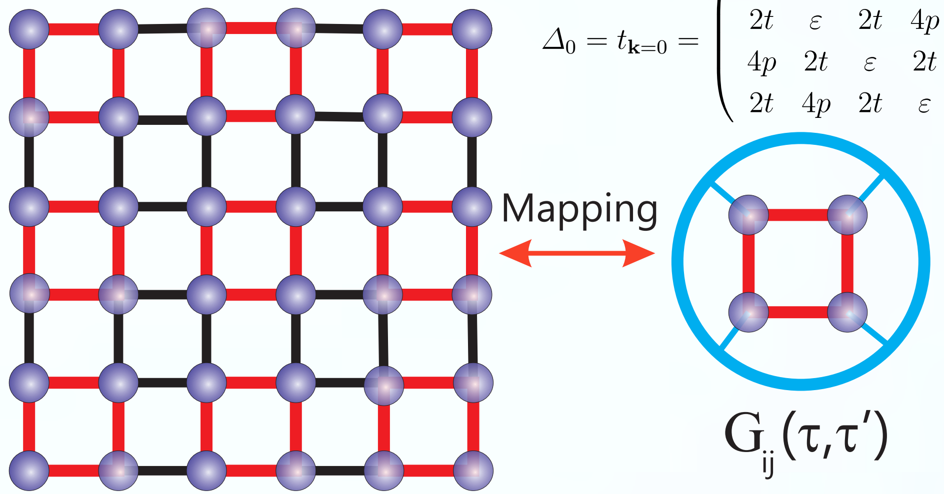
Plaquette DF-perturbation

$$t_{\mathbf{k}} = \begin{pmatrix} \varepsilon & tK^{0+} & pL^{-+} & tK^{-0} \\ tK^{0-} & \varepsilon & tK^{-0} & pL^{--} \\ pL^{+-} & tK^{+0} & \varepsilon & tK^{0-} \\ tK^{+0} & pL^{++} & tK^{0+} & \varepsilon \end{pmatrix}$$

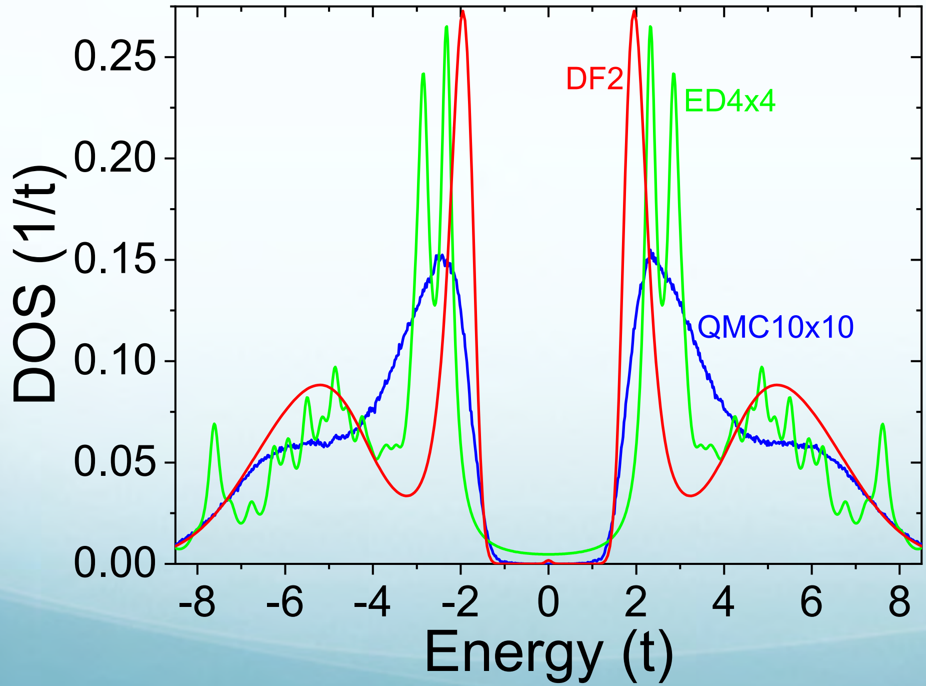
$$\begin{aligned} K_{\mathbf{k}}^{mn} &= 1 + e^{i(mk_x+nk_y)} \\ L_{\mathbf{k}}^{mn} &= 1 + e^{i(mk_x+nk_y)} + e^{imk_x} + e^{ink_y} \end{aligned}$$

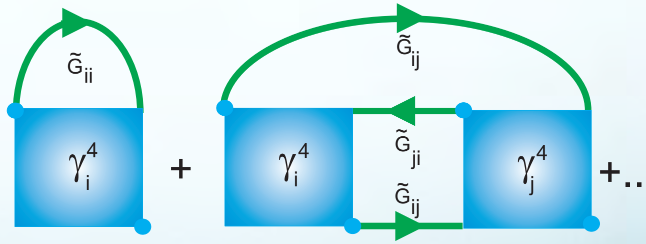
$$[m(n)] = -(1), 0, +(1)$$

$$\Delta_0 = t_{\mathbf{k}=0} = \begin{pmatrix} \varepsilon & 2t & 4p & 2t \\ 2t & \varepsilon & 2t & 4p \\ 4p & 2t & \varepsilon & 2t \\ 2t & 4p & 2t & \varepsilon \end{pmatrix}$$



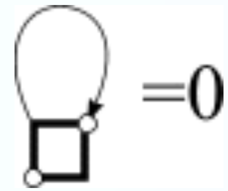
U=8
t=1
T=t/5





Condition for Δ and relation with DMFT

G^d=G^{DMFT}-g

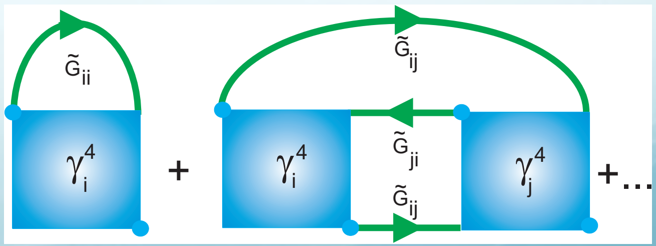


To determine Δ, we require
that Hartree correction in dual variables vanishes.
If no higher diagrams are taken into account, one obtains DMFT:

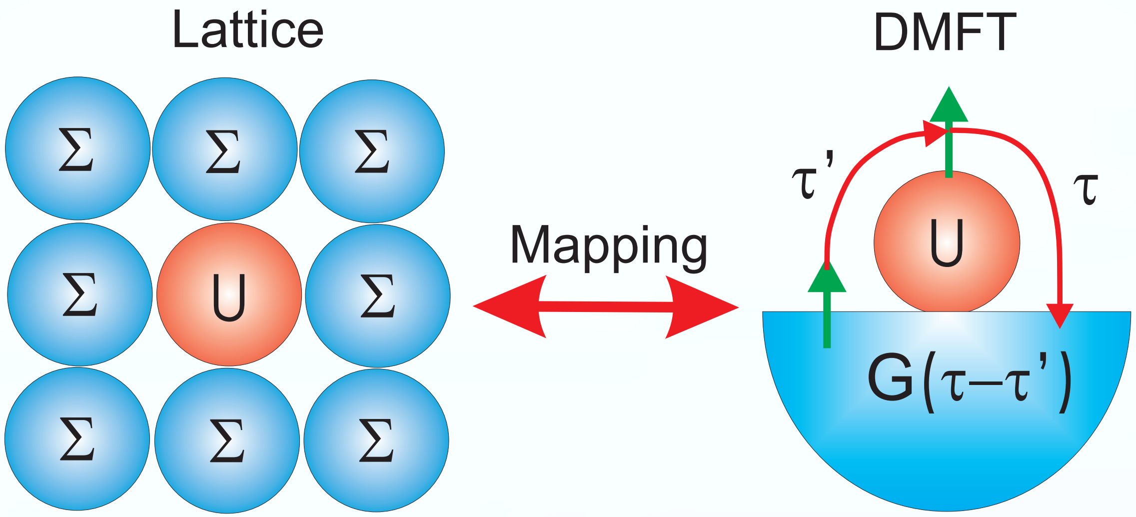
$$G_d = g\,\widetilde{G}\,g = G_{DMFT} - g \qquad G_{DMFT} = \left(g_\nu + \Delta_\nu - t_{\mathbf{k}}\right)^{-1}$$

$$\frac{1}{N}\sum_{\mathbf{k}} \tilde{G}^0_\omega(\mathbf{k}) = 0 \quad \Longleftrightarrow \quad \frac{1}{N}\sum_{\mathbf{k}} G^{\mathrm{DMFT}}_\omega(\mathbf{k}) = g_\omega$$

Higher-order diagrams give corrections
to the DMFT self-energy, and already
the leading-order correction is nonlocal.



Dinamical Mean Field Theory

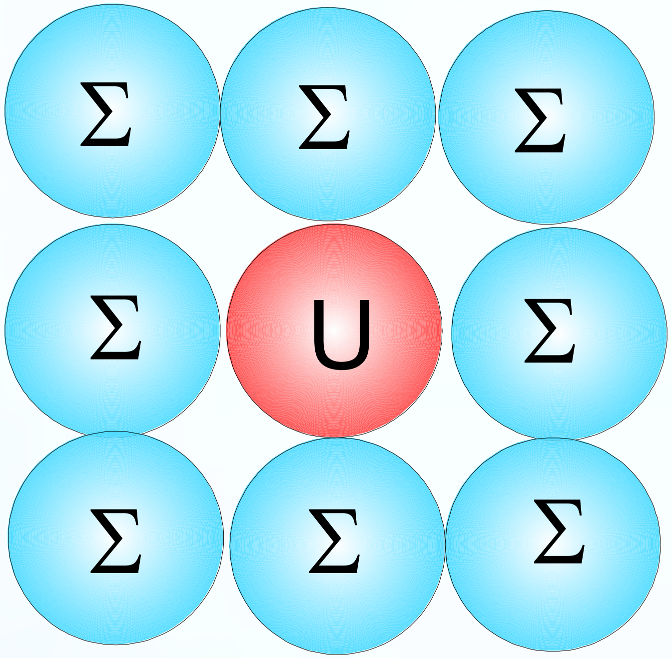


Self-consistent condition:

$$\sum_{\mathbf{k}} \left(g_\nu^{-1} + \Delta_\nu - t_{\mathbf{k}}\right)^{-1} = g_\nu$$

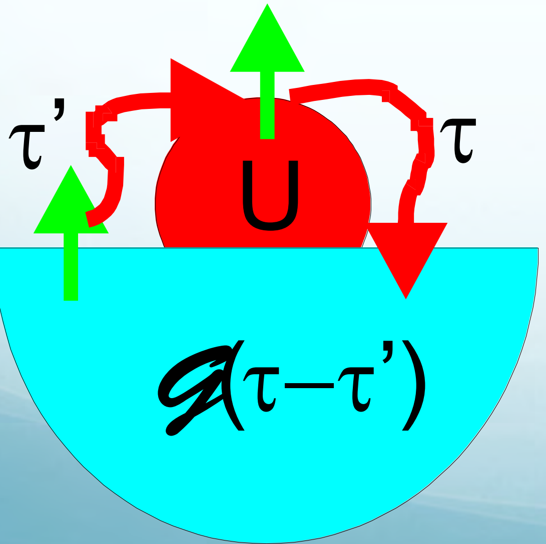
DMFT minimize “distance”: $|t_{\mathbf{k}} - \Delta_\nu|$

Quantum Impurity Solver



$$Z = \int \mathcal{D}[c^*, c] e^{-S_{simp}},$$

$$S_{simp} = -\sum_{I,J=0}^{N} \int_0^\beta d\tau \int_0^\beta d\tau' \, c^*_{I\sigma}(\tau) \left[\mathcal{G}_\sigma^{-1}(\tau-\tau')\right]_{IJ} c_{J\sigma}(\tau') + \sum_{I=1}^{N} \int_0^\beta d\tau U n_{I,\uparrow}(\tau) n_{I,\downarrow}(\tau),$$



What is a best scheme?
Quantum Monte Carlo !

CT-QMC: random walks in fermionic-det space

$Z = \ldots Z_{k-1} + Z_k + Z_{k+1} + \ldots.$

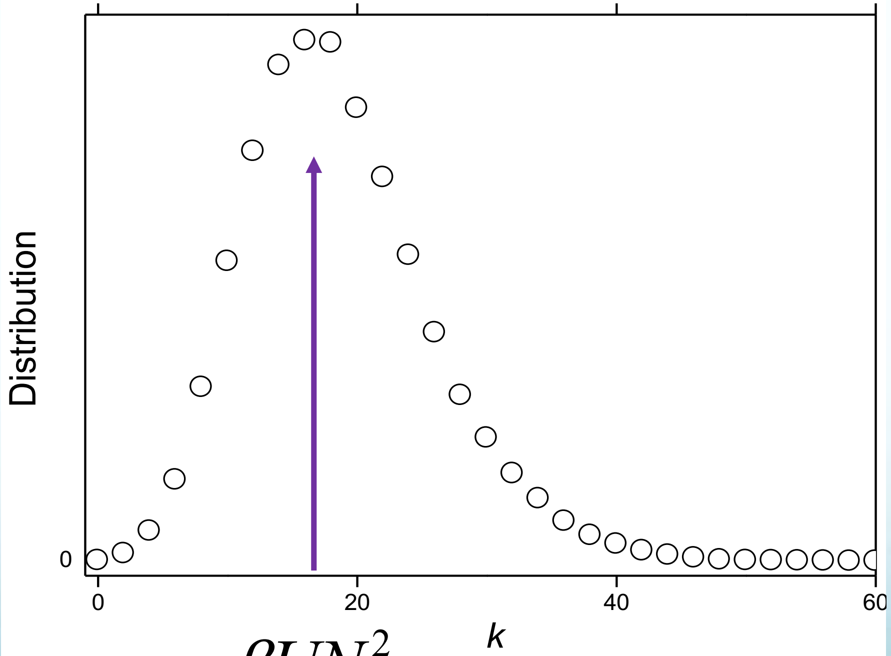
k-1 *k*+1

Acceptance ratio

decrease

Step *k*-1

$$\frac{k}{|w|}\frac{D^{k-1}}{D^k}$$



increase

Step *k*+1

$$\frac{|w|}{k+1}\frac{D^{k+1}}{D^k}$$

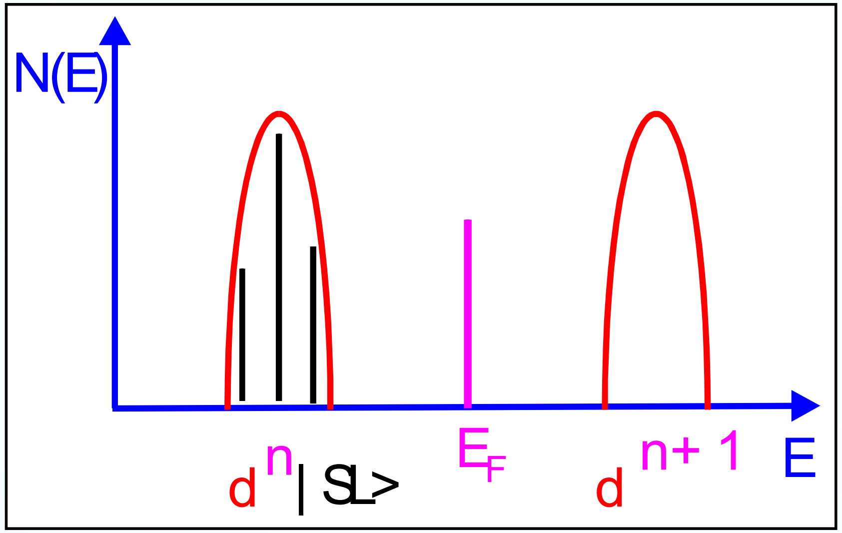
Maximum at $\beta U N^2$

A. Rubtsov and A.L., JETP Lett. 80, 67 (2004)

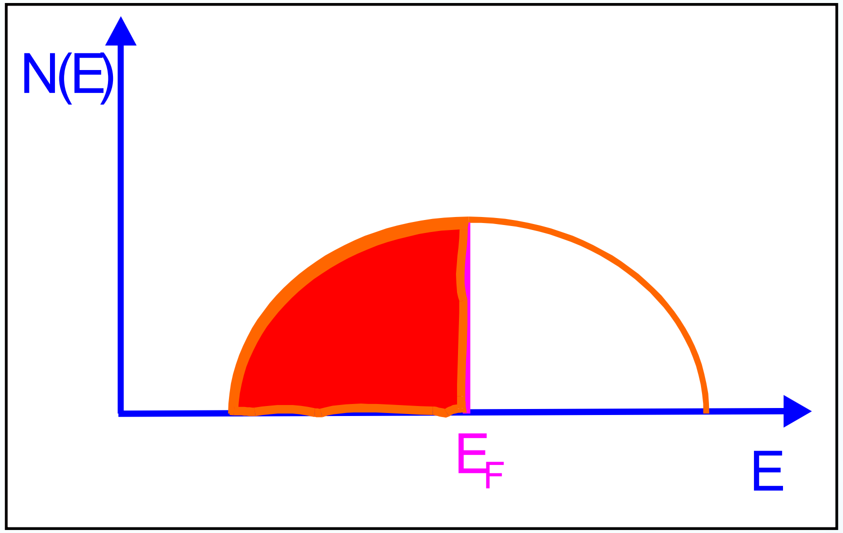
E. Gull, et al Rev. Mod. Phys. **83**, 349 (2011)

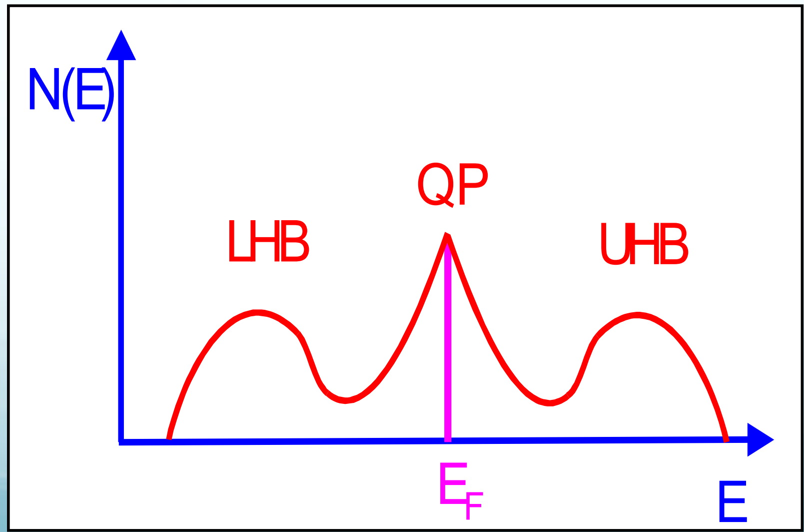
From Atom to Solid

Atomic physics (U)



Bands effects (LDA)

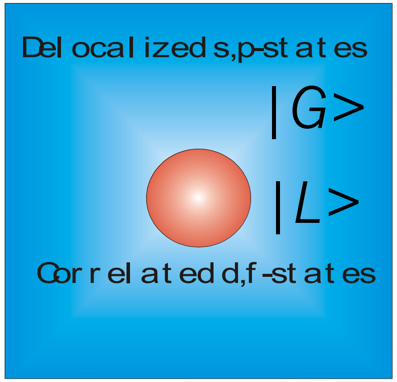




LDA+DMFT

General Projection formalism for LDA+DMFT

$$
\begin{aligned}
|L\rangle &= |il m\sigma\rangle \\
|G\rangle &= \left|n\,\vec{k}\,\sigma\right\rangle
\end{aligned}
\qquad
\begin{aligned}
\left\langle L_i | L_j \right\rangle &= \delta_{ij} \\
P_c &= \langle L|G\rangle
\end{aligned}
$$



P. Blöchl, PRB **50,** 17953 (1994)

$$G^{c}_{mm'}(i\omega) = \sum_{\vec{k}\,nn'} \langle L_m|G_n\rangle \left[(i\omega+\mu)\,\hat{1} - \widehat{H}_{KS}(\vec{k}) - \Delta\Sigma(i\omega)\right]^{-1}_{nn'} \langle G_{n'}|L_{m'}\rangle$$

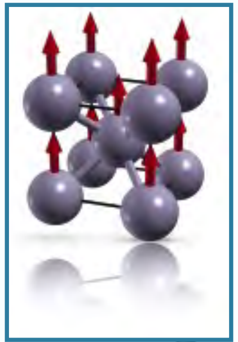
$$\Delta\Sigma_{nn'}(i\omega) = \sum_{mm'} \langle G_n|L_m\rangle\, \Delta\Sigma_{mm'}(i\omega)\, \langle L_{m'}|G_{n'}\rangle$$

$$
\begin{aligned}
\Sigma_{mm'}(i\omega) &= \left(G_0^{-1} - G^{-1}\right)_{mm'} \\
\Delta\Sigma_{mm'}(i\omega) &= \Sigma_{mm'}(i\omega) - \Sigma_{dc}
\end{aligned}
$$

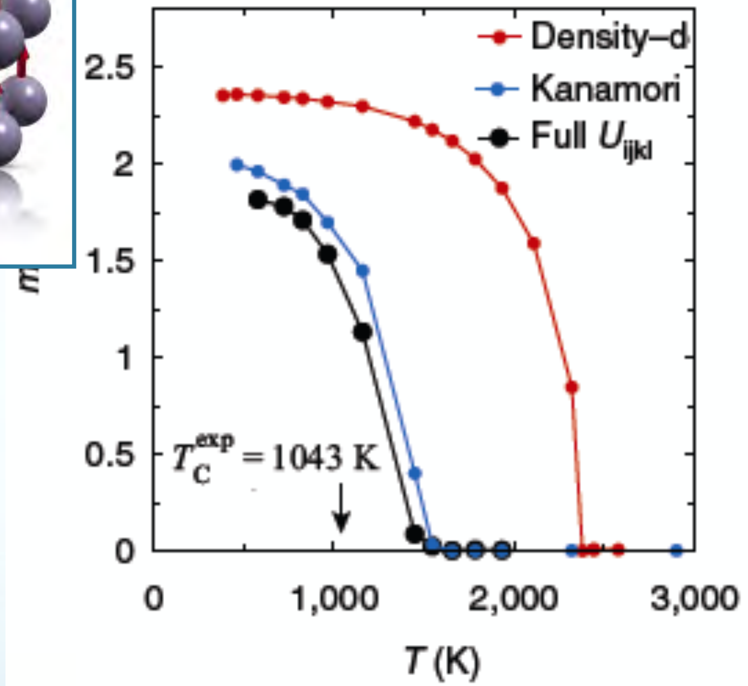
G. Trimarchi, *et al*. JPCM **20**,135227 (2008)
B. Amadon, *et al*. PRB **77**, 205112 (2008)

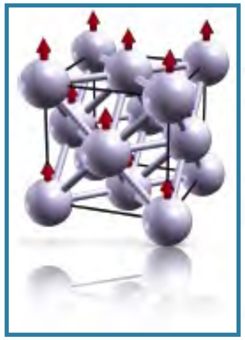
DFT+ DMFT: Curie Temperature

✧ calculations with Full U_{ijkl} from cRPA

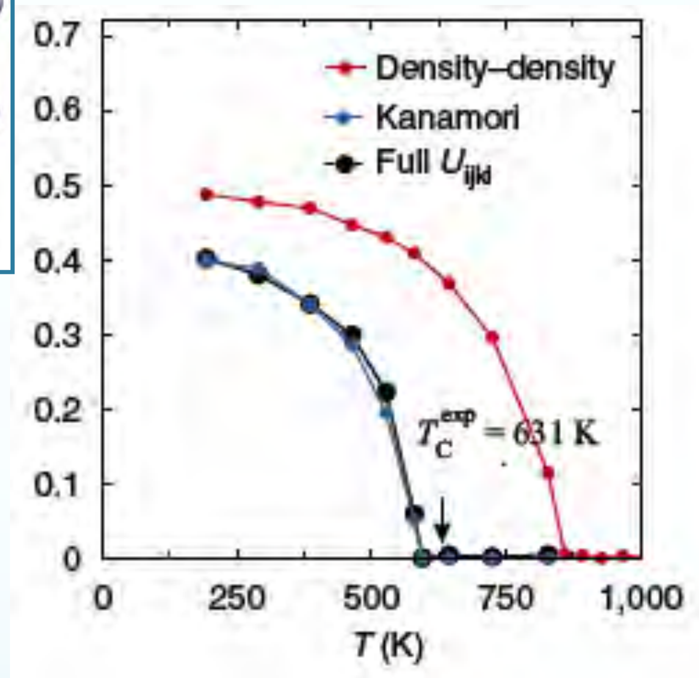


▪ Fe (bcc)





▪ Ni (fcc)

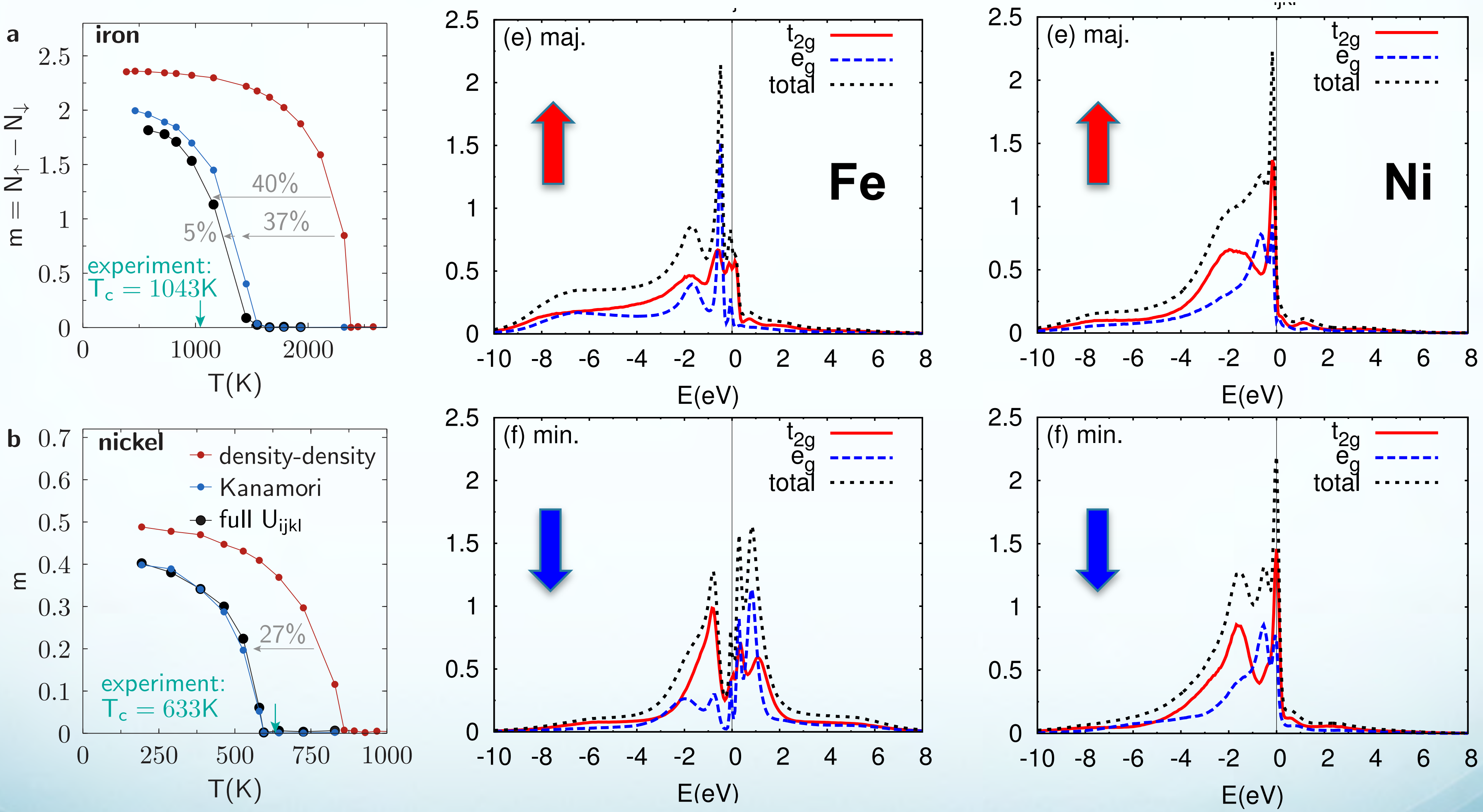


❖ **A.Hausoel, M. Karolak, E. Sasioglu, A. L., K. Held, A. Katanin, A.Toschi and G. Sangiovanni**

nature COMMUNICATIONS

DOI: 10.1038/ncomms16062

DFT+ DMFT: Curie Temperature



$$H_{\text{int}} = \frac{1}{2}\sum_{m,\sigma} U_{mm'm''m'''} c^{+}_{m\sigma} c^{+}_{m'\sigma'} c_{m'''\sigma'} c_{m''\sigma}$$

A. Hausoel, et al Nat. Comm. 8, 16062 (2017)

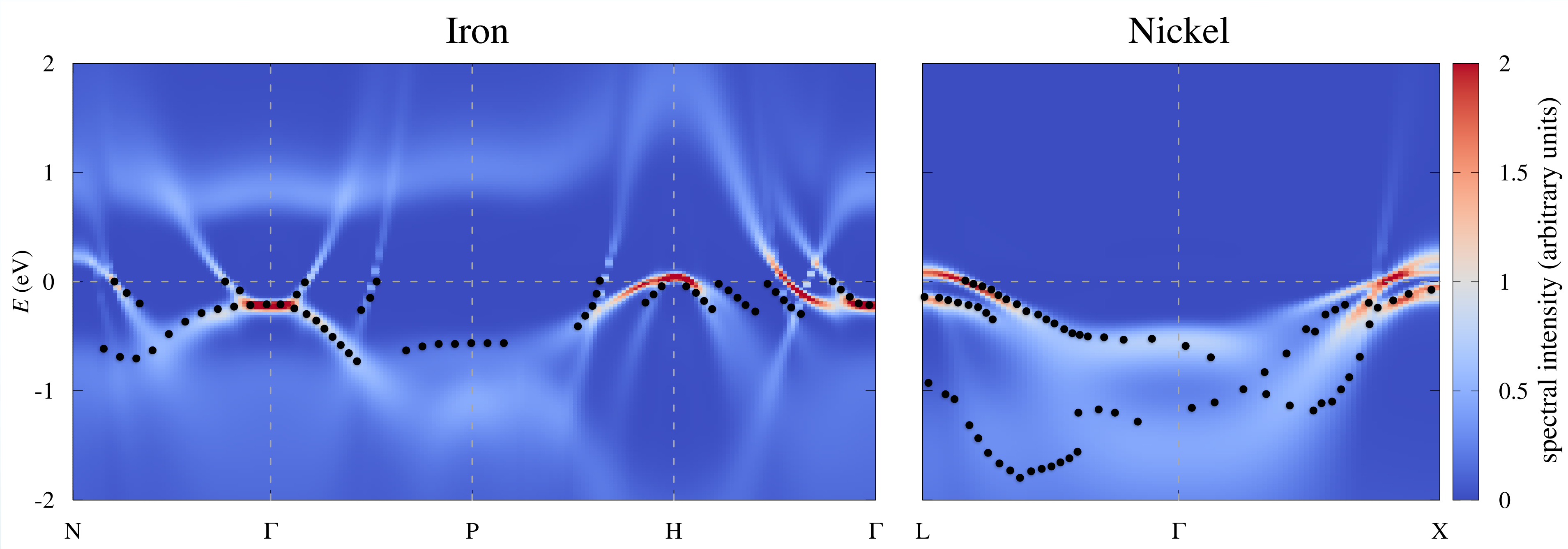
Ni:PES (DFT)

$W_{band} = 3(4)$ eV

$\Delta E_{ex} = 0.3(0.6)$ eV

$E_{sat} = -6(?)$ eV

Spectral Function for Fe and Ni paramagnetic DFT+DMFT

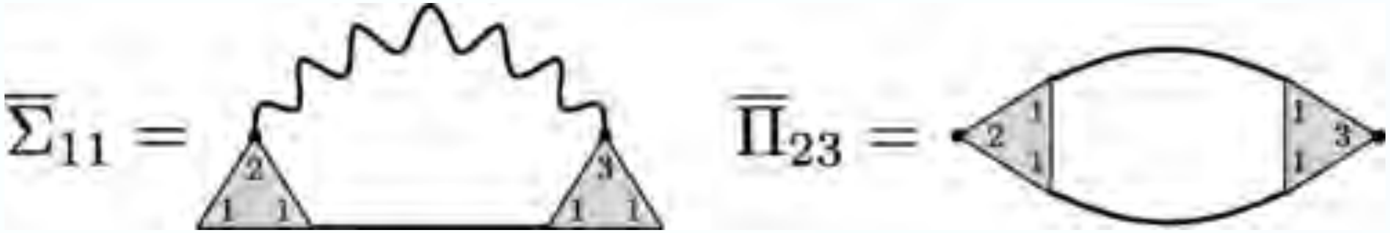


A. Hausoel, M. Karolak, E. Sasioglu, A. L., K. Held, A. Katanin, A.Toschi and G. Sangiovanni
Nat. Comm. 8, 16062 (2017)

Magnetic fluctuations and Hund coupling

E. Stepanov, Y. Nomura, A.L., and S. Biermann, PRL **127**, 207205 (2021)

D-TRILEX



3D-3-orbital t_{2g}-model ($C_\alpha = \cos k_\alpha$)

$l = \{\alpha\beta\}$

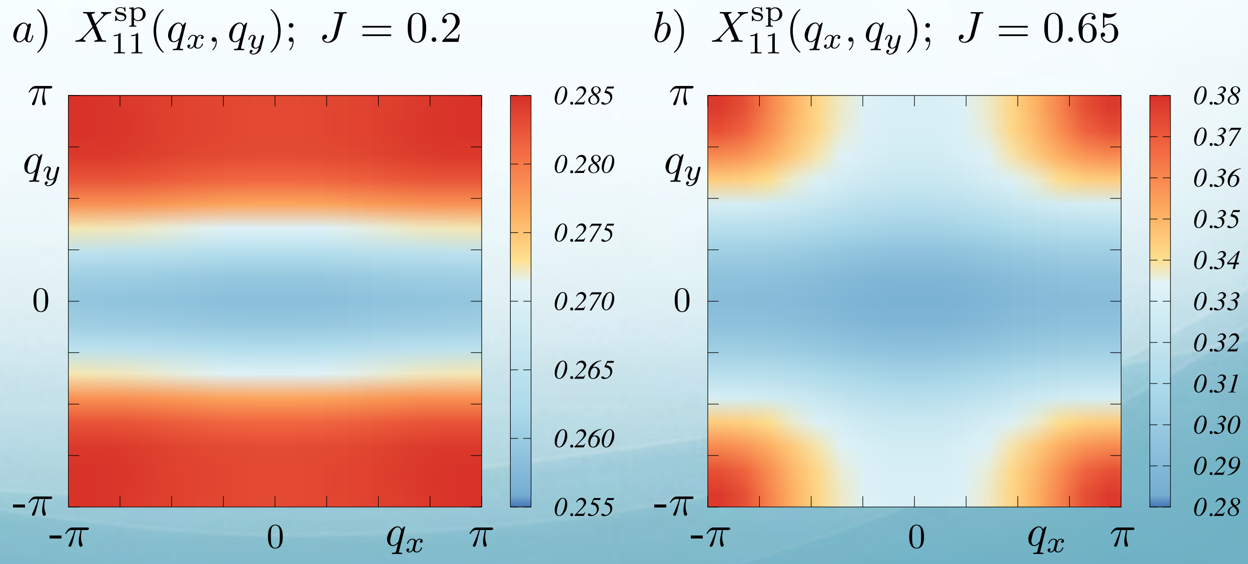
$1 = yz,$

$2 = zx$, and $3 = xy$

$$\mathcal{H} = -\sum_{ij,l,\sigma} t^{ll}_{ij} c^{\dagger}_{il\sigma} c_{jl\sigma} + \frac{1}{2}\sum_{i,ll'} \left(U^{\mathrm{ch}}_{ll'} n_{il} n_{il'} + U^{\mathrm{sp}}_{ll'} m_{il} m_{il'} \right)$$

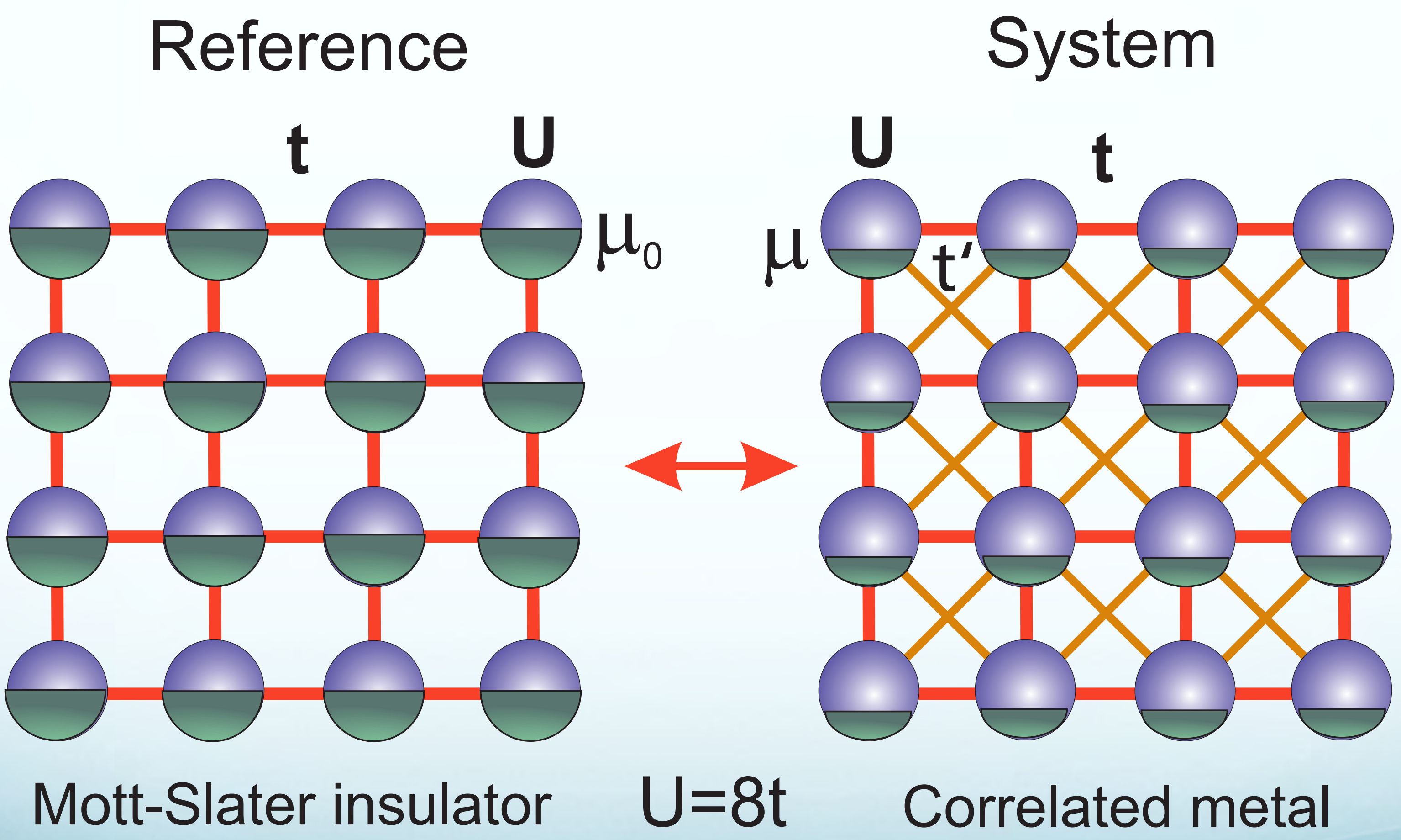
$$t_{ll}(\mathbf{k}) = \epsilon + 2t_\pi(C_\alpha + C_\beta) + 2t_\delta C_\gamma + 4t_\sigma C_\alpha C_\beta$$

Spin susceptibility (yz-component)



Super-perturbation: DF-QMC

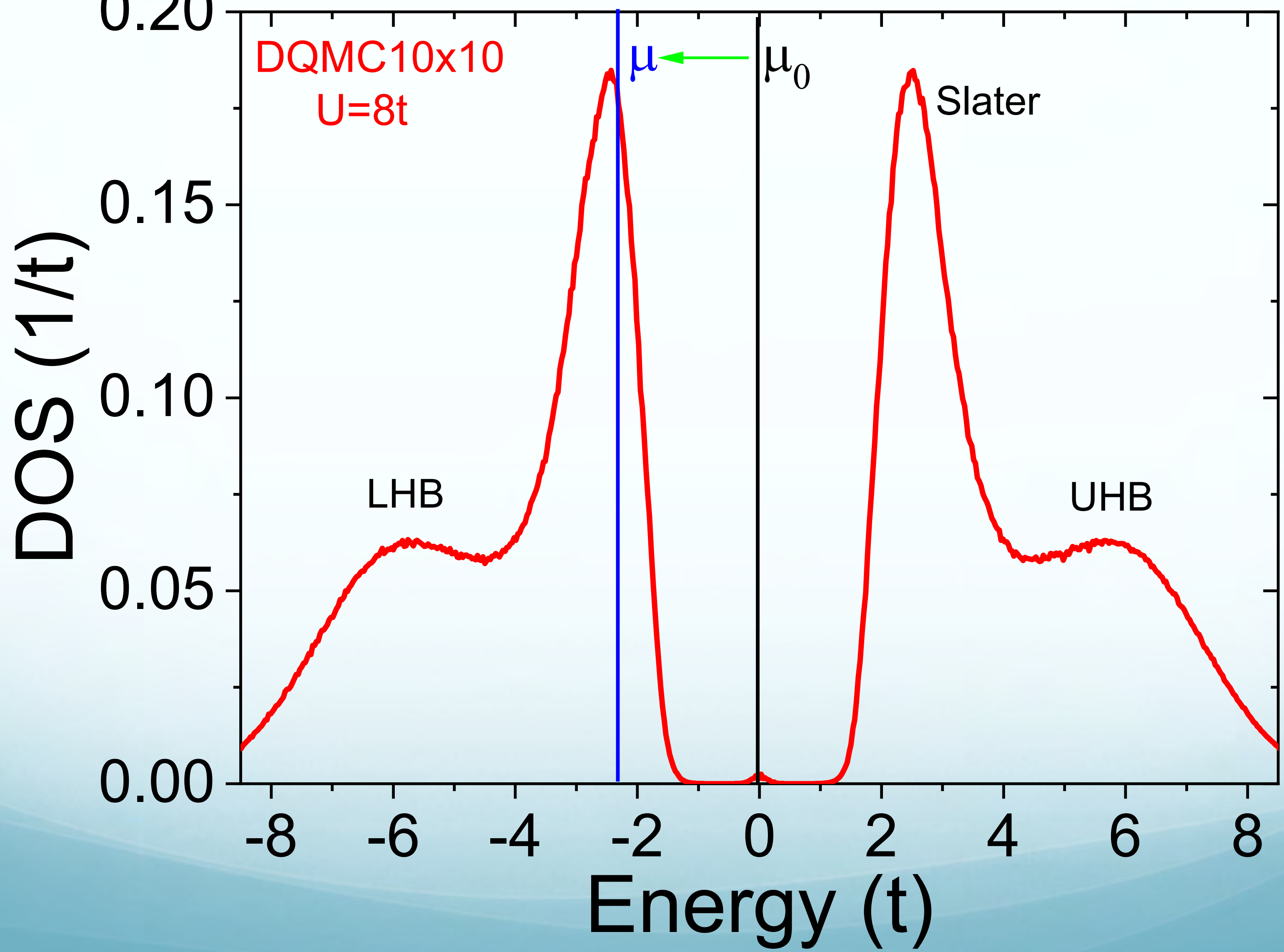
- Controllable perturbative solution of doped Hubbard model for HTSC
- Developed DF expansion around DQMC for N=1, t'=0



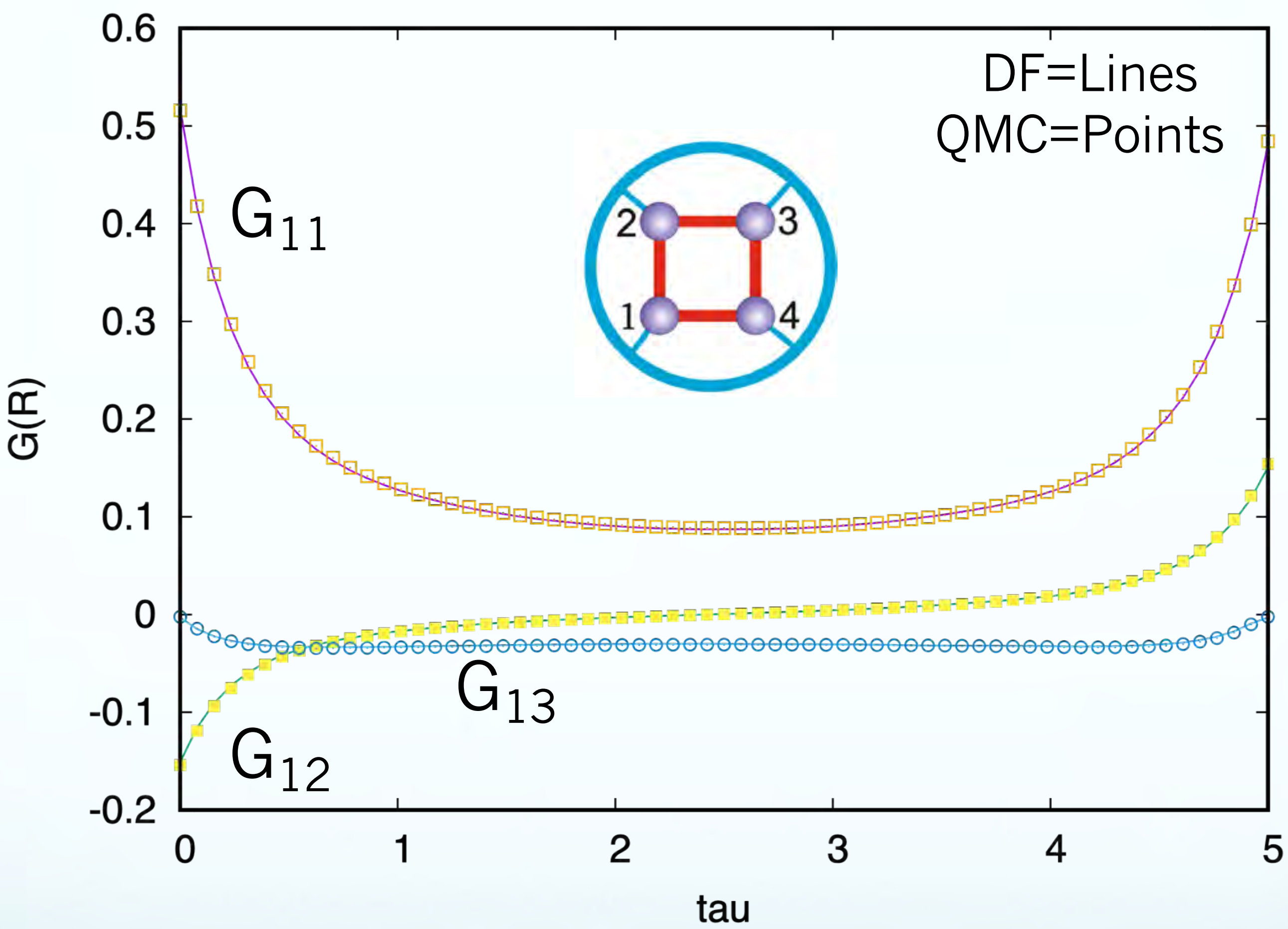
DQMC – no sign problem

DF-superperturbation

DOS for Reference System



Super-DF-QMC 2x2 test DQMC-Hirsch



DF-pert:

μ=-0.3

t'=0

U=5.56

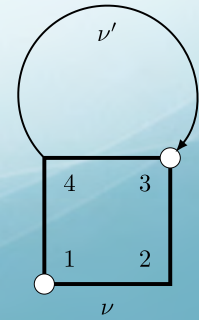
β=5

L=64

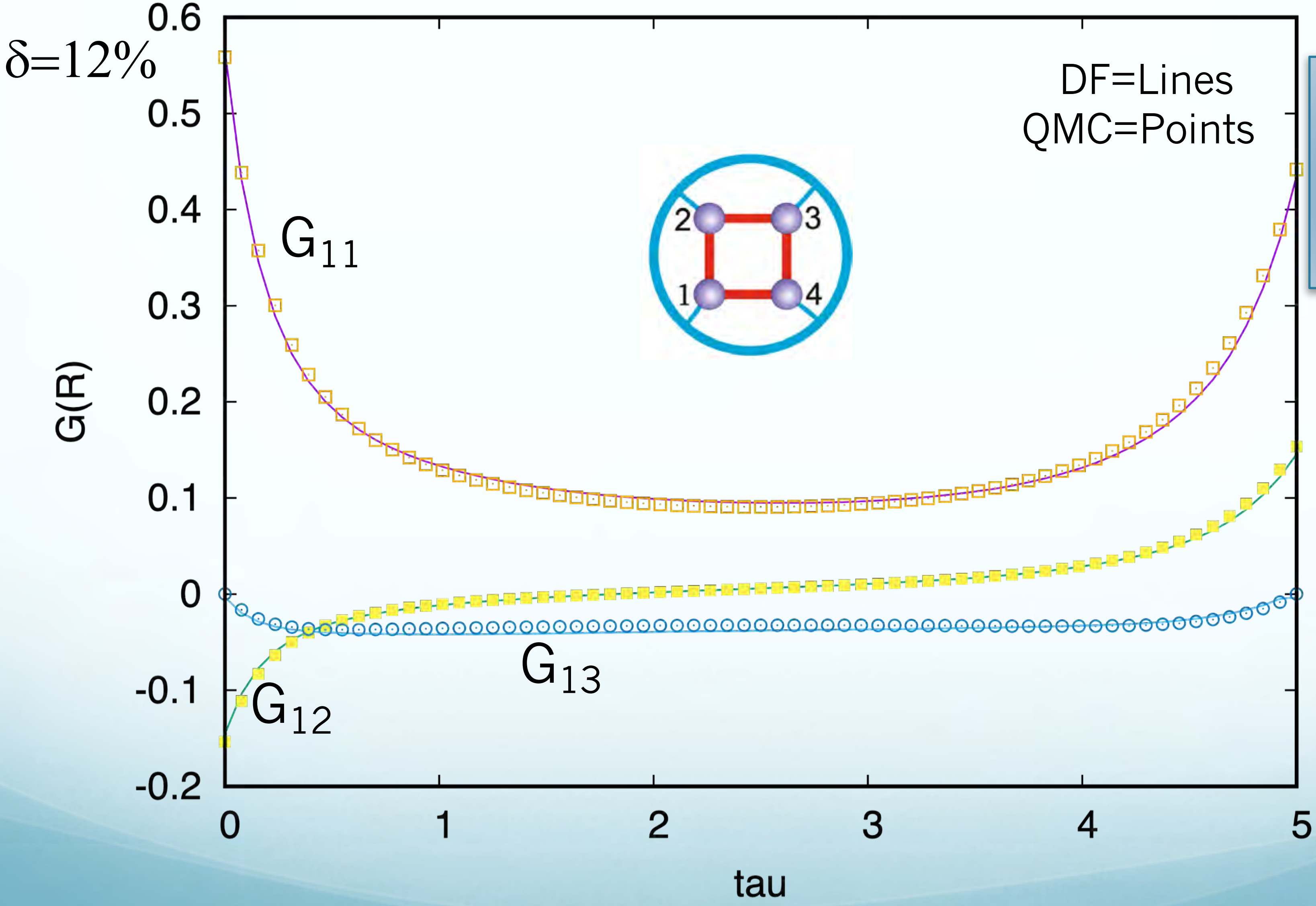
(ℓ =1,L)

$$\Sigma_{12} \sim \frac{1}{Z}\mathop{\mathrm{Tr}}_{\{s\}} \langle c_1 c_2^* c_3 c_4^* \rangle_{\{s\}} \tilde{G}_{43} \qquad 1 \equiv (i, \ell, \sigma)$$

$$\langle c_1 c_2^* c_3 c_4^* \rangle_{\{s\}} = \langle c_1 c_2^* \rangle_{\{s\}} \langle c_3 c_4^* \rangle_{\{s\}} - \langle c_1 c_4^* \rangle_{\{s\}} \langle c_3 c_2^* \rangle_{\{s\}}$$



Super-DF-QMC 2x2 compare with exact QMC



DF-pert:

$\mu=-1.3$

t'/t=-0.15

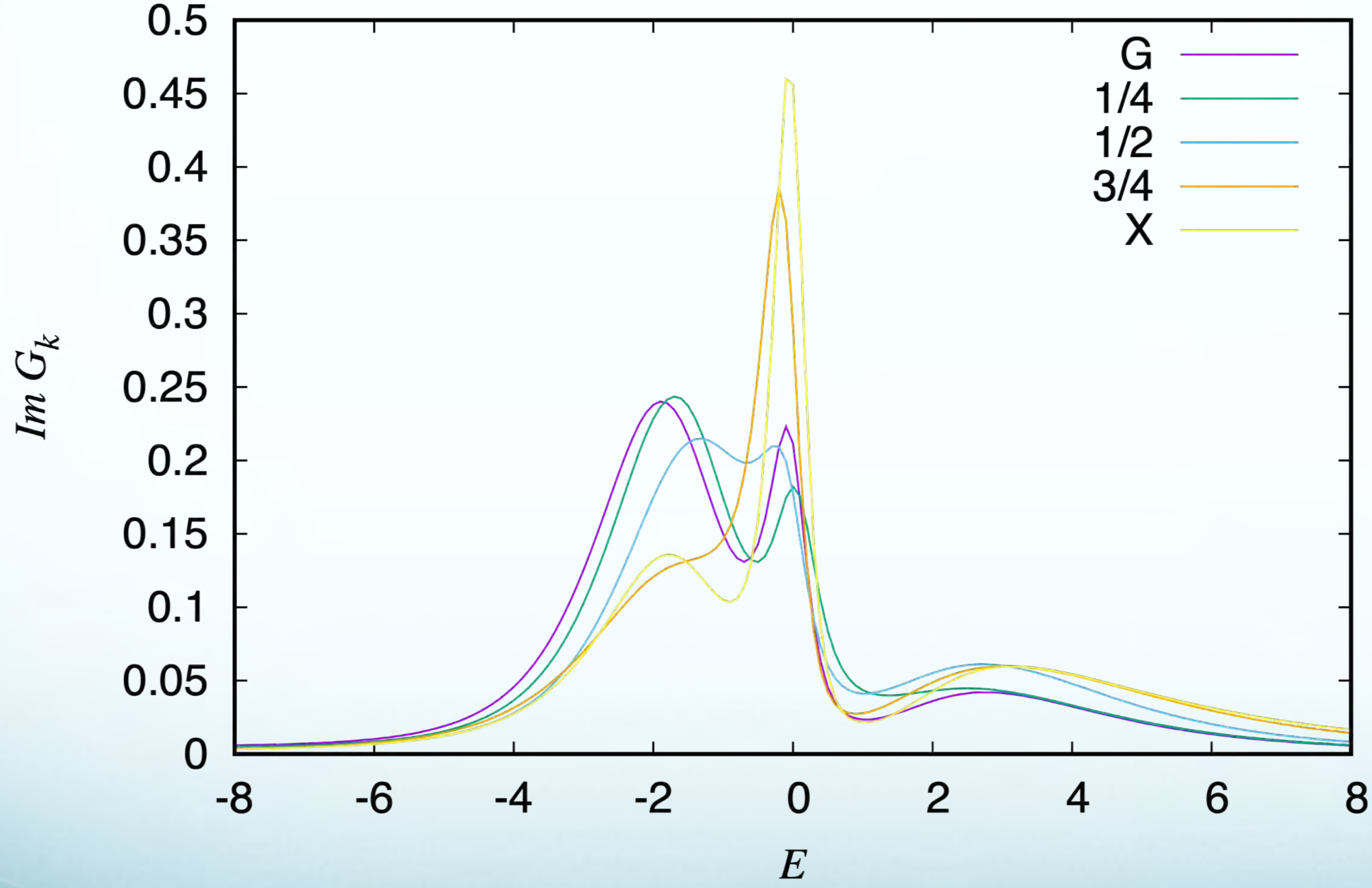
U=5.56

β=5

L=64

(ℓ=1,L)

DFQ for 8x8: Spectral Function



T=t/5
t'/t=-0.3
U=6

μ=-1.3

β=5

Conclusions

- Local correlations well described with the CT-QMC impurity solver: basis for DFT+DMFT
- Multiorbital D-TRILEX is most efficient scheme for non-local correlation in realistic systems
- DF-theory can be combined with Lattice DQMC to describe strongly correlated materials

Collaborations with:

Viktor Harkov, Matteo Vandelli (Hamburg)
Erik G.C.P. van Loon (Lund)
Sergei Iskakov (Michigan)
Evgeny Stepanov (Paris)
Mikhail Katsnelson (Nijmegen)