TECHNISCHE UNIVERSITÄT WIEN
Vienna University of Technology

European Research Council
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(electronic) correlations at the nano-scale

Angelo Valli

"ab-initio DΓA" ERC kick-off

1-4 September 2013

intro

- electronic correlations at the nanoscale
- model system

methods

- extending DMFT & DΓA to nano
- solving the parquet

  remarks: parquet formalism

applications

- local & non-local correlations in quasi 1D rings

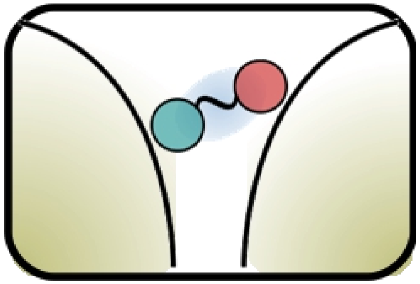
  benchmark nano-DMFT

  understand role of non-local correlations

outlook

- technical improvements
- understanding & employing vertex functions

- spatial confinement & low-dimensionality: **enhanced correlations**
- **competing** (bare) **energy scales**



- spatial confinement & low-dimensionality: **enhanced correlations**
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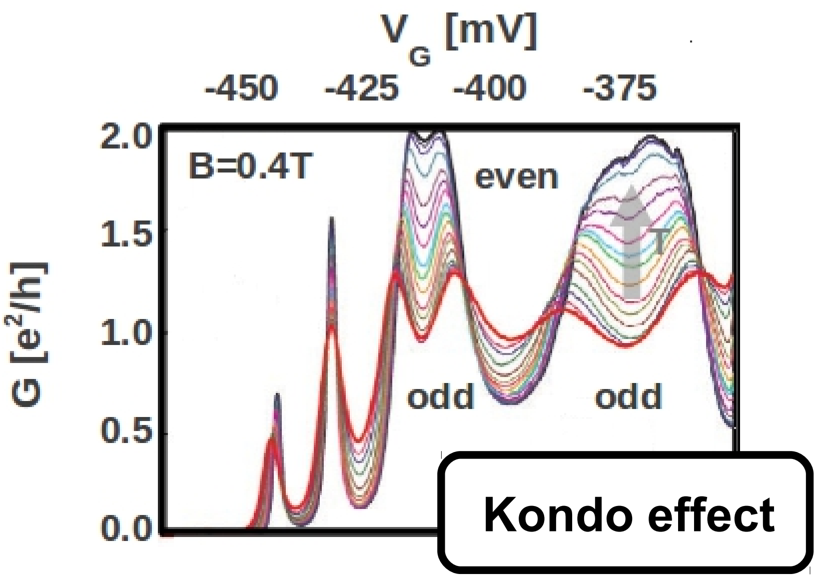
⇩

- fascinating phenomena (experiments)



Van der Wiel *et al.*, Science **289** (2000)

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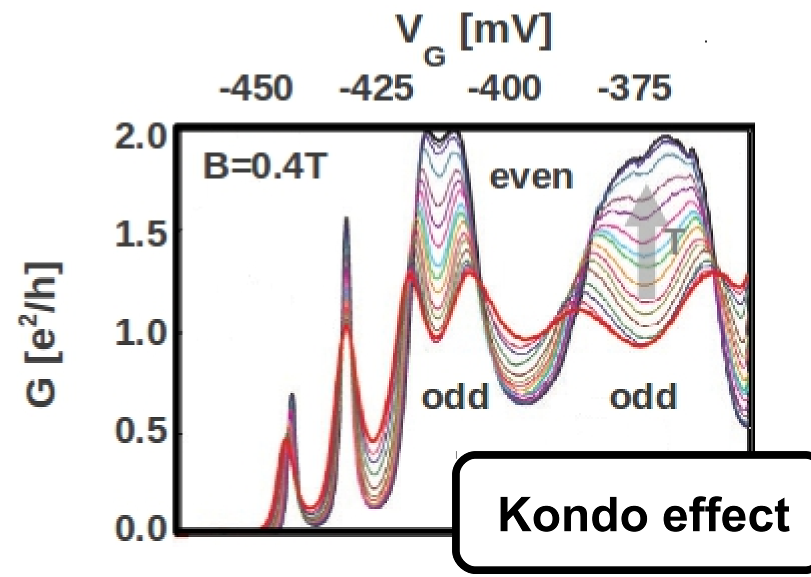

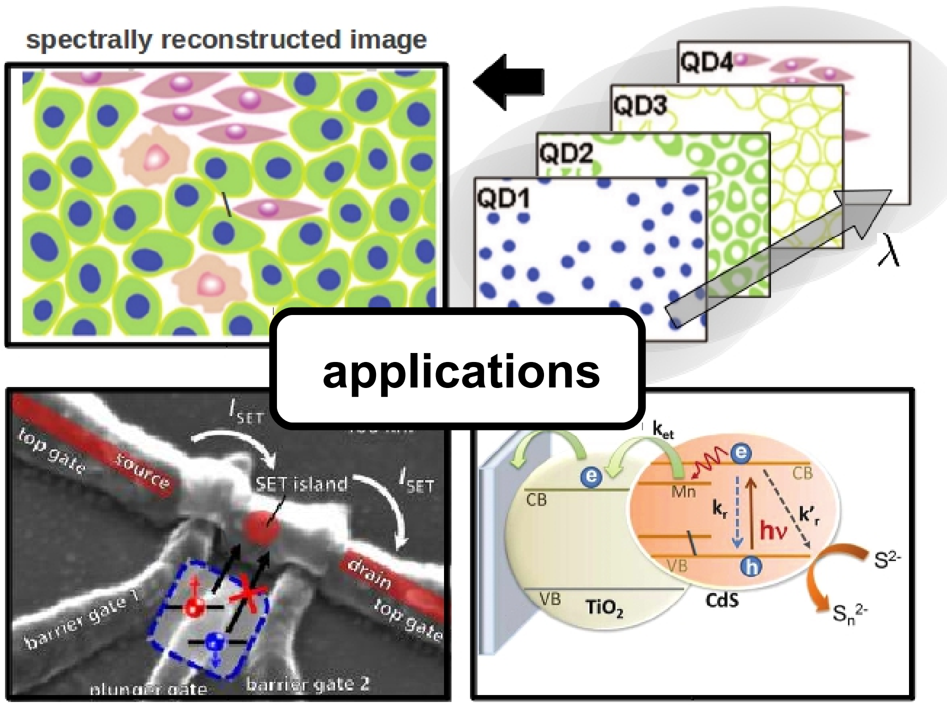

Van der Wiel *et al.*, Science **289** (2000)
True *et al.*, JMD **9** (2007)
Morello *et al.*, Nature **467** (2010)
Santra *et al.*, JACS **134** (2012)

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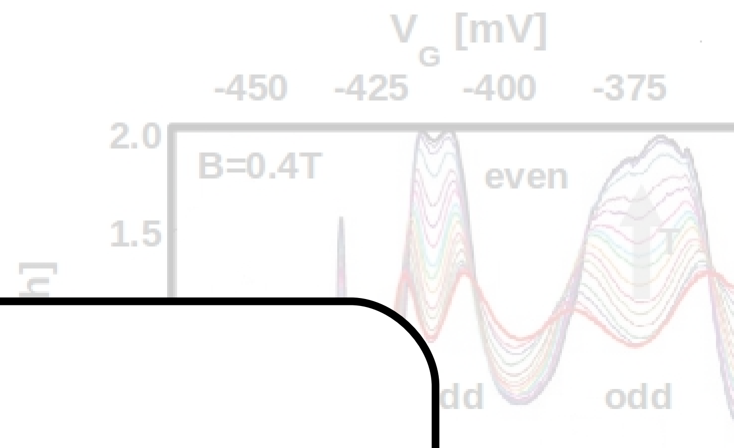


Kondo effect

qualitative understanding

...few suitable models...

► **frontier research:** development of tools for strongly correlated nanostructures

…ence **289** (2000)

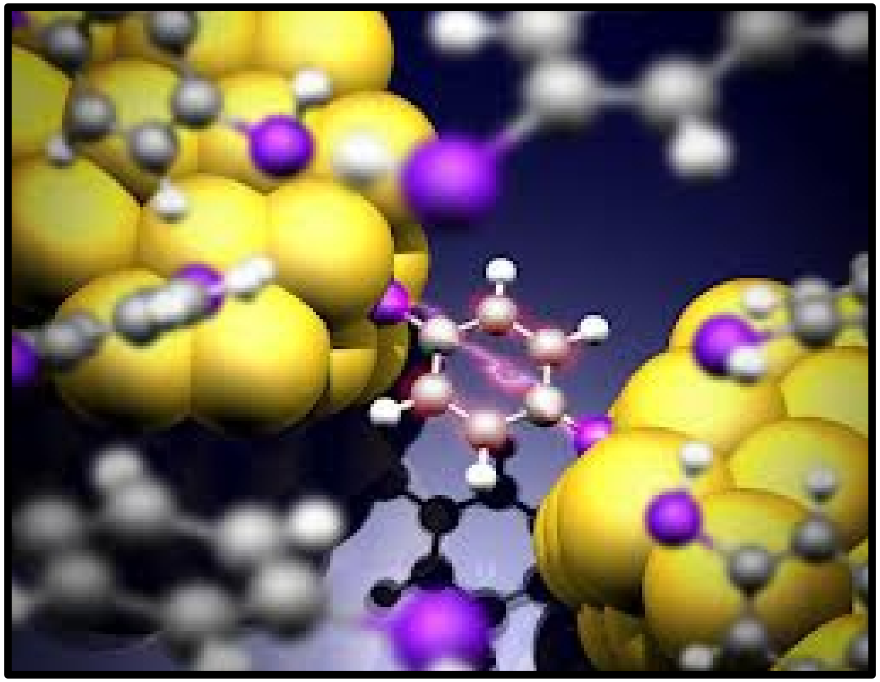
True *et al.*, JMD **9** (2007)

Morello *et al.*, Nature **467** (2010)

Santra *et al.*, JACS **134** (2012)

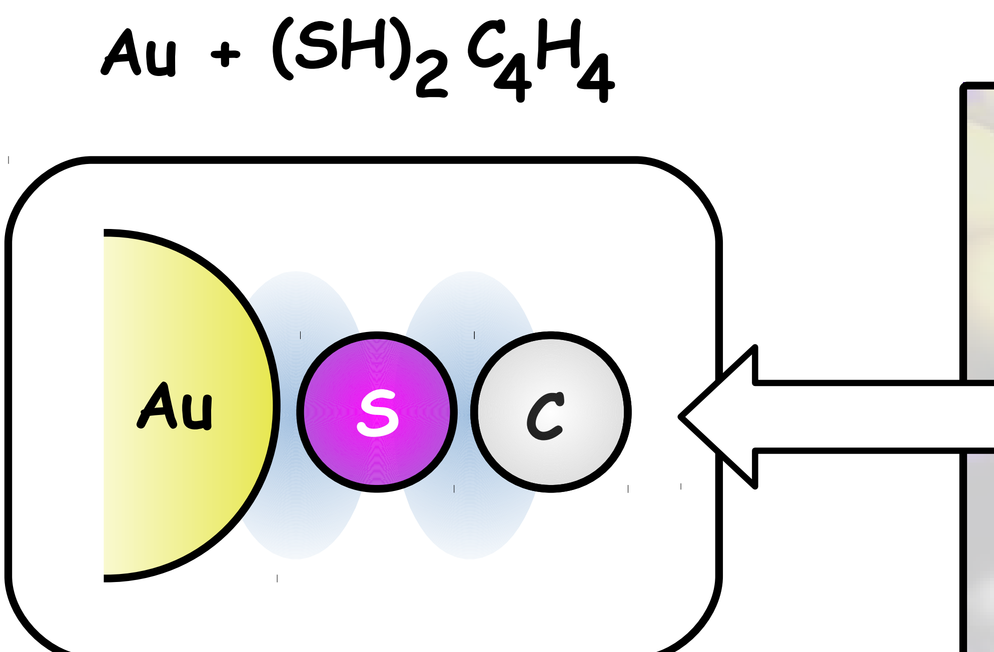
“staying in tune” with experiments?

1,4-BDT **molecular junction**

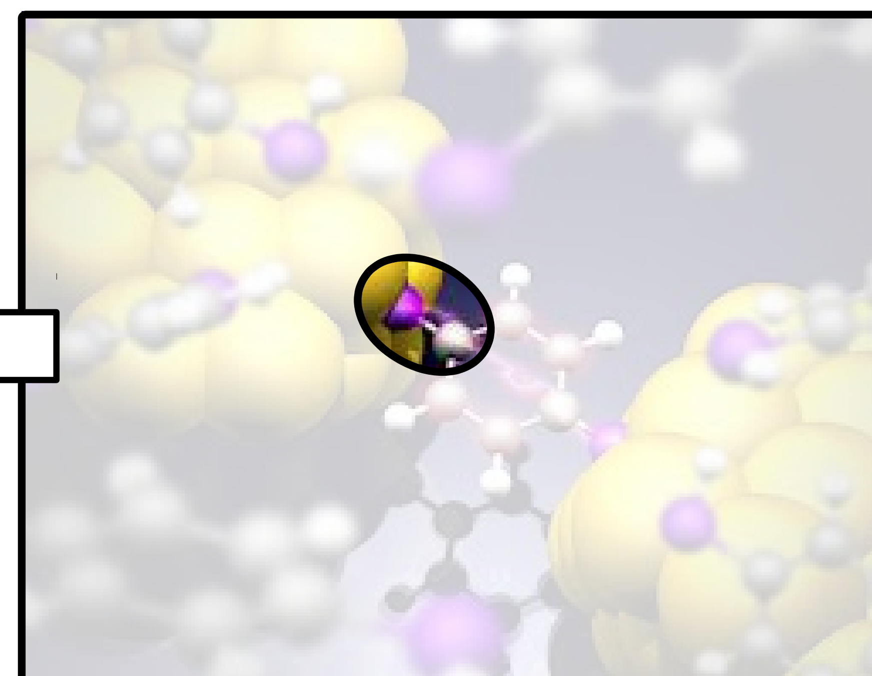


- **take “chemistry” into account**

  state-of-the-Art: quantum chemistry, DFT+NEGF



1,4-BDT **molecular junction**



- **take “chemistry” into account**

  state-of-the-Art: quantum chemistry, DFT+NEGF

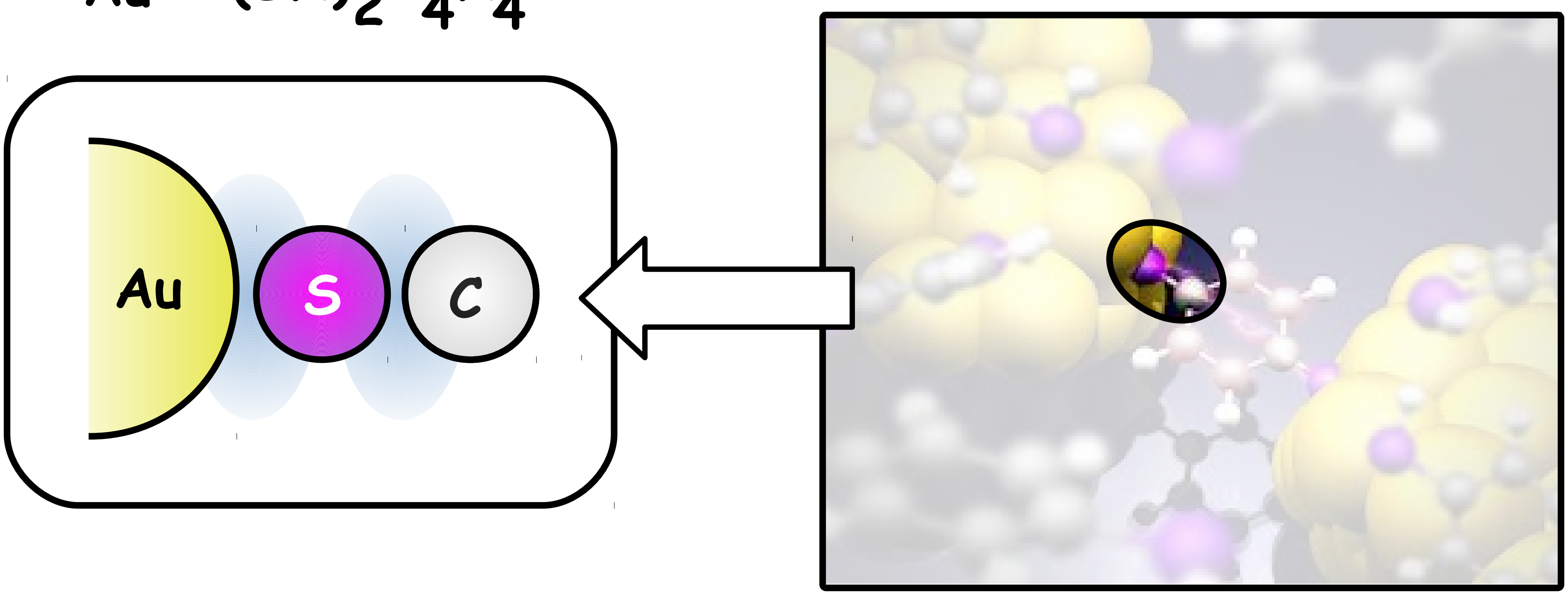
- **electronic correlations?**

  non-systematically

  doubtful perturbative approaches

$Au + (SH)_2 C_4H_4$

1,4-BDT **molecular junction**



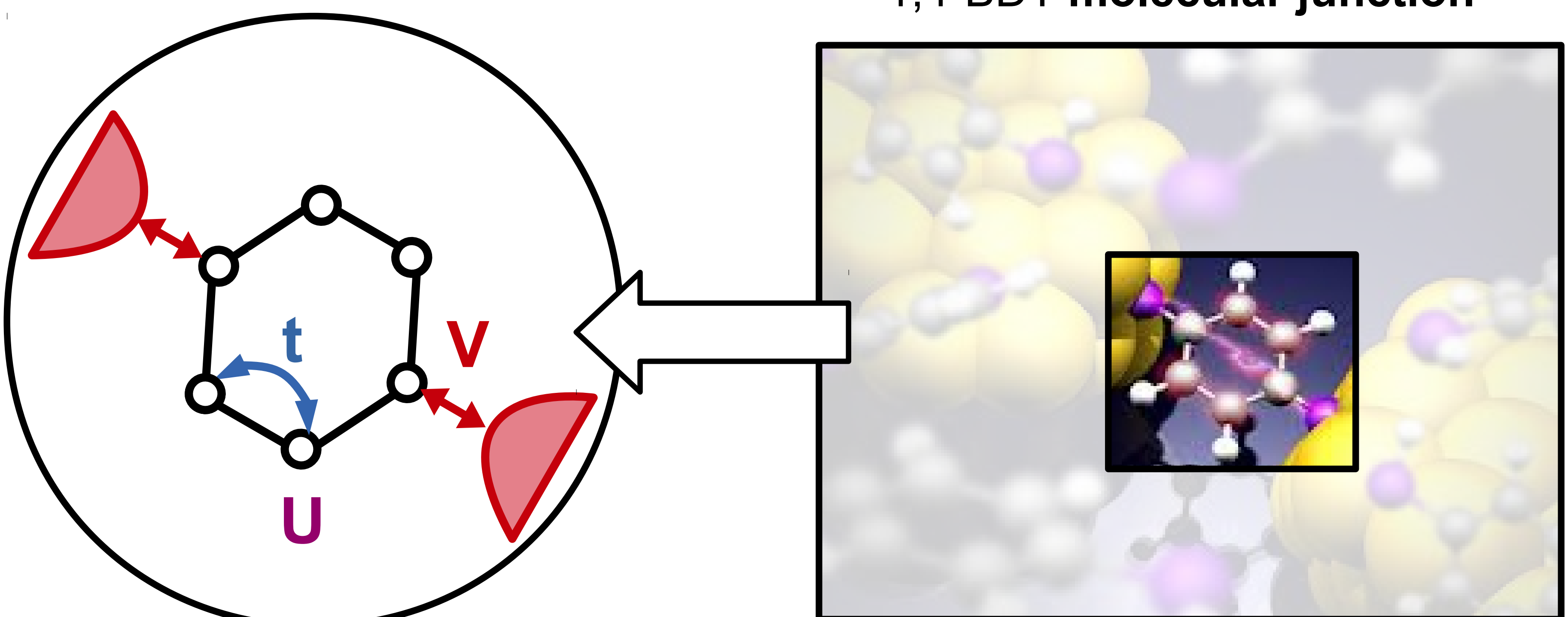
intro
methods
applications
outlook

- **understand electronic correlations**

caveat: realism may pretend more complex interaction

$$\mathcal{H} = -\sum_{ij\sigma} t_{ij} c_{i\sigma} c^{\dagger}_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$
$$+ \sum_{i\eta k\sigma} V_{i\eta k} \left(c^{\dagger}_{i\sigma} l_{\eta k\sigma} + \mathrm{h.c} \right) + \sum_{\eta k\sigma} \epsilon_{\eta k} l^{\dagger}_{k\sigma} l_{k\sigma}$$

1,4-BDT **molecular junction**

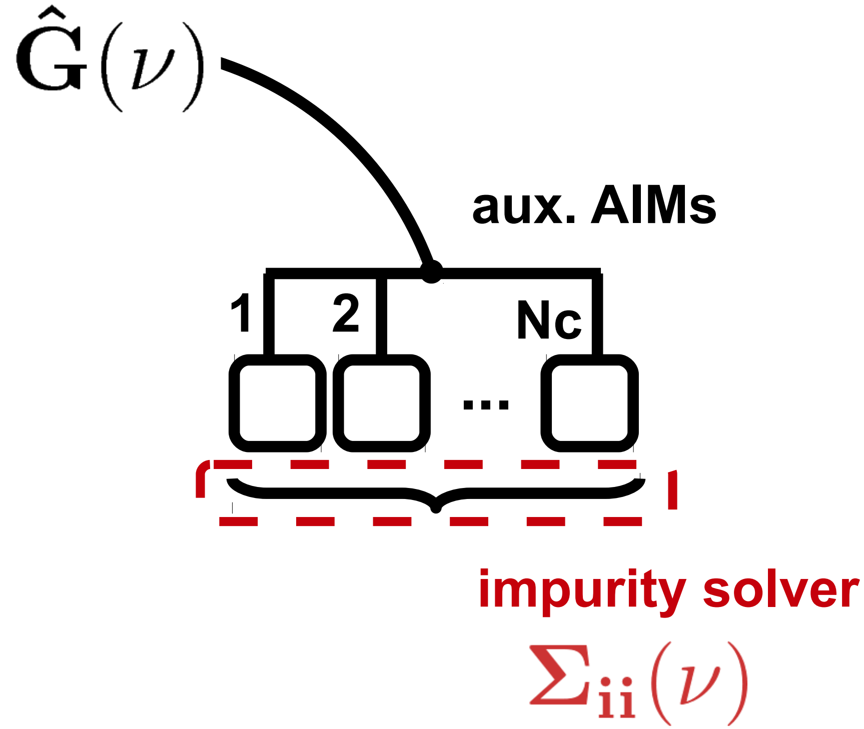


nano-DMFT

(1-particle self-consistent)

Valli *et al.*, PRL **104** (2010)

Valli *et al.*, PRB **86** (2012)



DMFT-like

Nc x Nc → Nc (1x1)

$$\mathcal{G}_{0i}^{-1} = \{G_{ii}\}^{-1} + \Sigma_{ii}$$

in the same spirit:

Potthoff & Nolting, PRB **60** (1999)

Biermann *et al.* PRL **87** (2001)

Snoek *et al.* NJP **10** (2008)

Jacob *et al.* PRL **103** (2009), PRB **82** (2010)

Schwabe *et al.* PRL **109** (2012)

Titvinidze *et al.* PRB **86** (2012)

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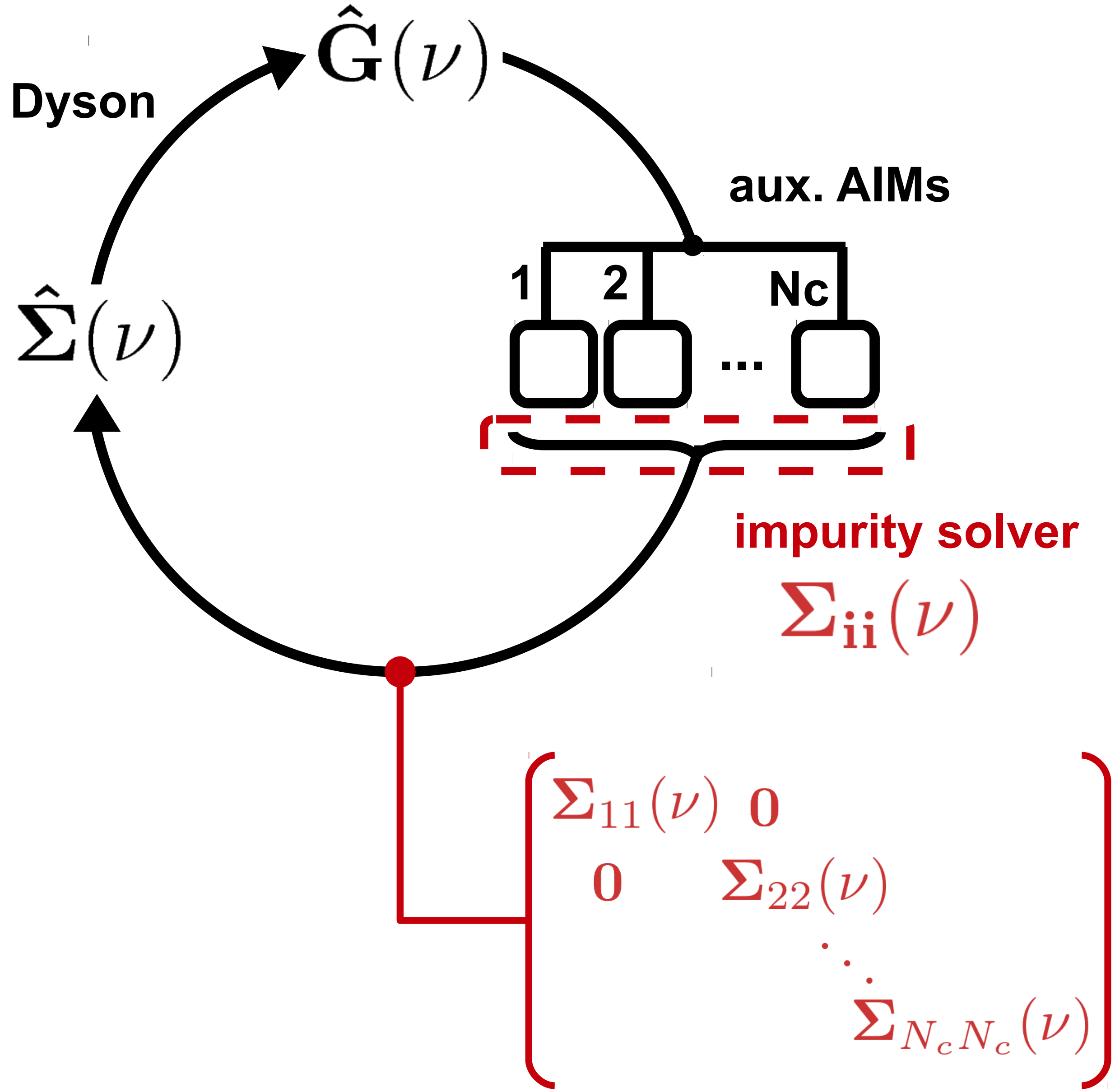

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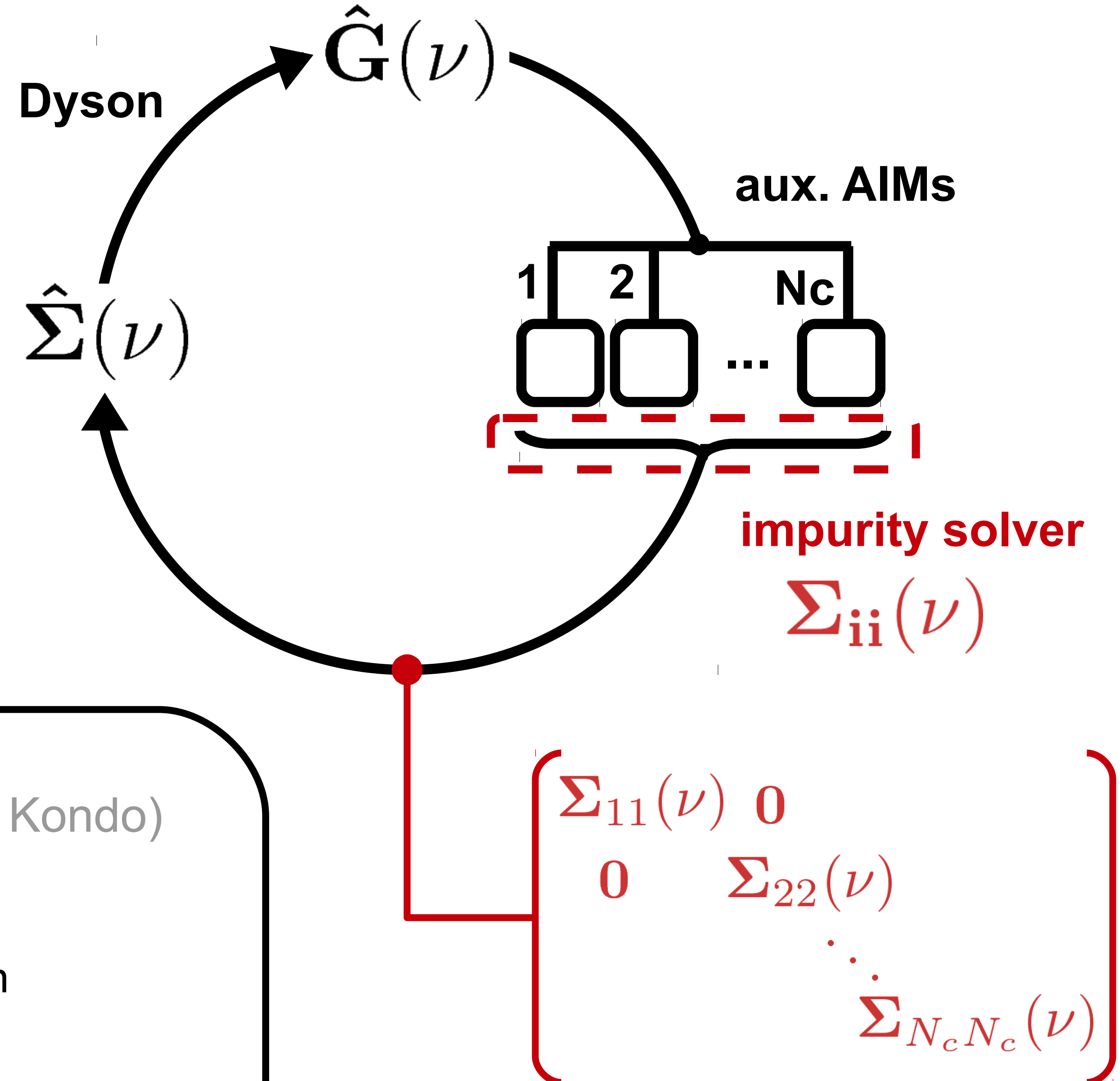
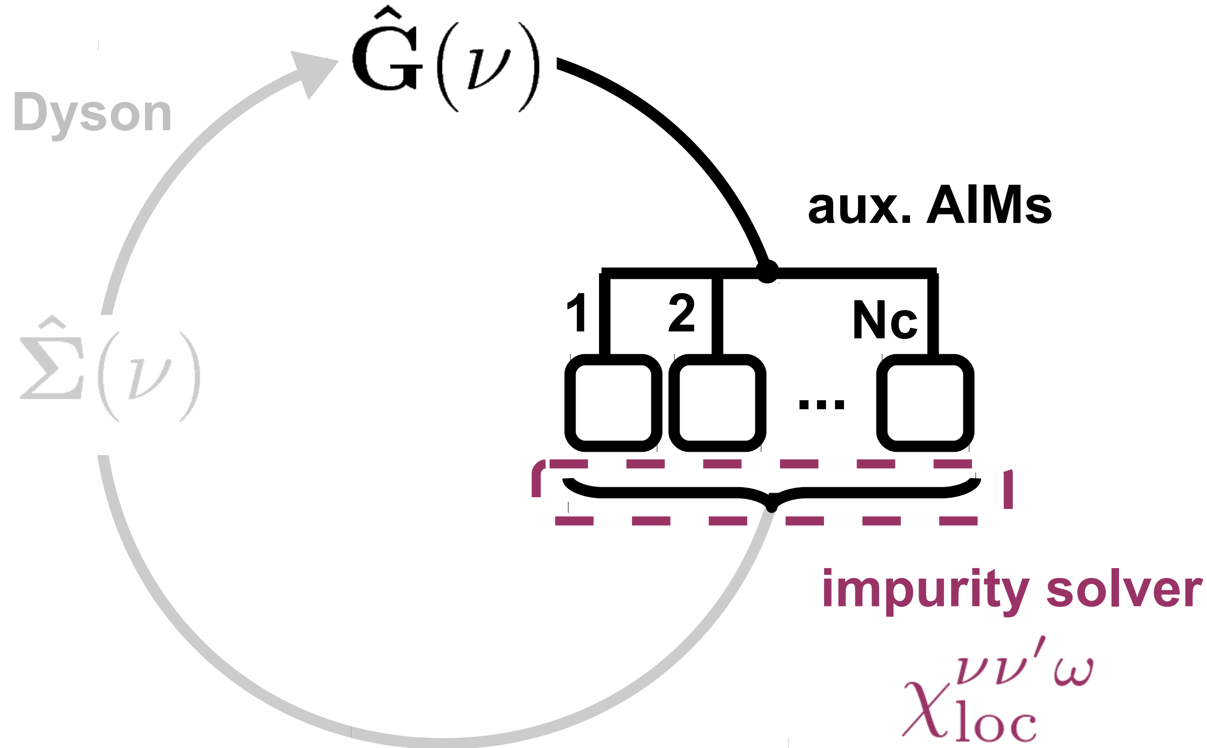


- ✔ **local** correlations (e.g., Kondo)
- ✔ flexible
  - structure implementation
  - solver-independent
- ✔ scales with Nineq ≤ Nc
- ✘ non-local correlations neglected

nano-DΓA

(2-particle self-consistent)



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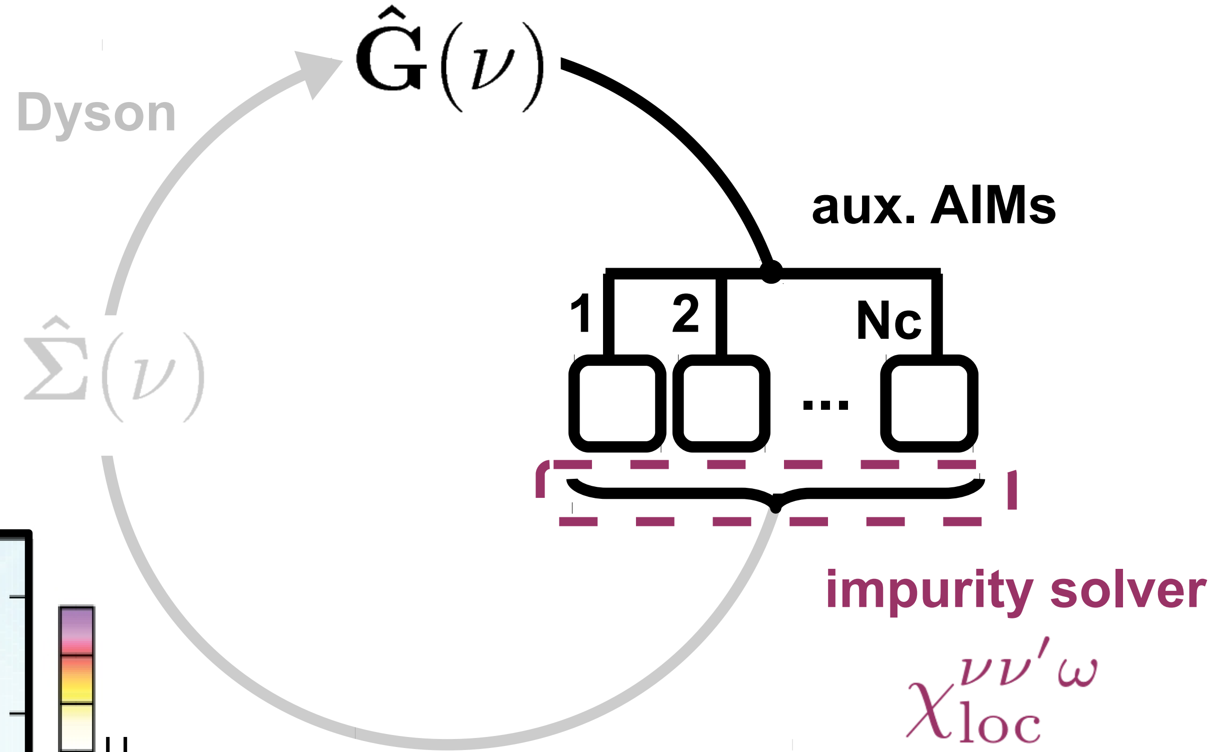
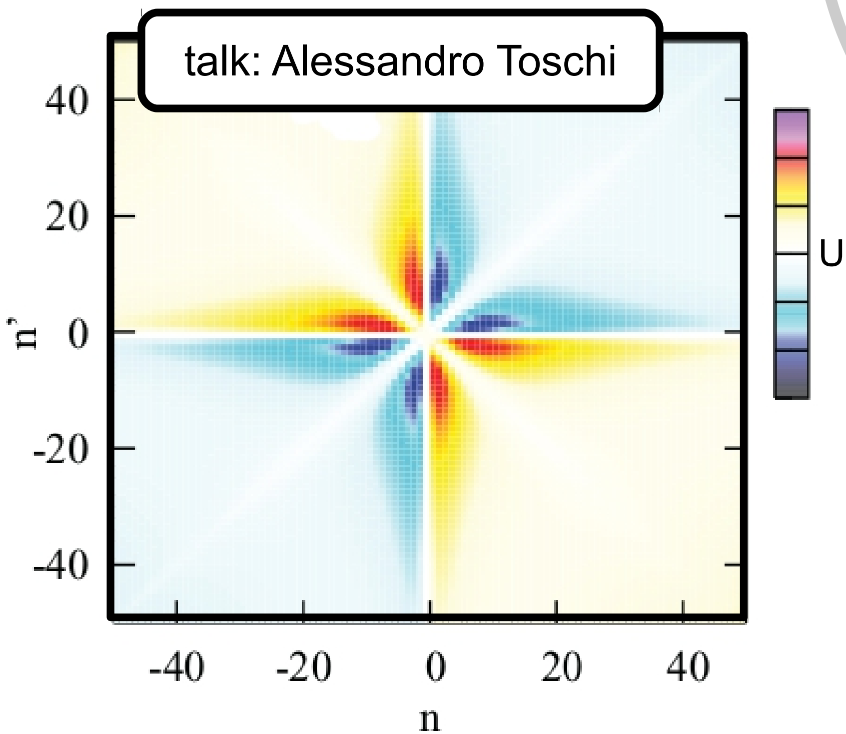
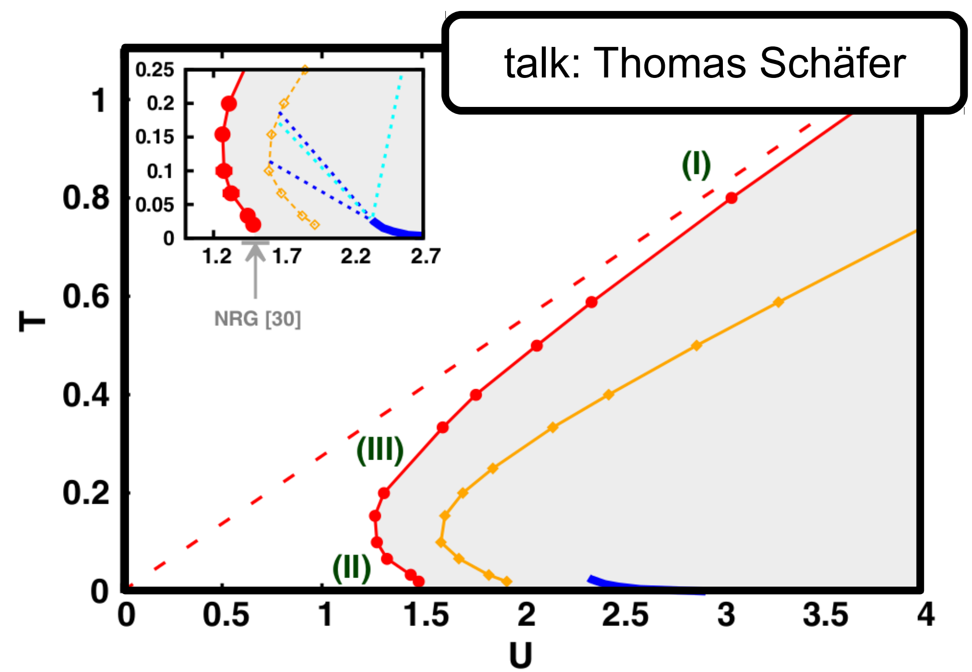
nano-DΓA

(2-particle self-consistent)





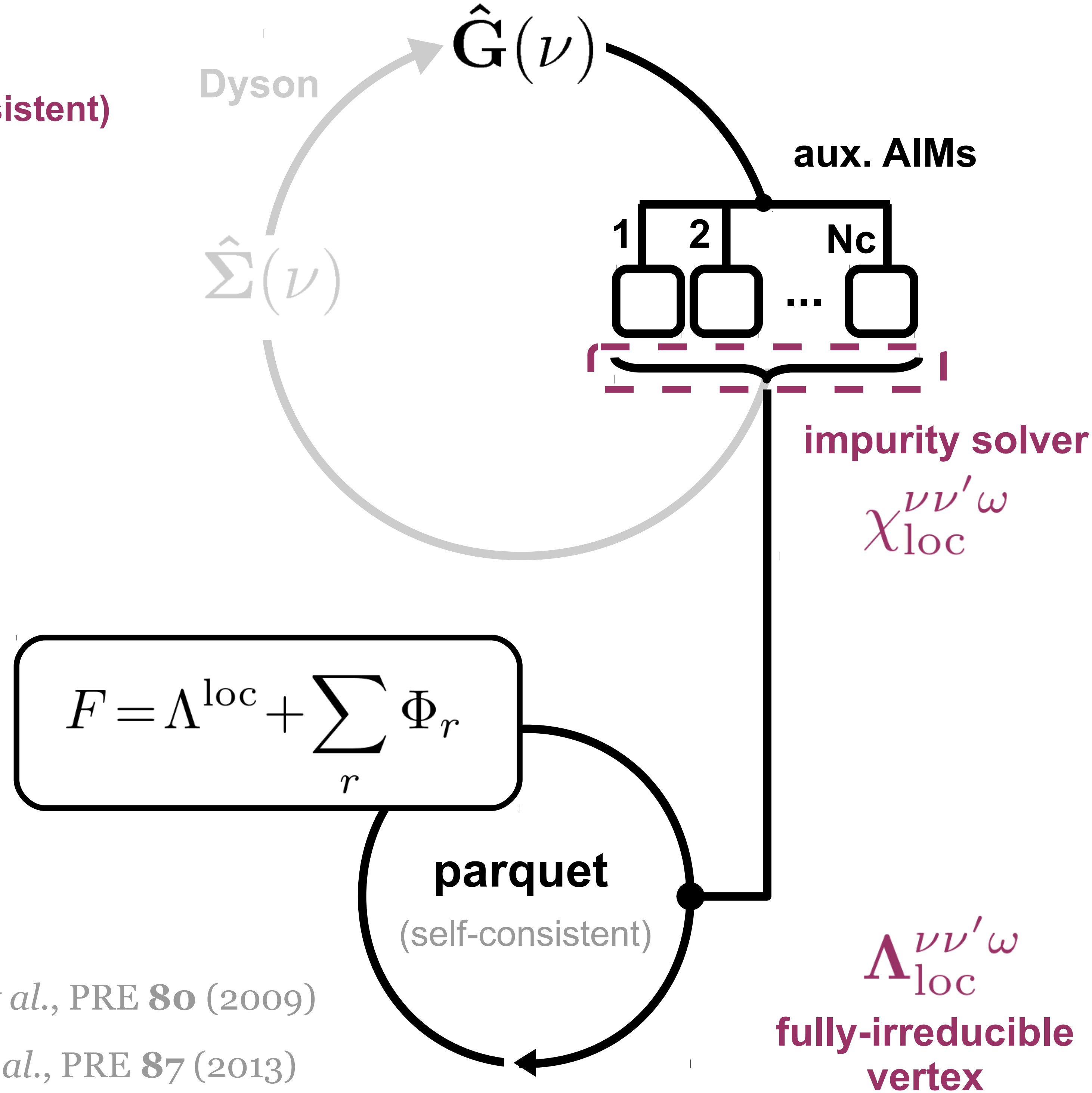


Rohringer, Valli, & Toschi. PRB **86** (2012)

Schäfer *et al.*, PRL **110** (2013)

nano-DΓA

(2-particle self-consistent)



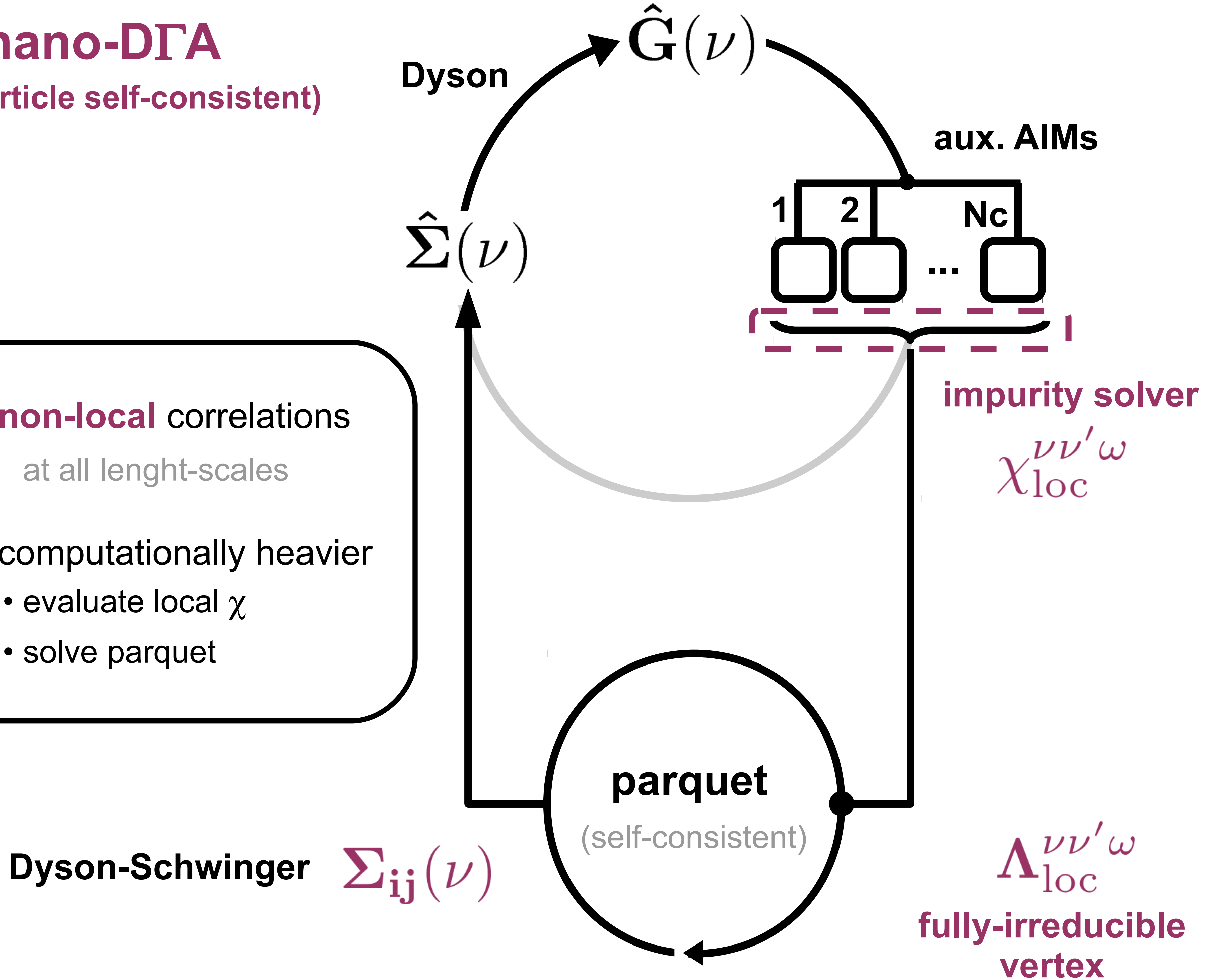
Yang *et al.*, PRE **80** (2009)

Tam *et al.*, PRE **87** (2013)

nano-DΓA

(2-particle self-consistent)

- ✔ **non-local** correlations
  at all lenght-scales
- ✖ computationally heavier
  - evaluate local χ
  - solve parquet



closed set of equations

- **parquet**
- **Bethe-Salpeter**
- **Dyson-Schwinger (eq. of motion)**
- **Dyson**

> “[...] alllow the vertex corrections and the self-energy to be calculated in a self-consistent manner, given the input of the fully irreducible vertex”

Tam *et al.*, PRE **87** (2013)

closed set of equations

- **parquet**
- Bethe-Salpeter
- Dyson-Schwinger (eq. of motion)
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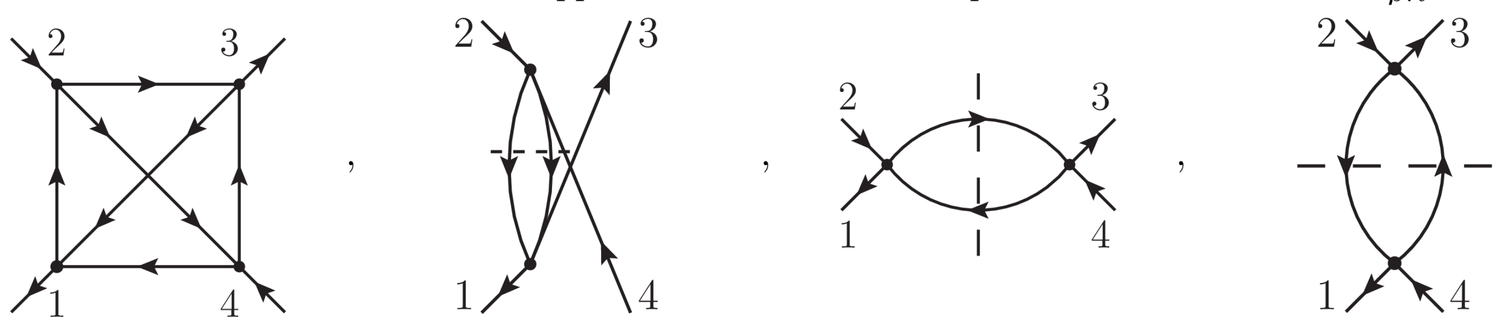
"decomposition" of two-particle vertex function

$$F = \Lambda + \Phi_{pp} + \Phi_{ph} + \Phi_{\overline{ph}}$$

e.g.:



all diagrams | **fully irreducible** | **reducible in a given (ph or pp) channel**

Rohringer, Valli, & Toschi PRB **86** (2012)

closed set of equations

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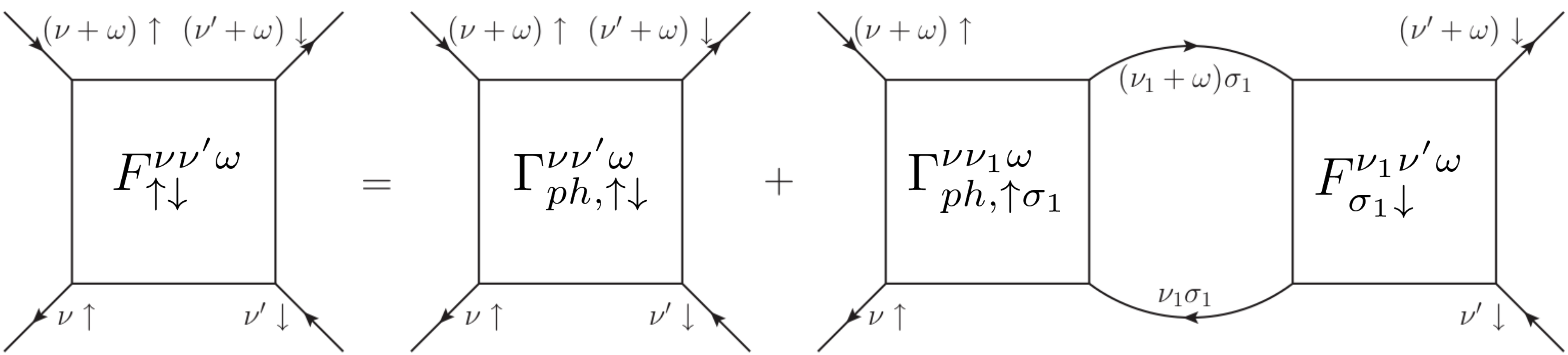

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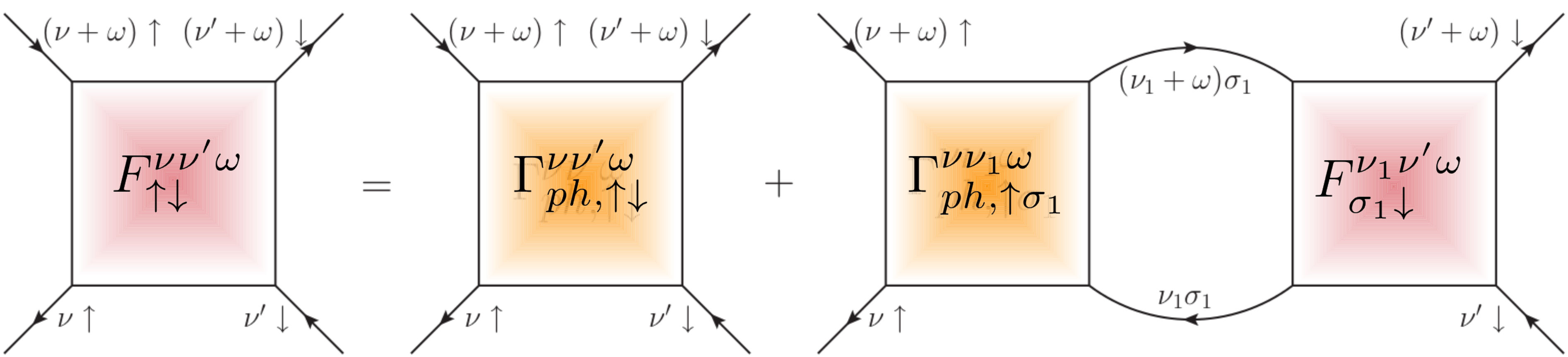

irreducible

Rohringer, Valli, & Toschi PRB **86** (2012)

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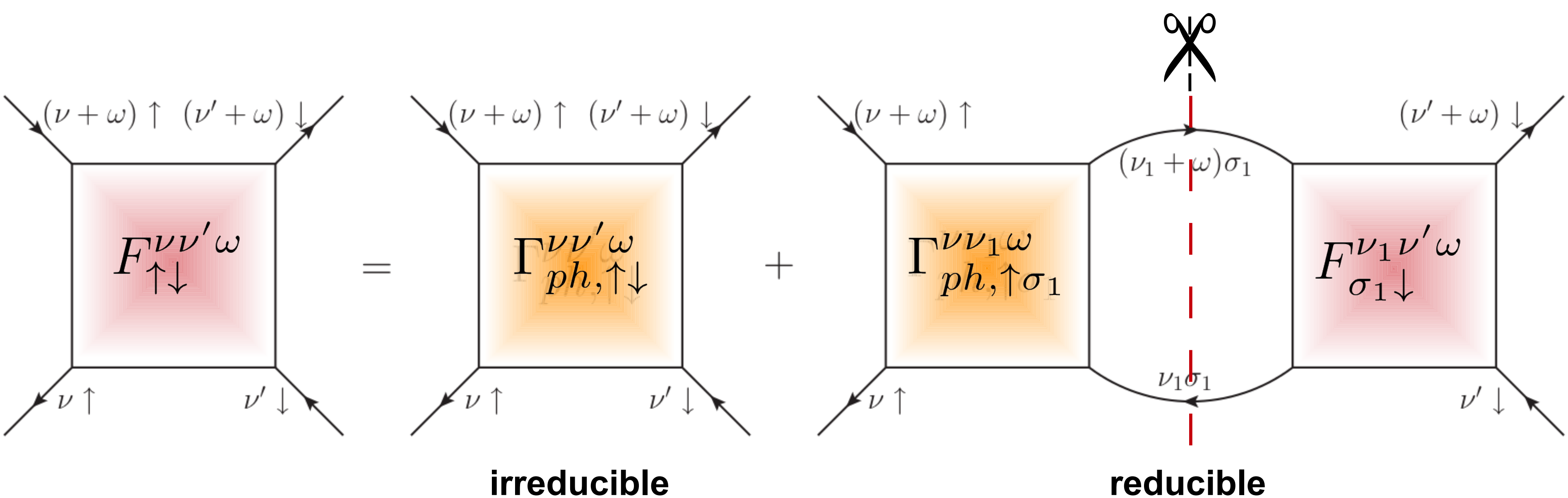

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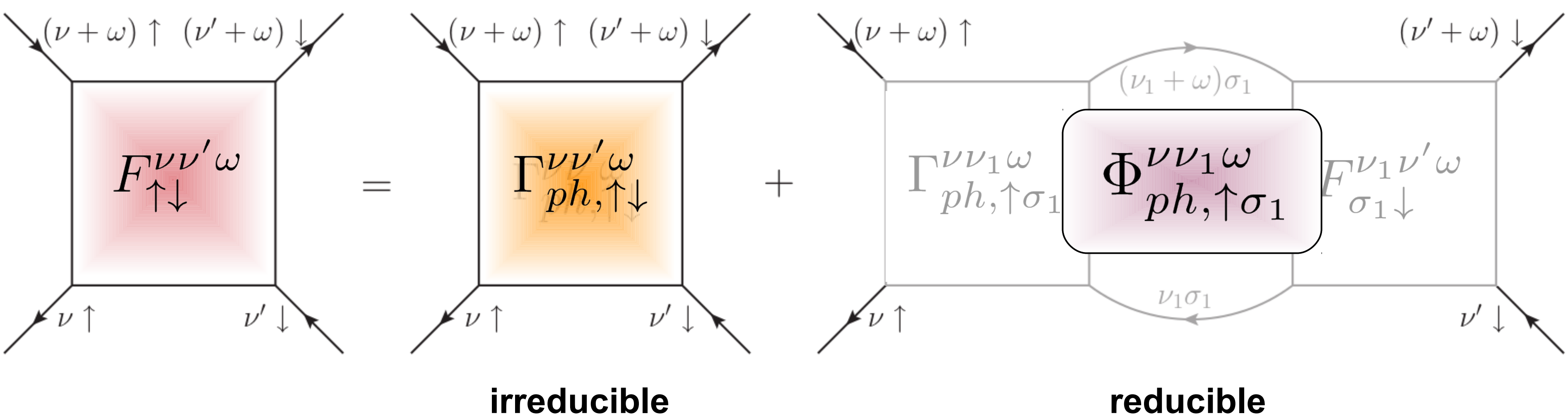

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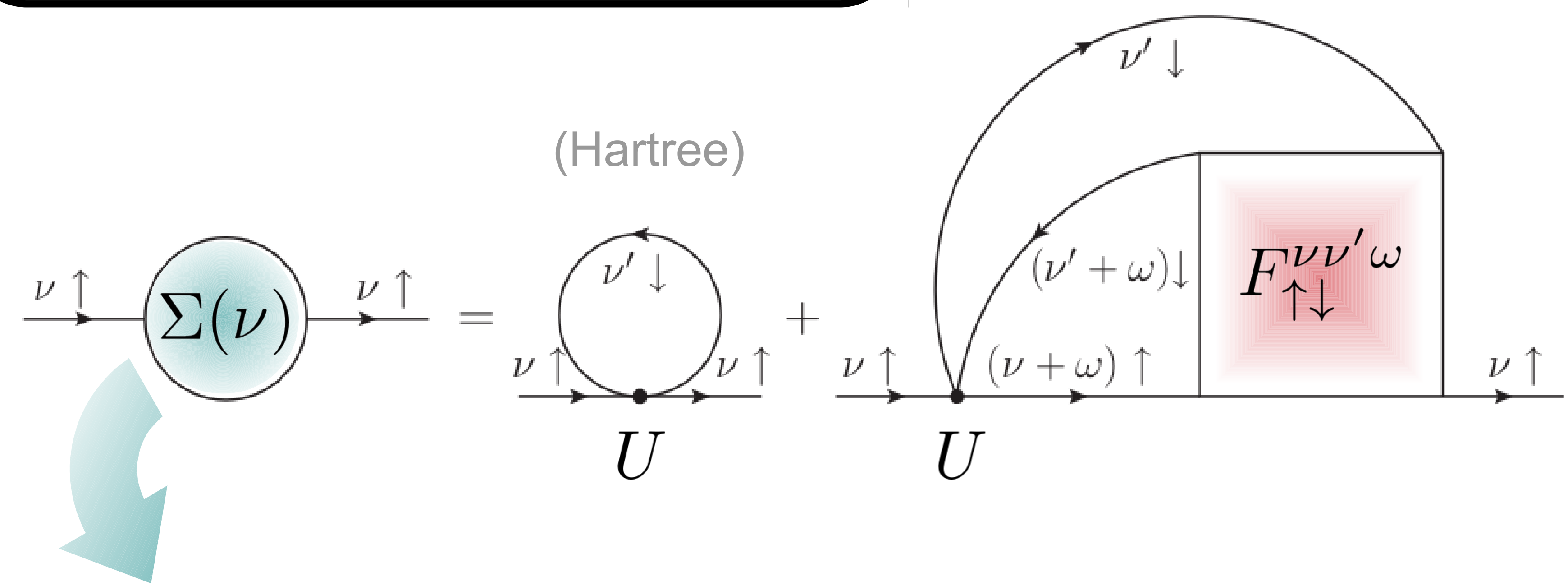


$$G = \left(G_0^{-1} - \Sigma\right)^{-1}$$

Rohringer, Valli, & Toschi PRB **86** (2012)

ϵ_k, Σ

Dyson

$$G = \left(G_0^{-1} - \Sigma\right)^{-1}$$

solving the parquet: flowchart

ϵ_k, Σ

Dyson

$$G = (G_0^{-1} - \Sigma)^{-1}$$

$$\chi_0 = -\beta G G$$

parquet

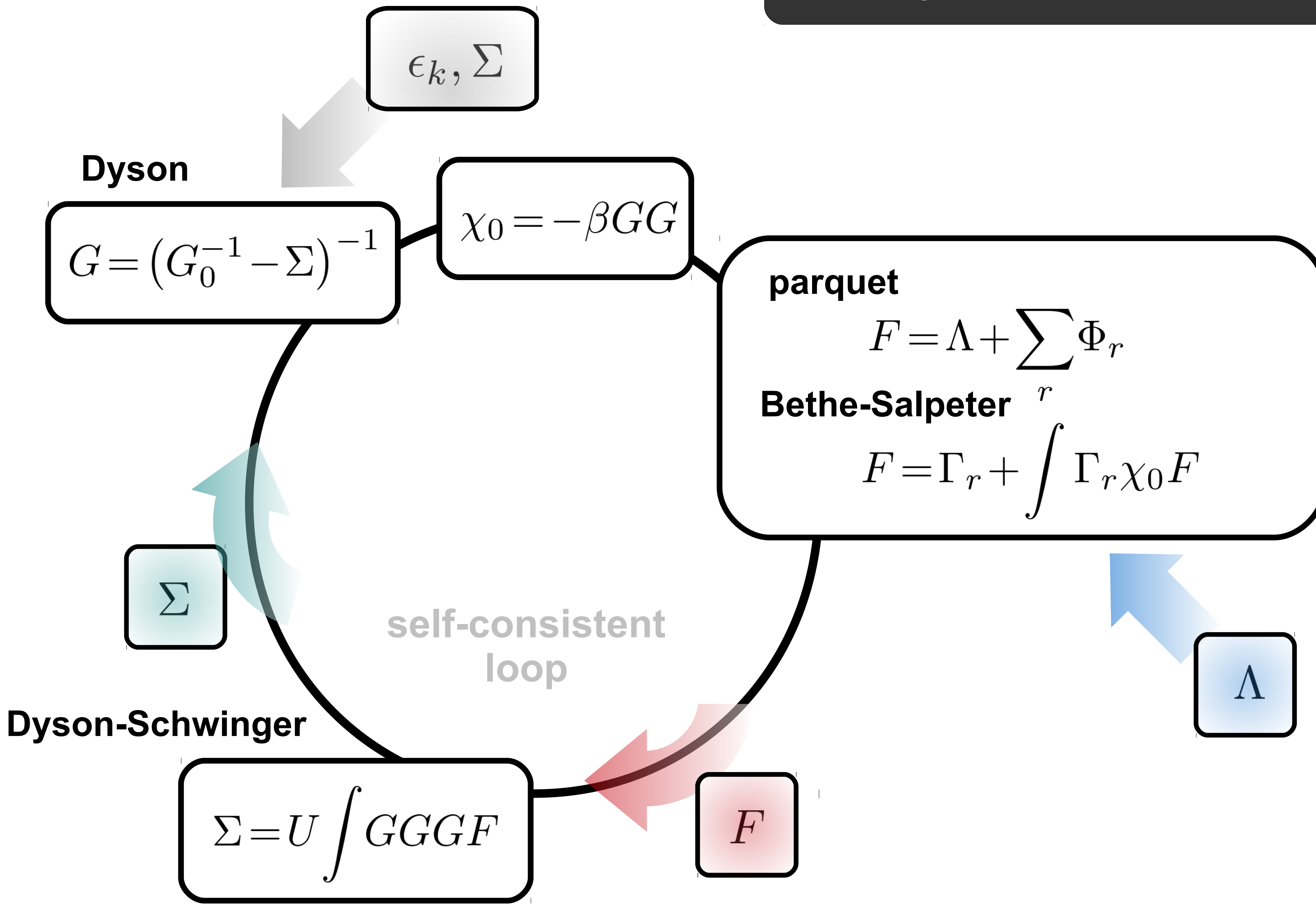
$$F = \Lambda + \sum_r \Phi_r$$

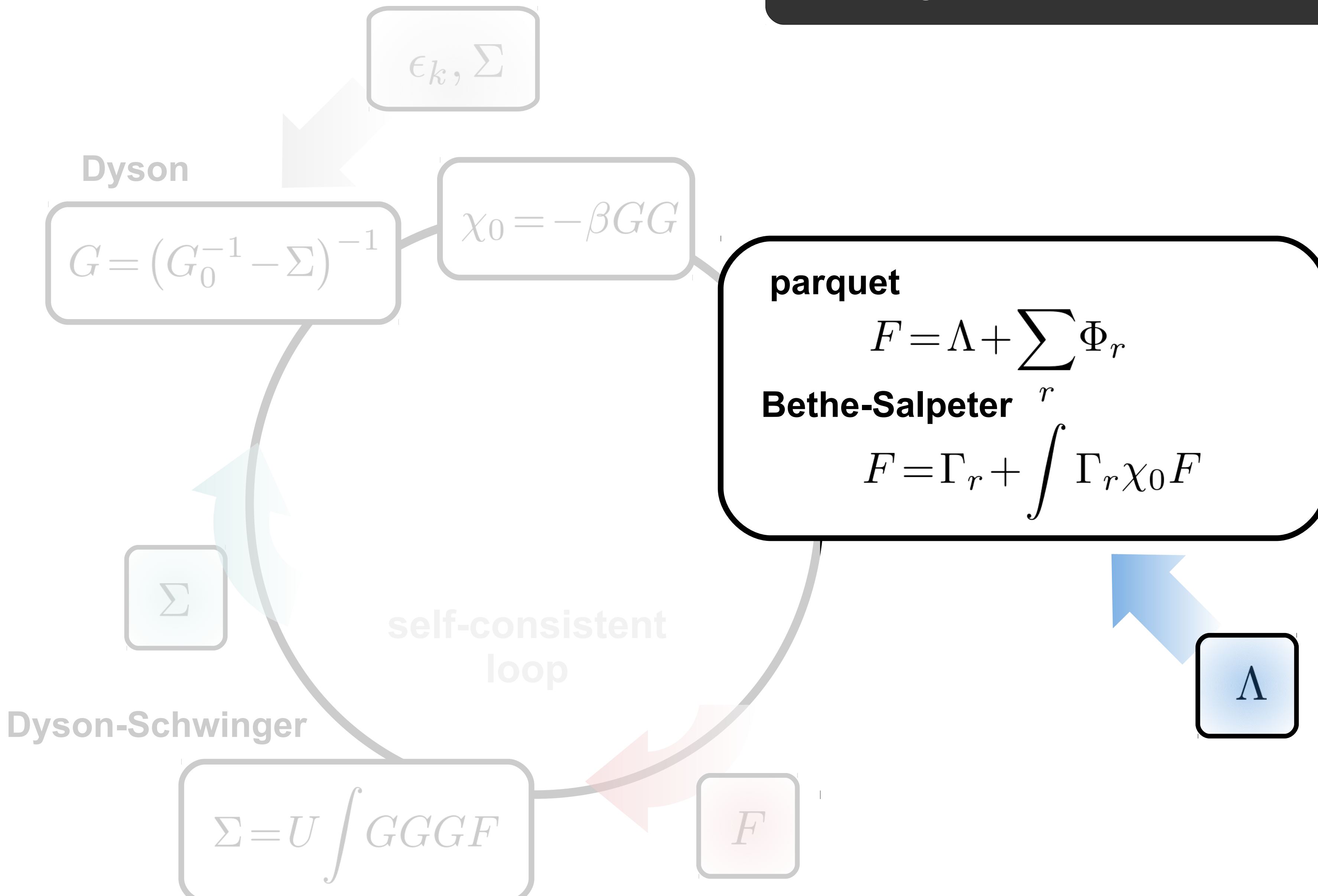
Bethe-Salpeter

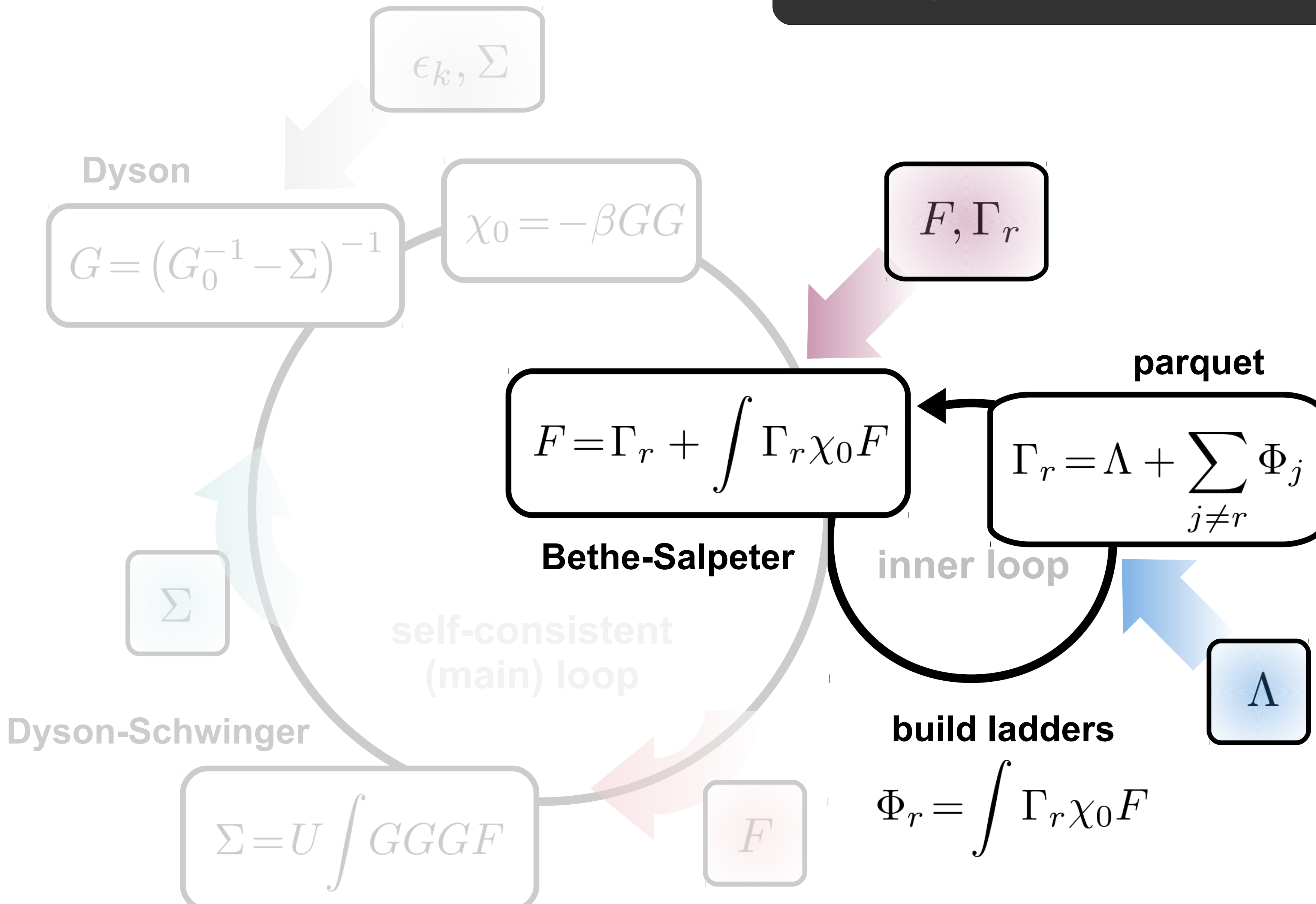
$$F = \Gamma_r + \int \Gamma_r \chi_0 F$$

Λ

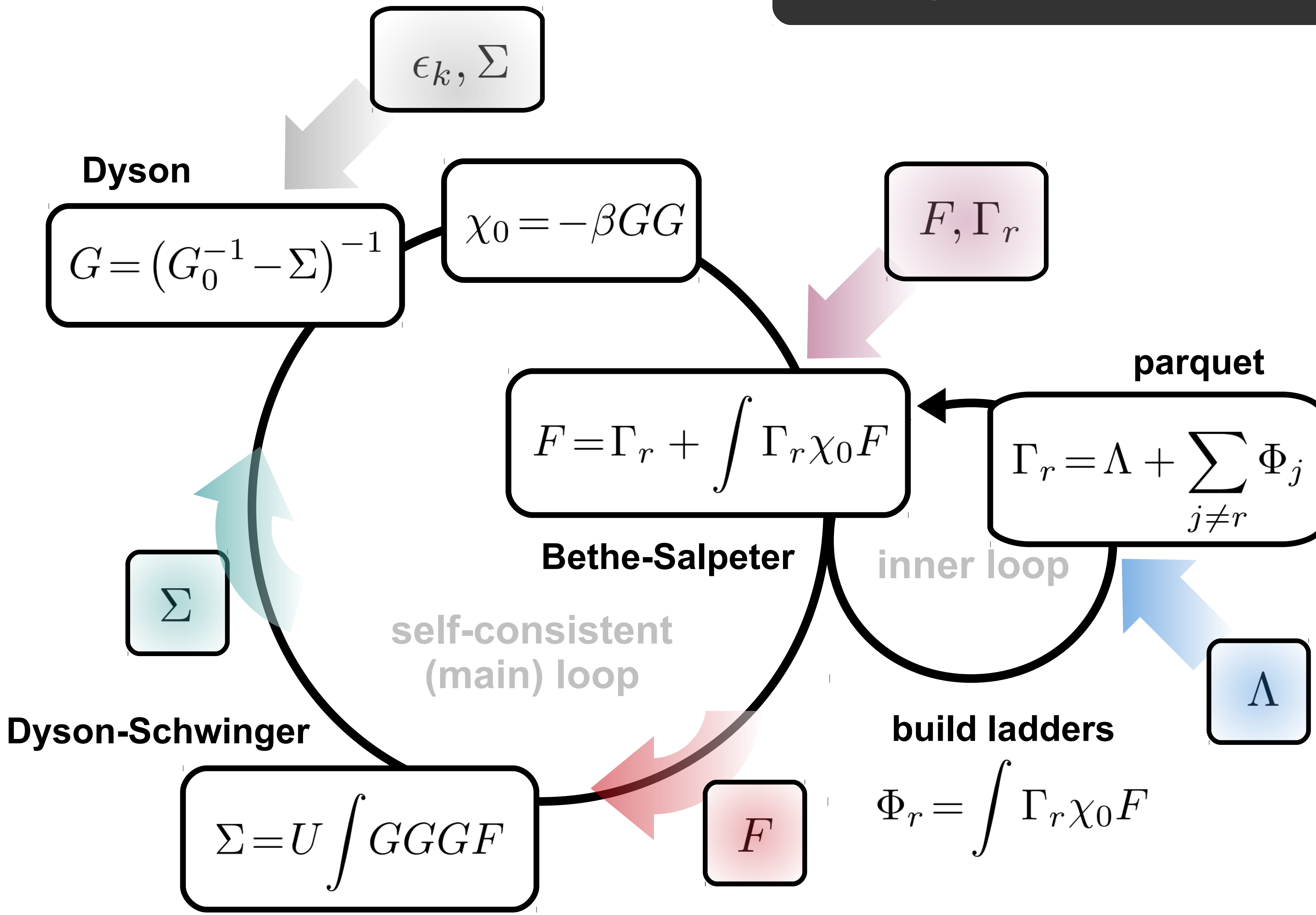
solving the parquet: flowchart




ϵ_k, Σ
Dyson
$G = (G_0^{-1} - \Sigma)^{-1}$
$\chi_0 = -\beta GG$
parquet
$F = \Lambda + \sum_r \Phi_r$
Bethe-Salpeter
$F = \Gamma_r + \int \Gamma_r \chi_0 F$
Σ
self-consistent loop
Λ
Dyson-Schwinger
$\Sigma = U \int GGGF$
F


ϵ_k, Σ
Dyson
$G = (G_0^{-1} - \Sigma)^{-1}$
$\chi_0 = -\beta GG$
F, Γ_r
parquet
$F = \Gamma_r + \int \Gamma_r \chi_0 F$
$\Gamma_r = \Lambda + \sum_{j \neq r} \Phi_j$
Bethe-Salpeter
inner loop
Σ
self-consistent (main) loop
Λ
build ladders
$\Phi_r = \int \Gamma_r \chi_0 F$
Dyson-Schwinger
$\Sigma = U \int GGGF$
F

solving the parquet: flowchart



take into account competing instabilities

non-locality of each channel, neglected in ladder-DΓA

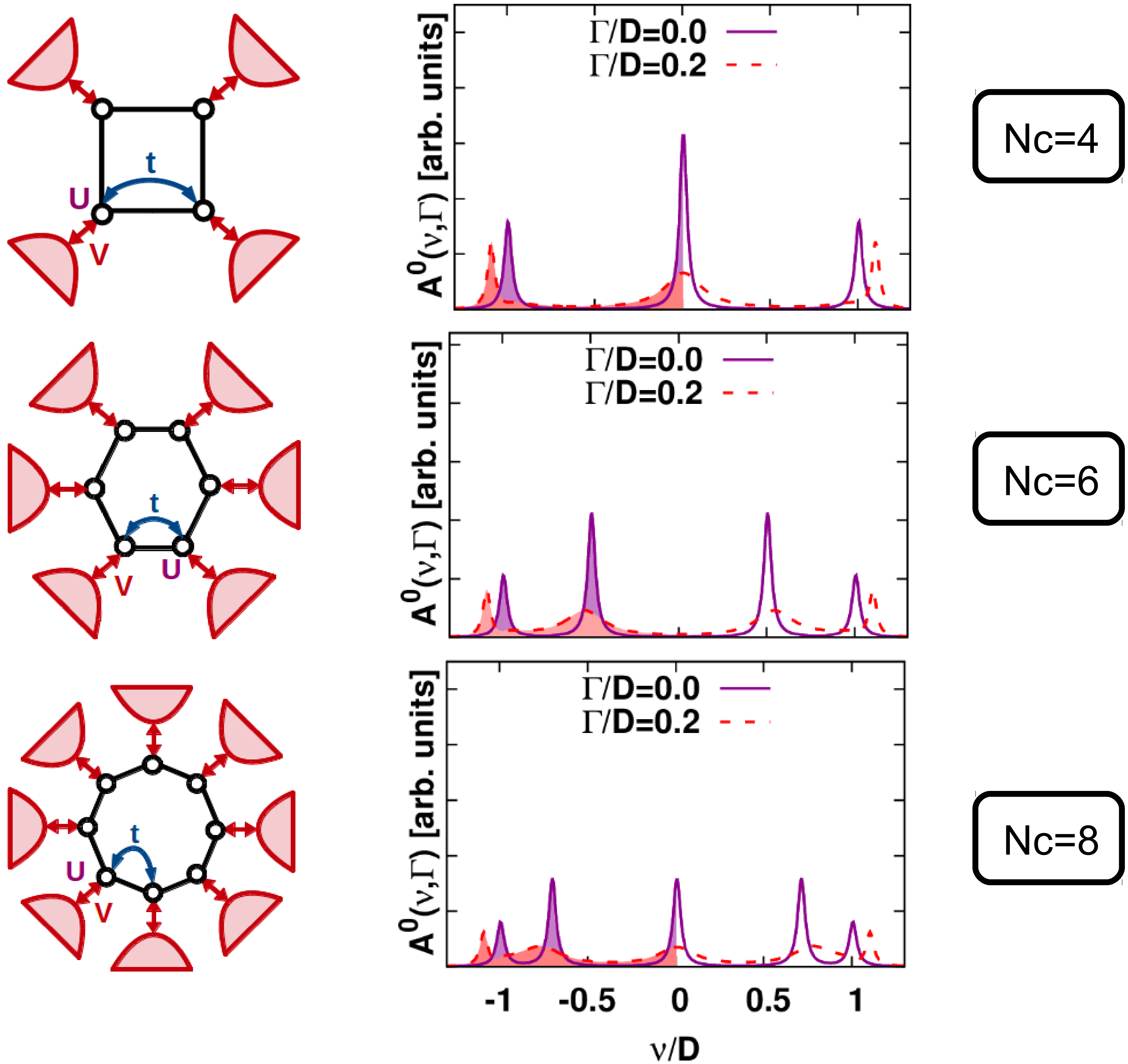
huge memory requirements: $F^{\nu\nu'\omega}_{kk'q}$ $\Gamma^{\nu\nu'\omega}_{kk'q}$ $\Phi^{\nu\nu'\omega}_{kk'q}$

- Nc (Nk): system size
- nf, nb: frequency range (critical to invert Bethe-Salpeter)

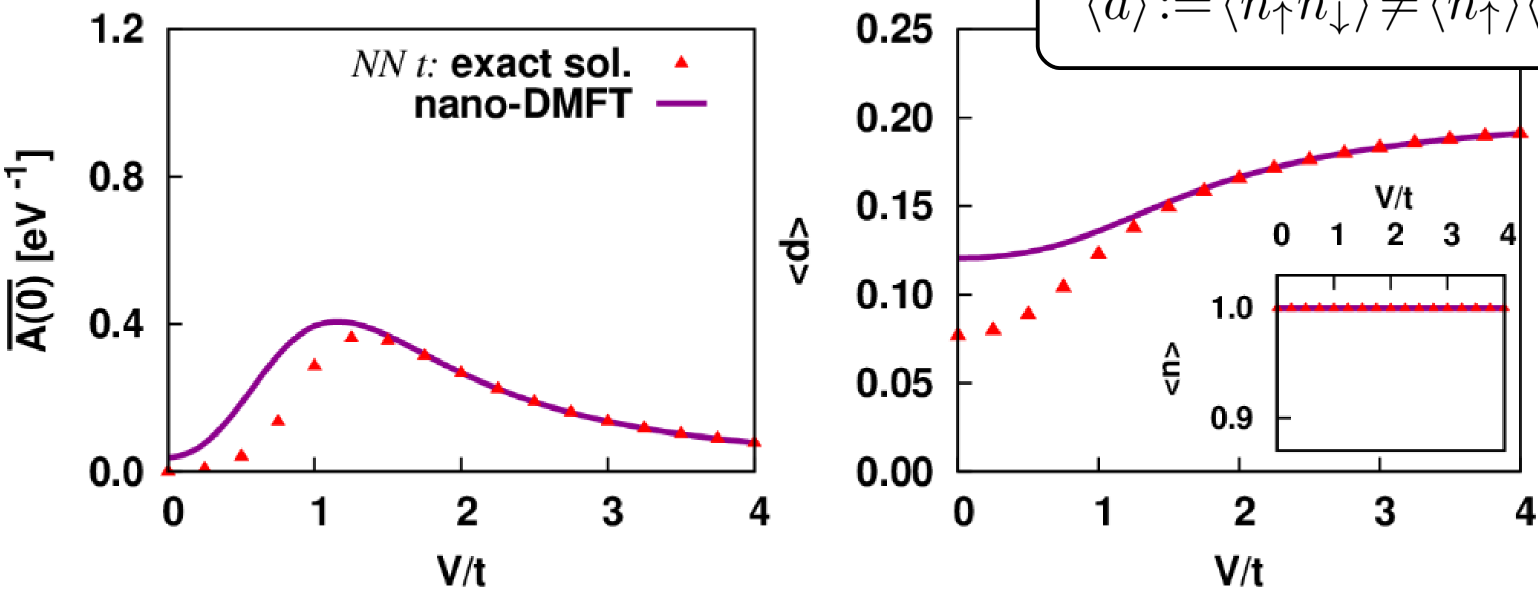
numerical instabilities

- strong coupling
- low temperature

local & non-local correlations in Q1D rings



nano-DMFT @ work



$$\langle d \rangle := \langle n_\uparrow n_\downarrow \rangle \neq \langle n_\uparrow \rangle \langle n_\downarrow \rangle$$

parameters:

U/D=2.5

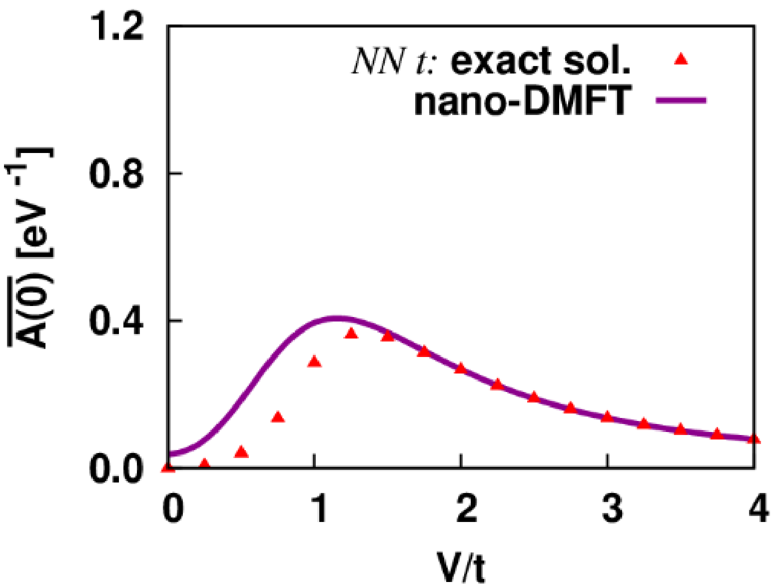
T/D=0.025

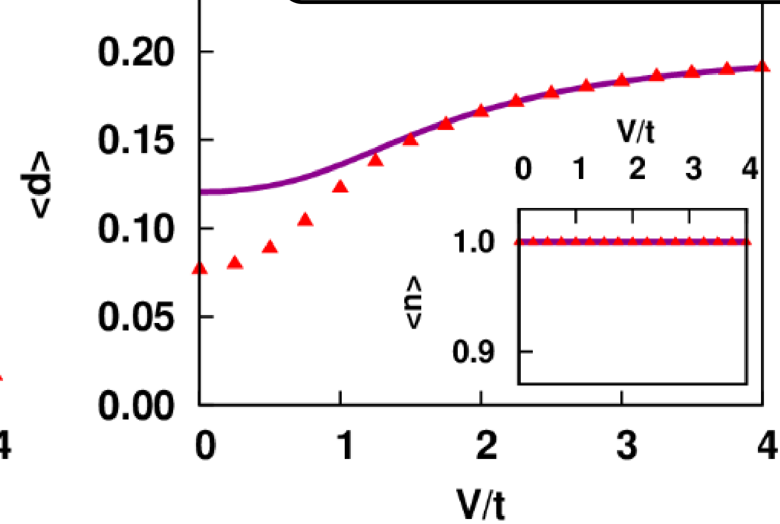
impurity solver:

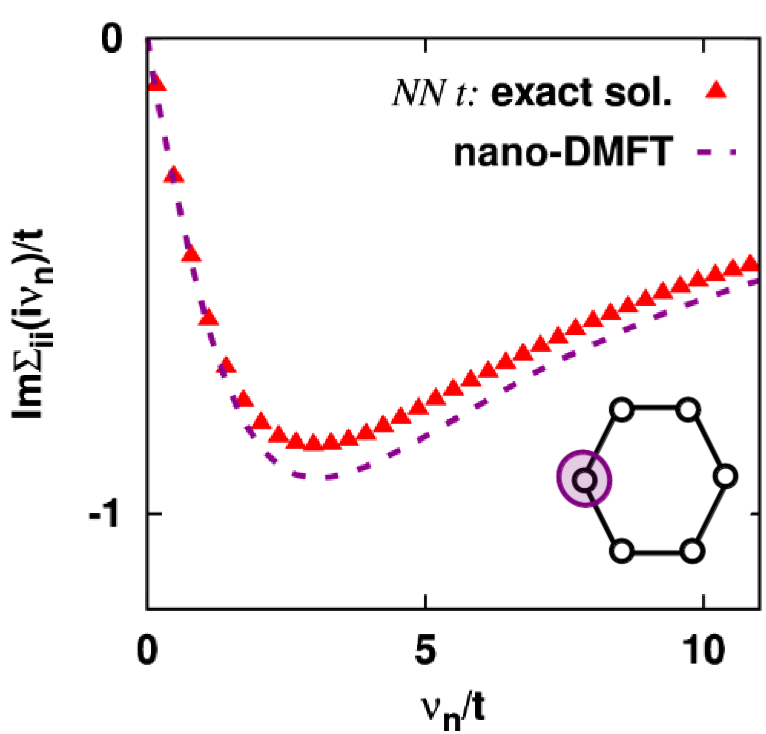
Hirsch-Fye QMC (both nano-DMFT & exact sol.)

nano-DMFT @ work

$$\langle d \rangle := \langle n_\uparrow n_\downarrow \rangle \neq \langle n_\uparrow \rangle \langle n_\downarrow \rangle$$

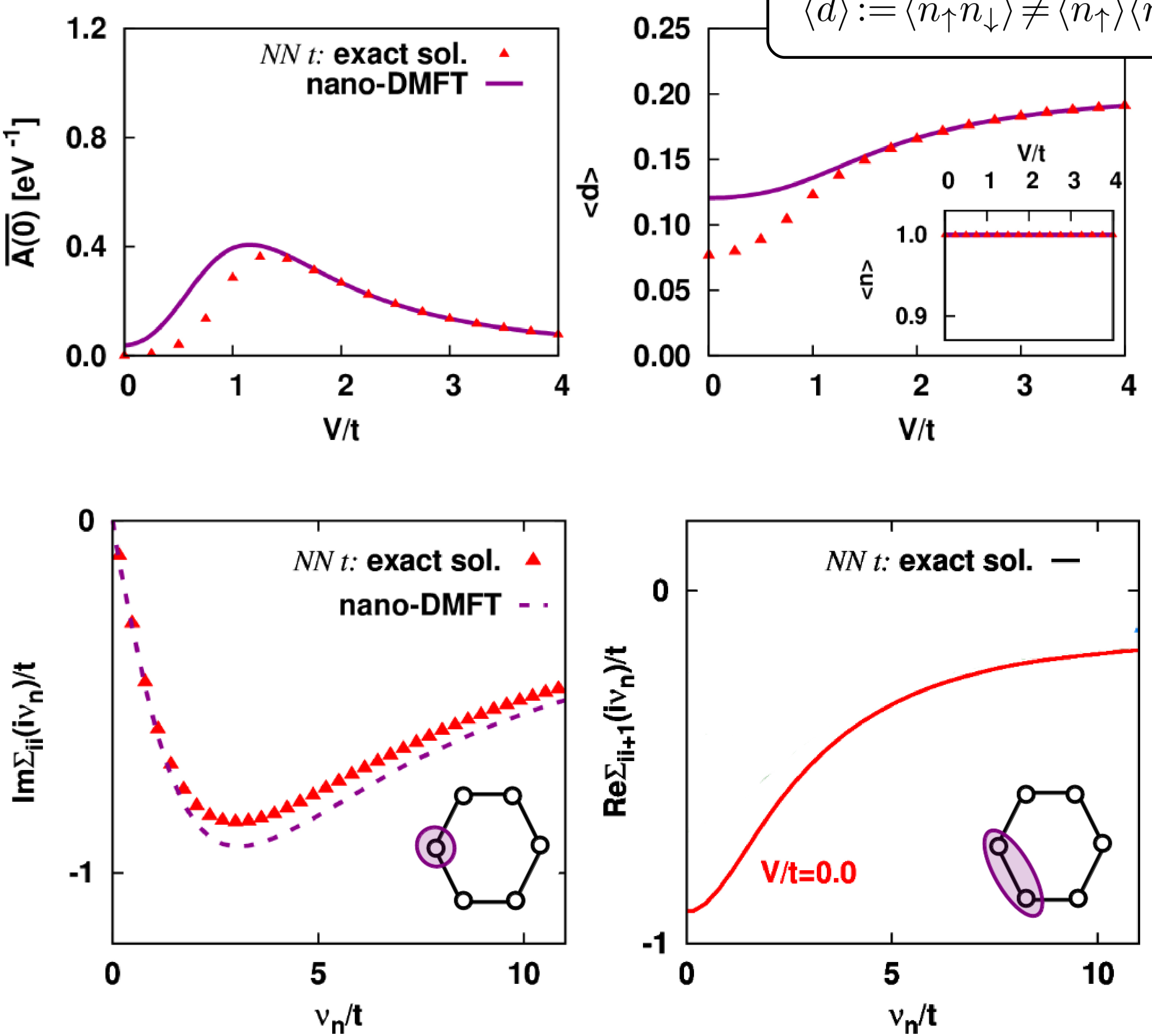






nano-DMFT @ work

$$\langle d \rangle := \langle n_{\uparrow} n_{\downarrow} \rangle \neq \langle n_{\uparrow} \rangle \langle n_{\downarrow} \rangle$$



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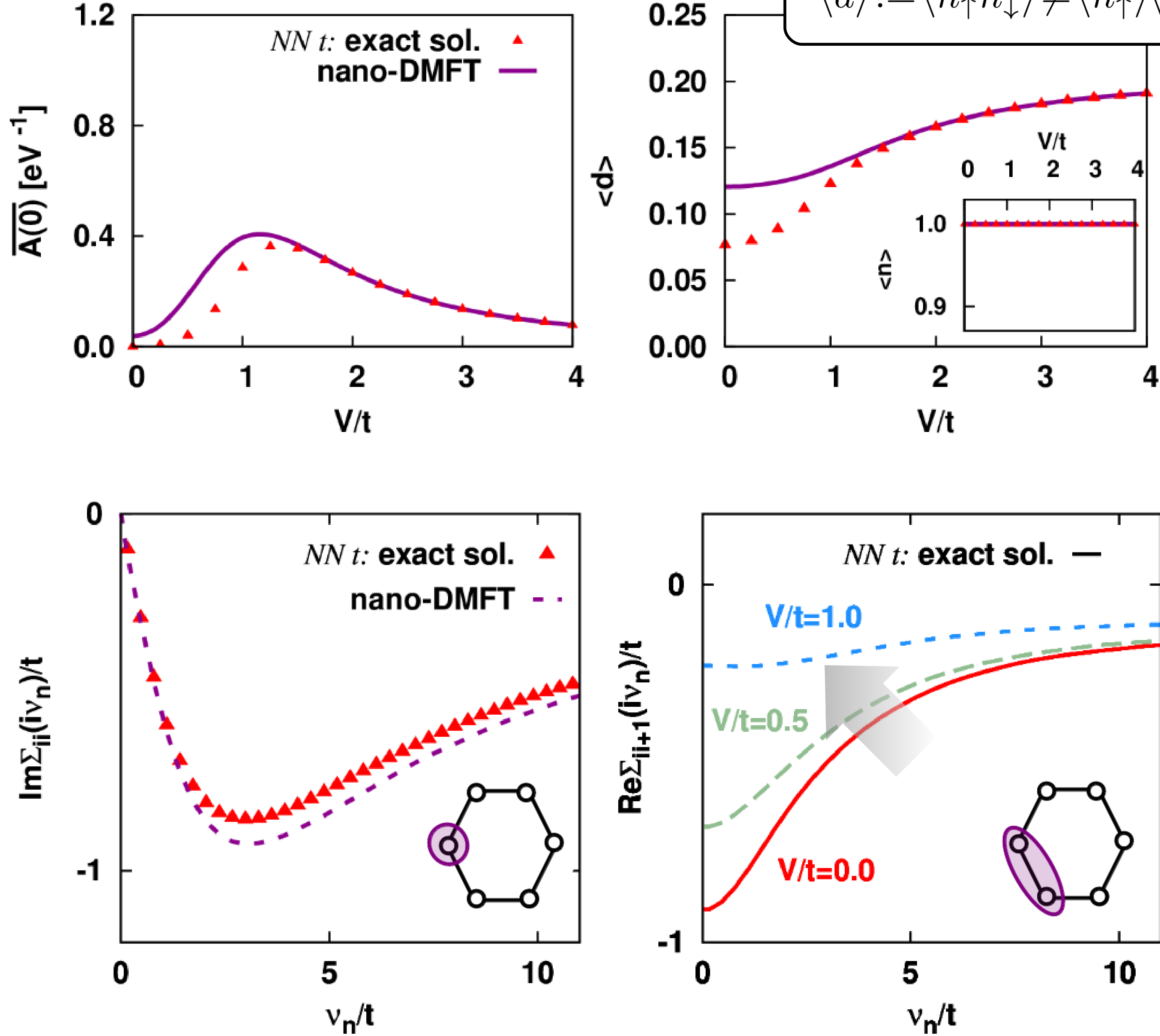

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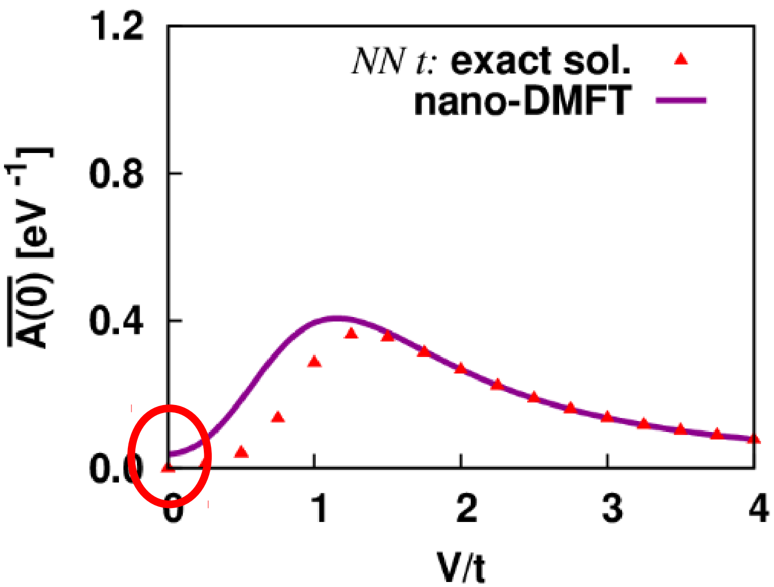

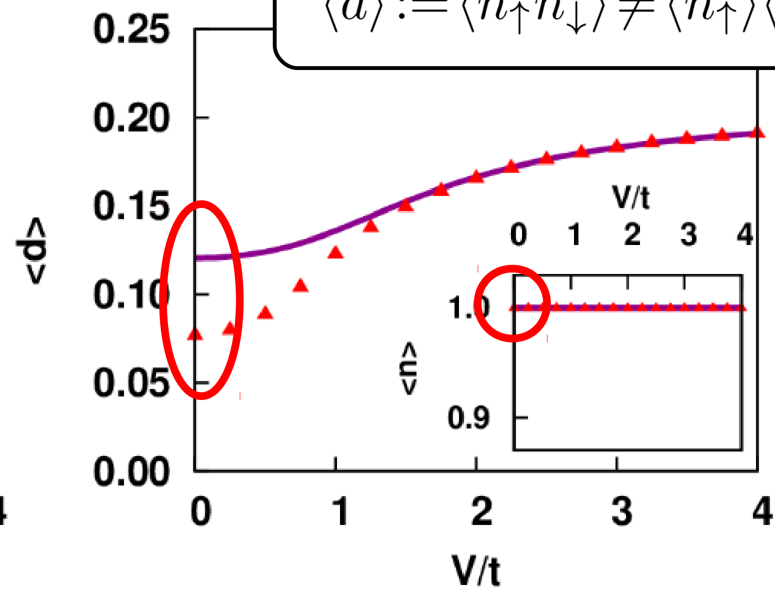

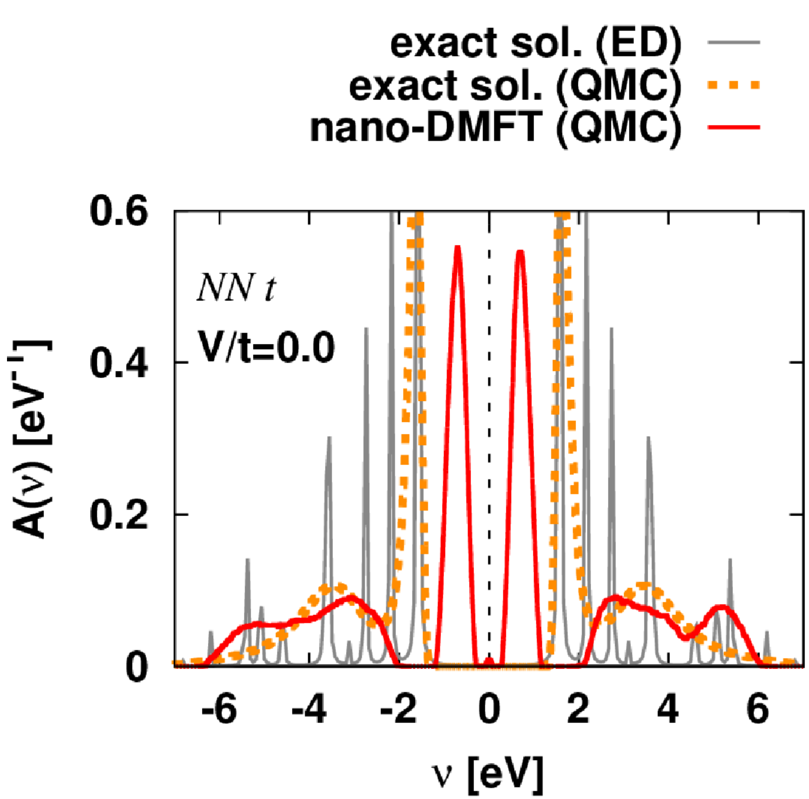

nano-DMFT @ work

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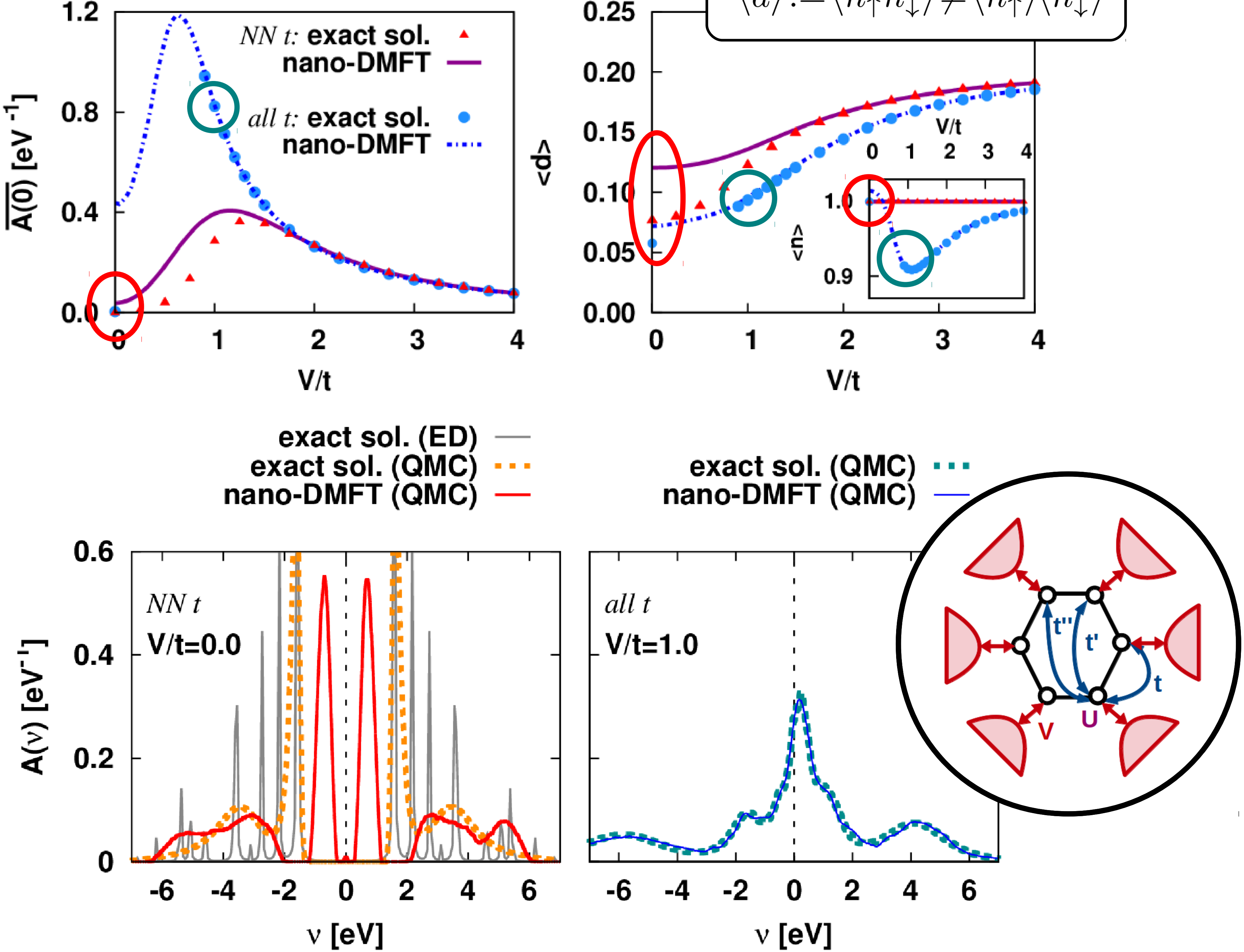
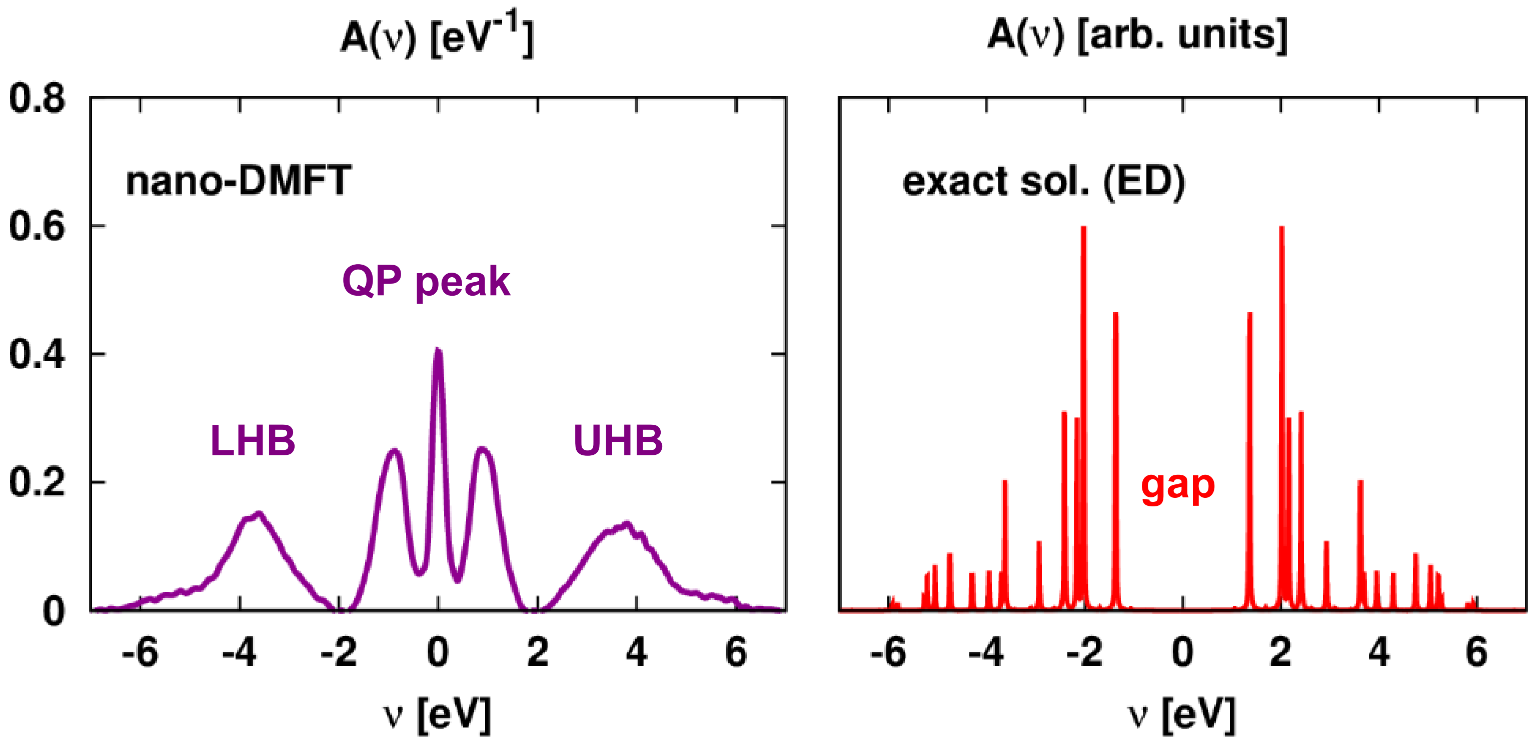


nano-DMFT @ work



parameters:

U/D=2.5

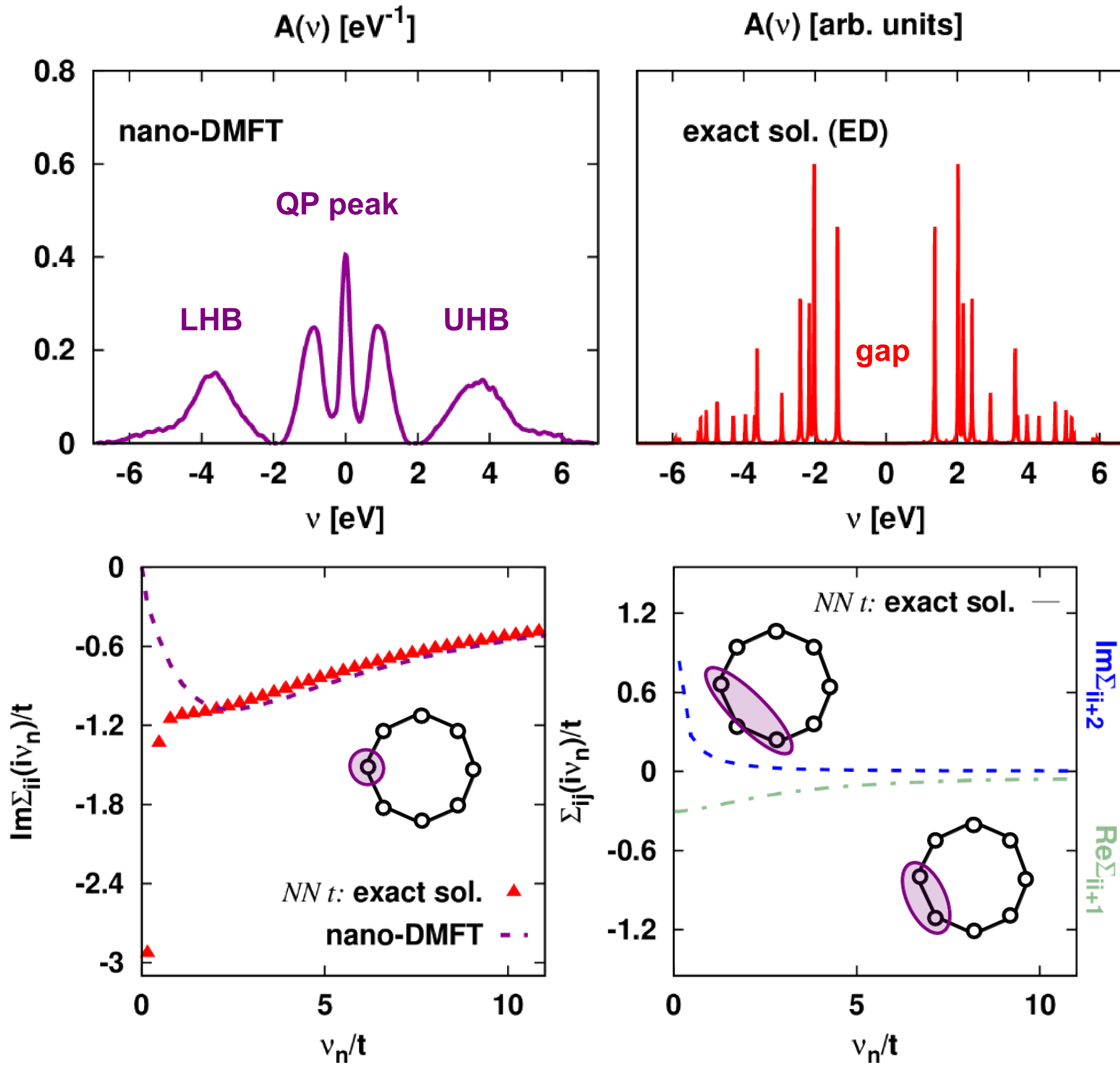
T/D=0.025

V/D=0 (non-local correlations most important)

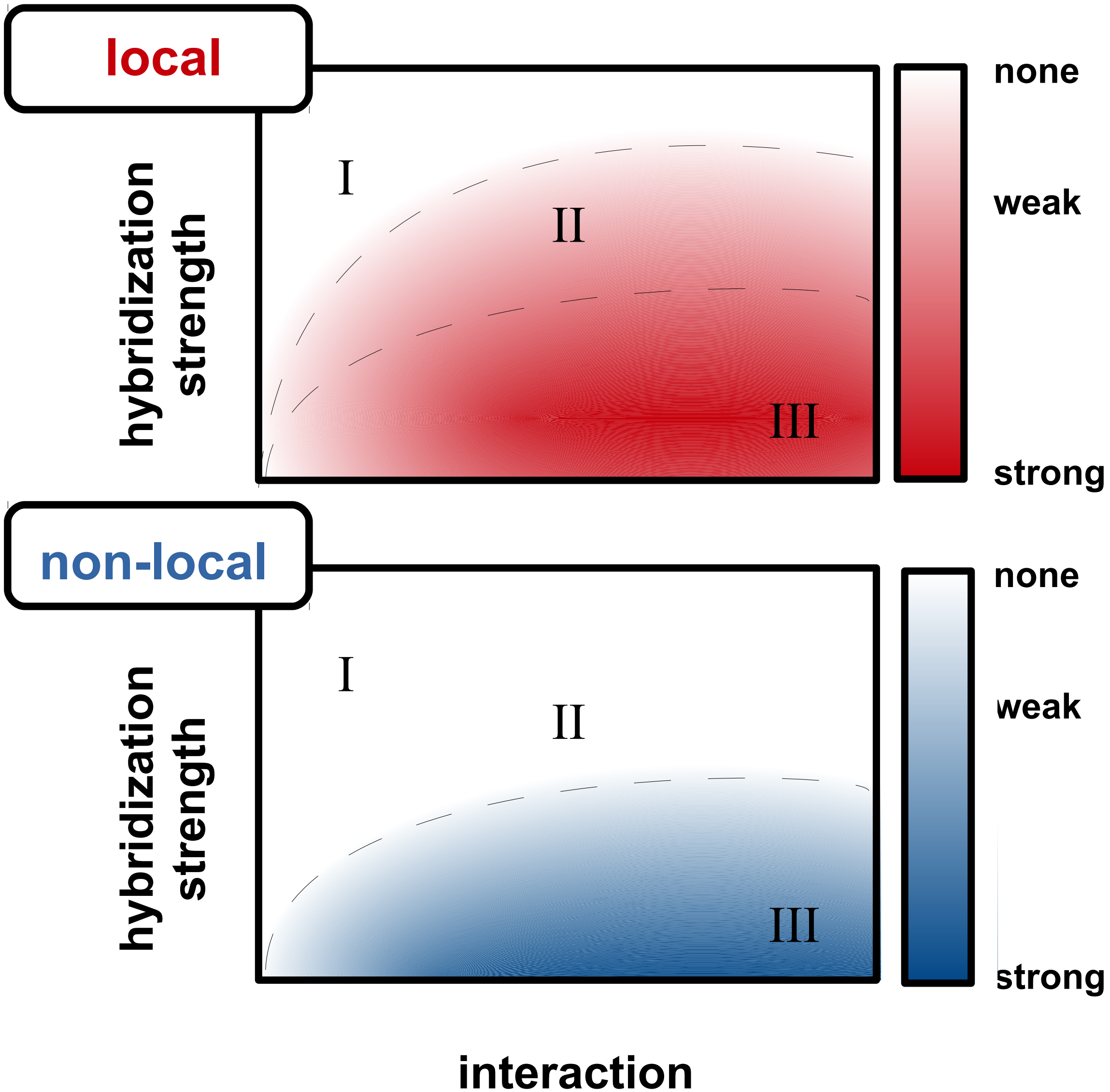
impurity solver:

Hirsch-Fye QMC (both nano-DMFT & exact sol.)

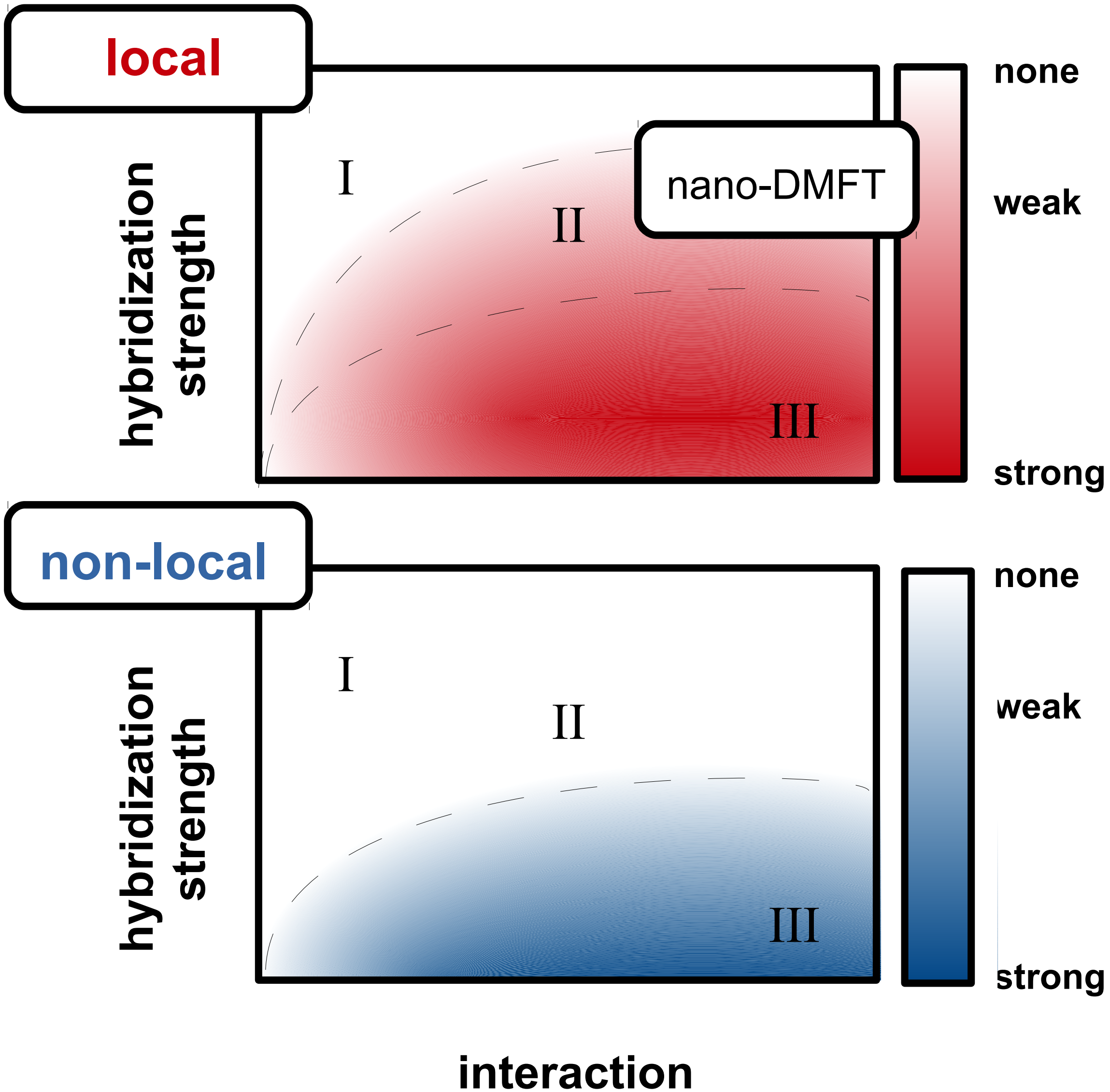
nano-DMFT @ work

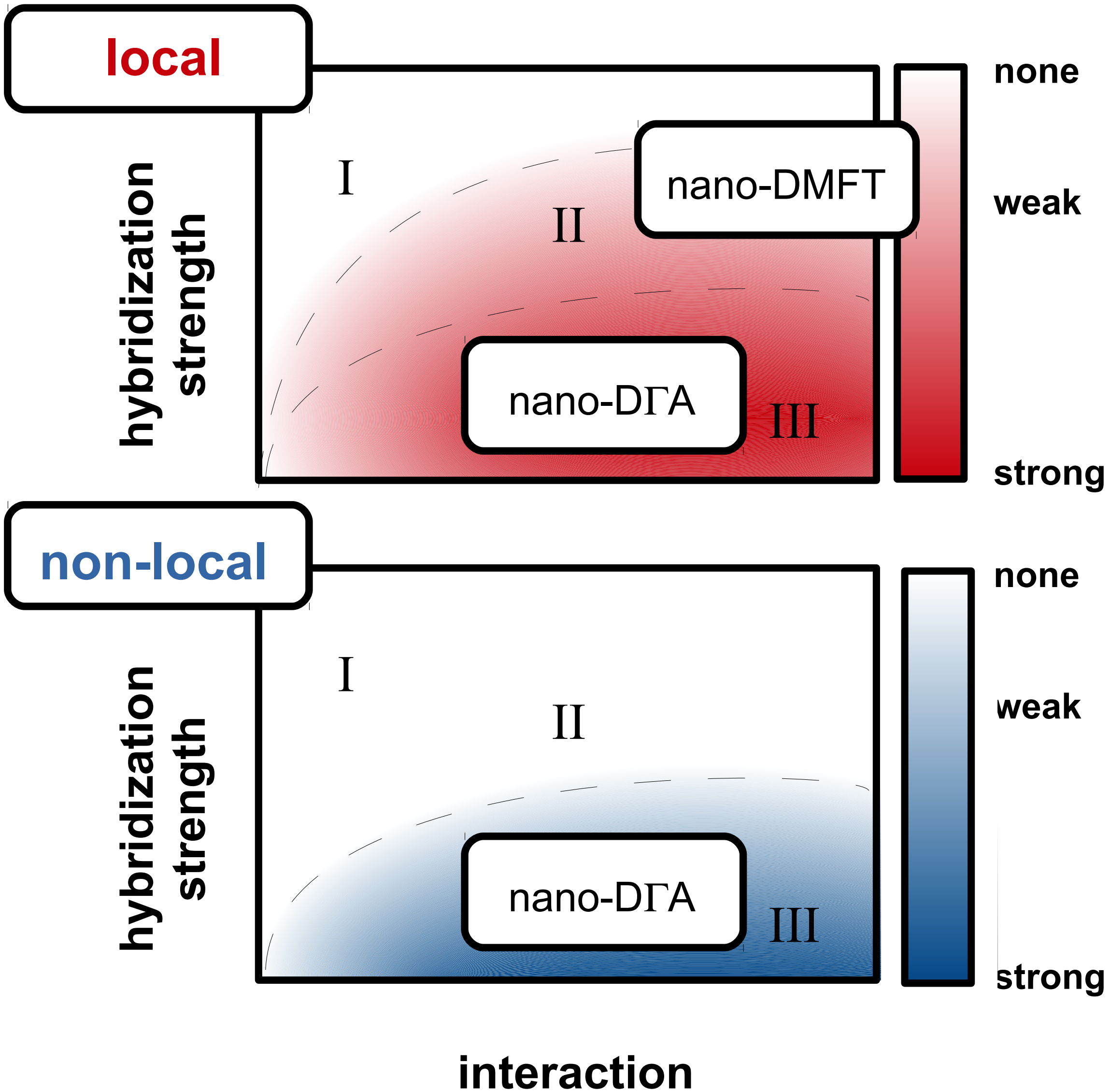


correlations "phase diagram"

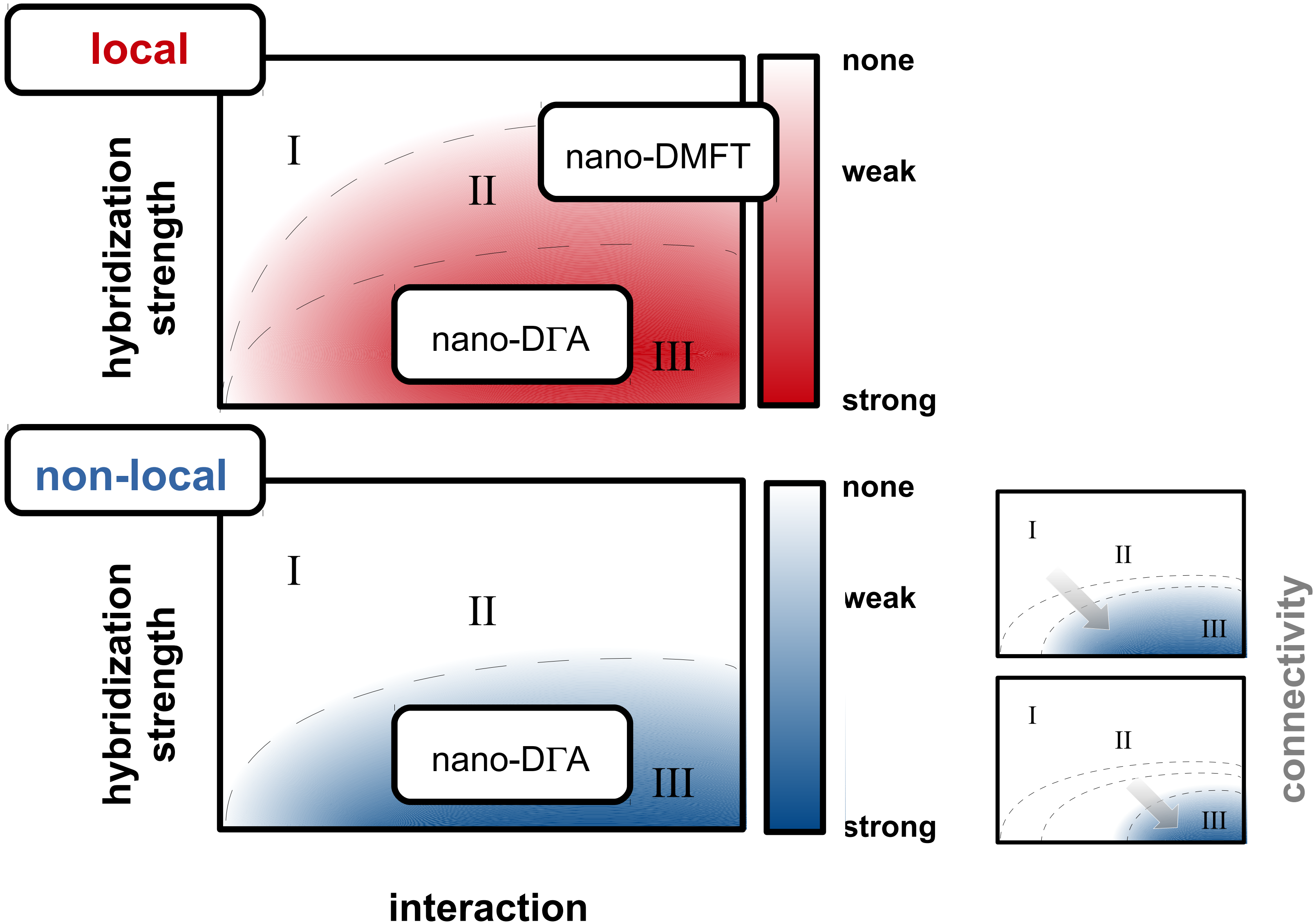


correlations "phase diagram"



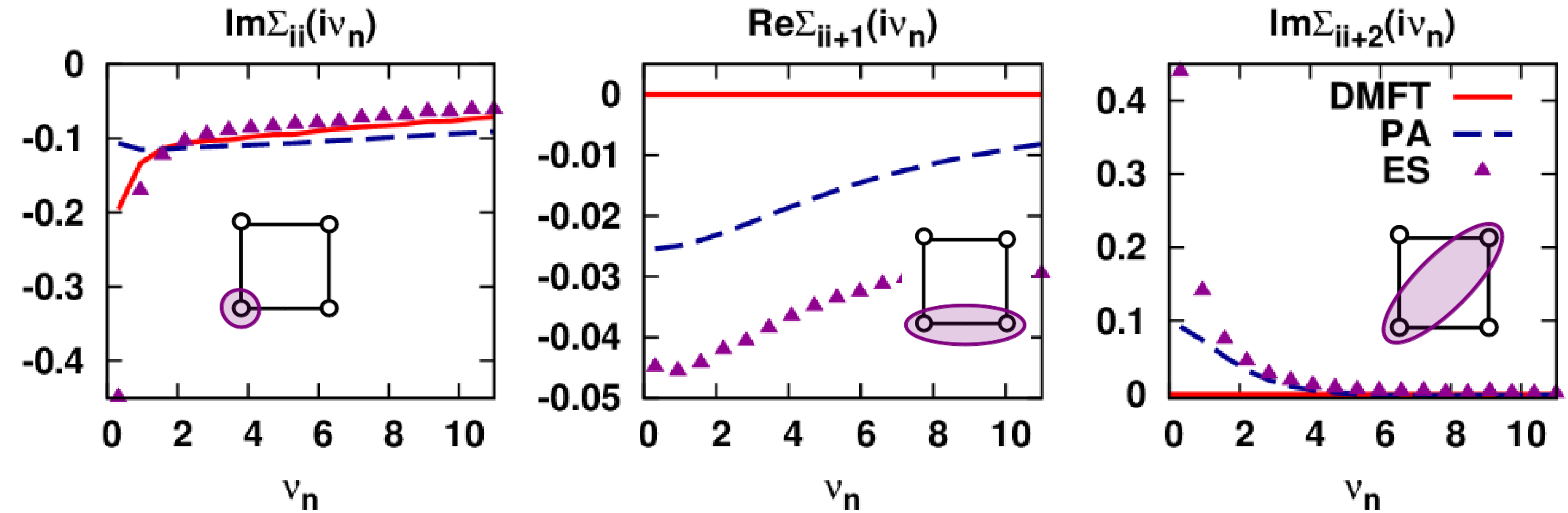

local
I
II
III
nano-DMFT
nano-DΓA
hybridization strength
none
weak
strong
non-local
I
II
III
nano-DΓA
hybridization strength
none
weak
strong
interaction

correlations “phase diagram”



U/D=1.0 T/D=0.05

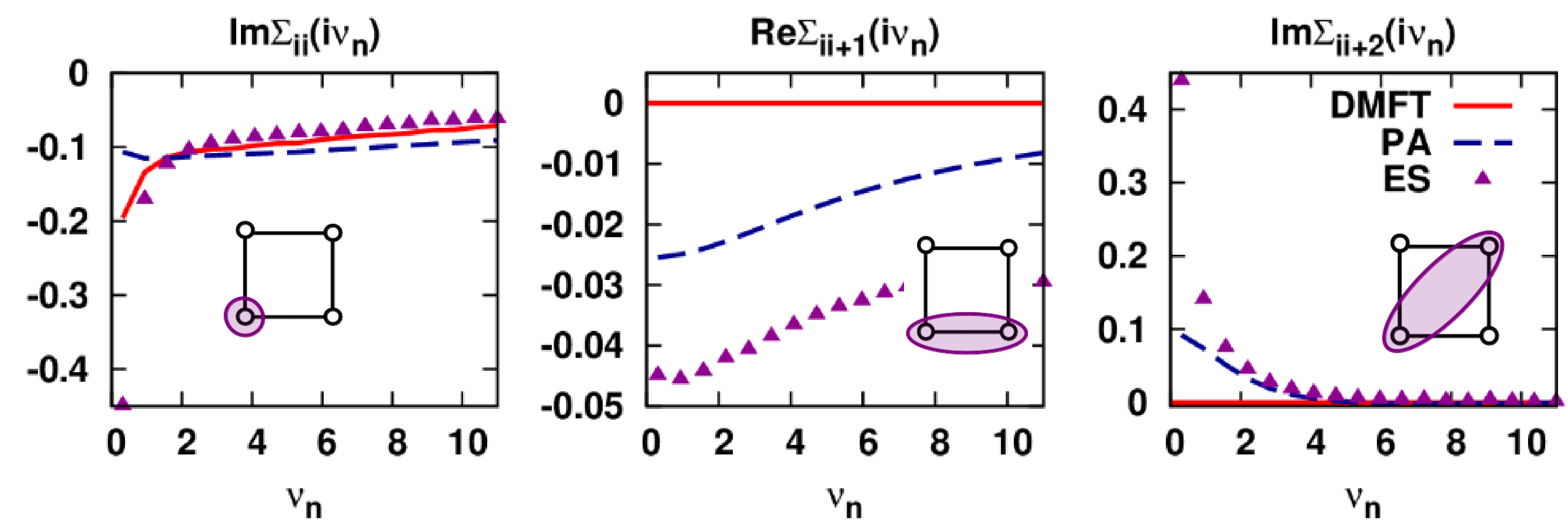
parquet approximation (PA): Λ=U



Nc=4

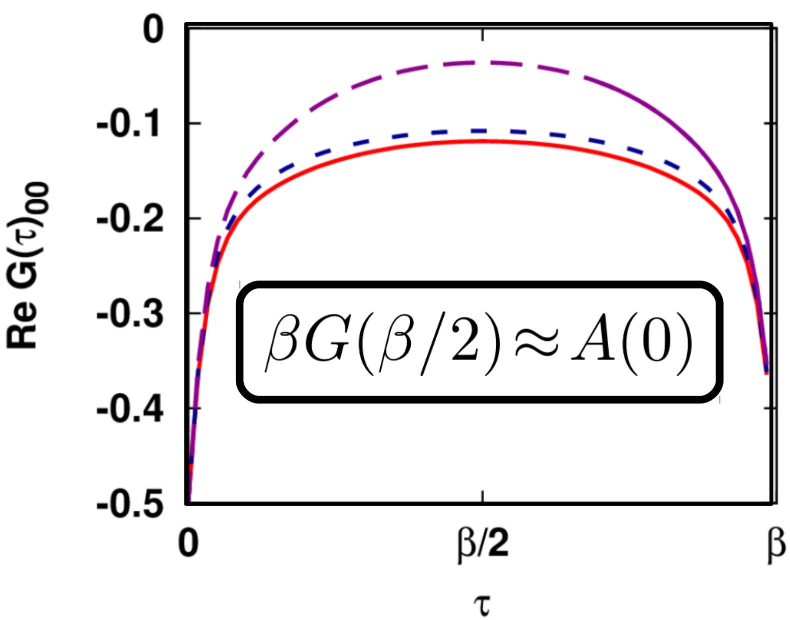
parquet approximation (PA): Λ=U

U/D=1.0 T/D=0.05



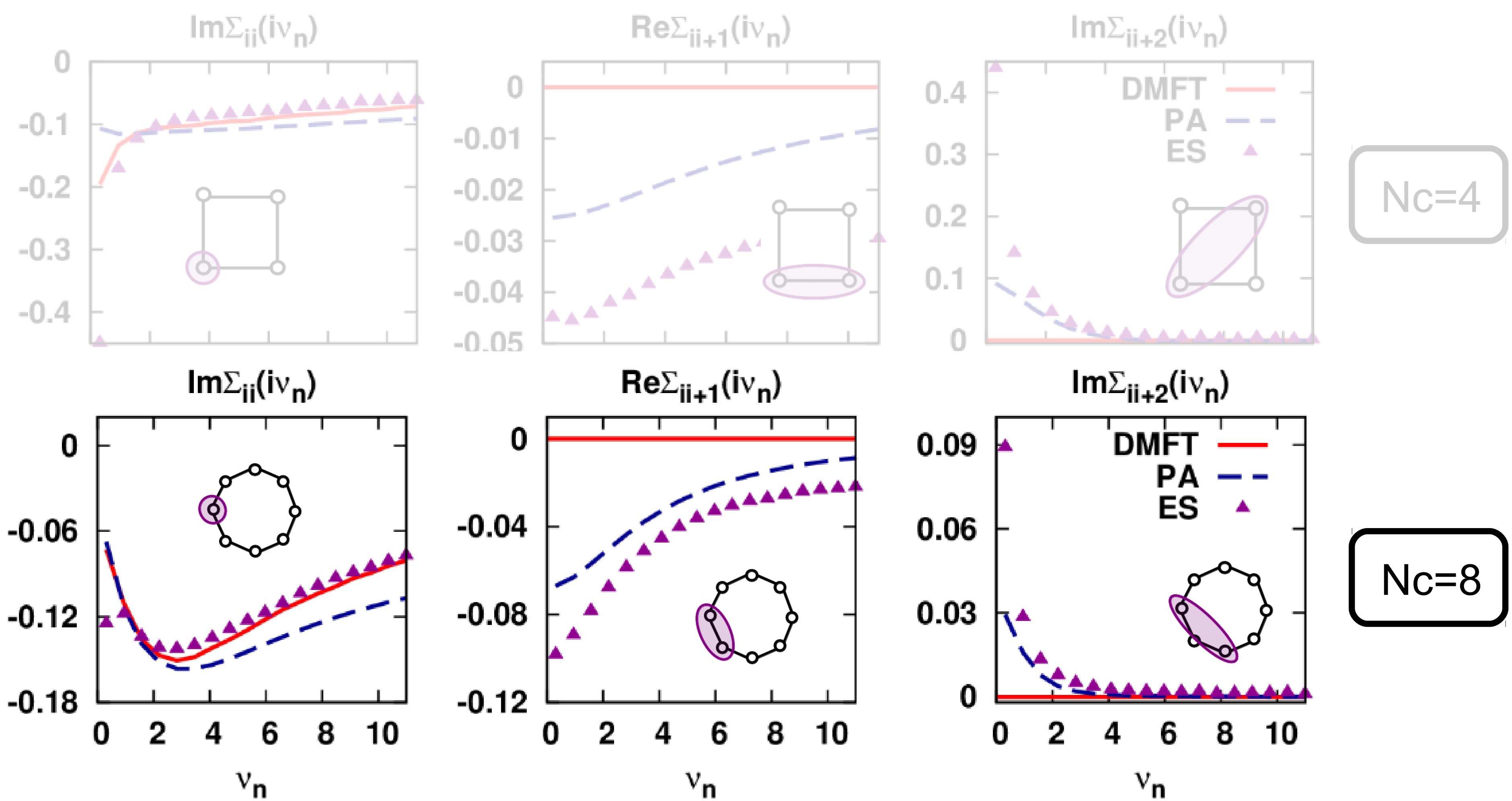
Nc=4

nano-DMFT capture (to some extent) insulating behavior?



U/D=1.0 T/D=0.05

parquet approximation (PA): Λ=U



intro

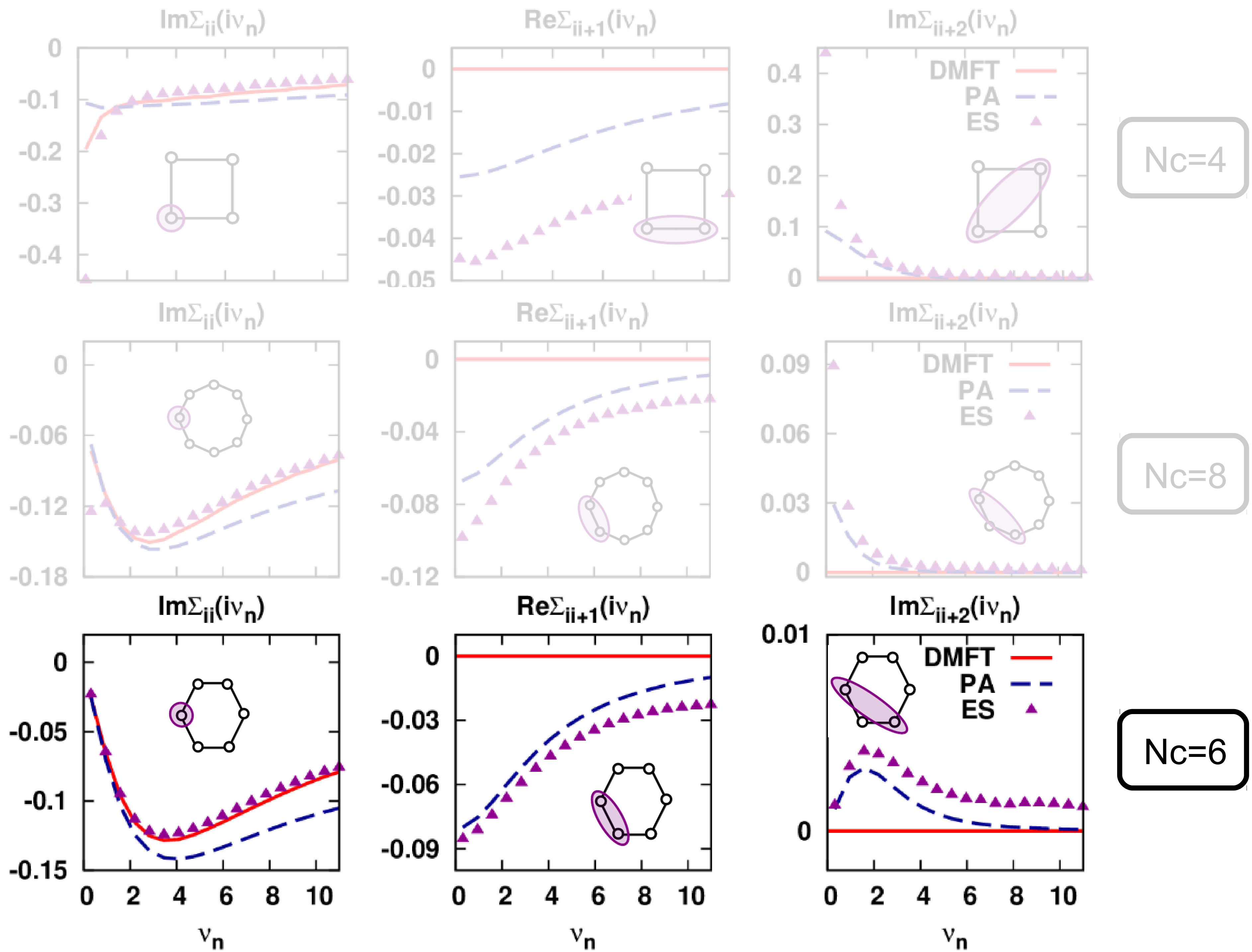
methods

applications

outlook

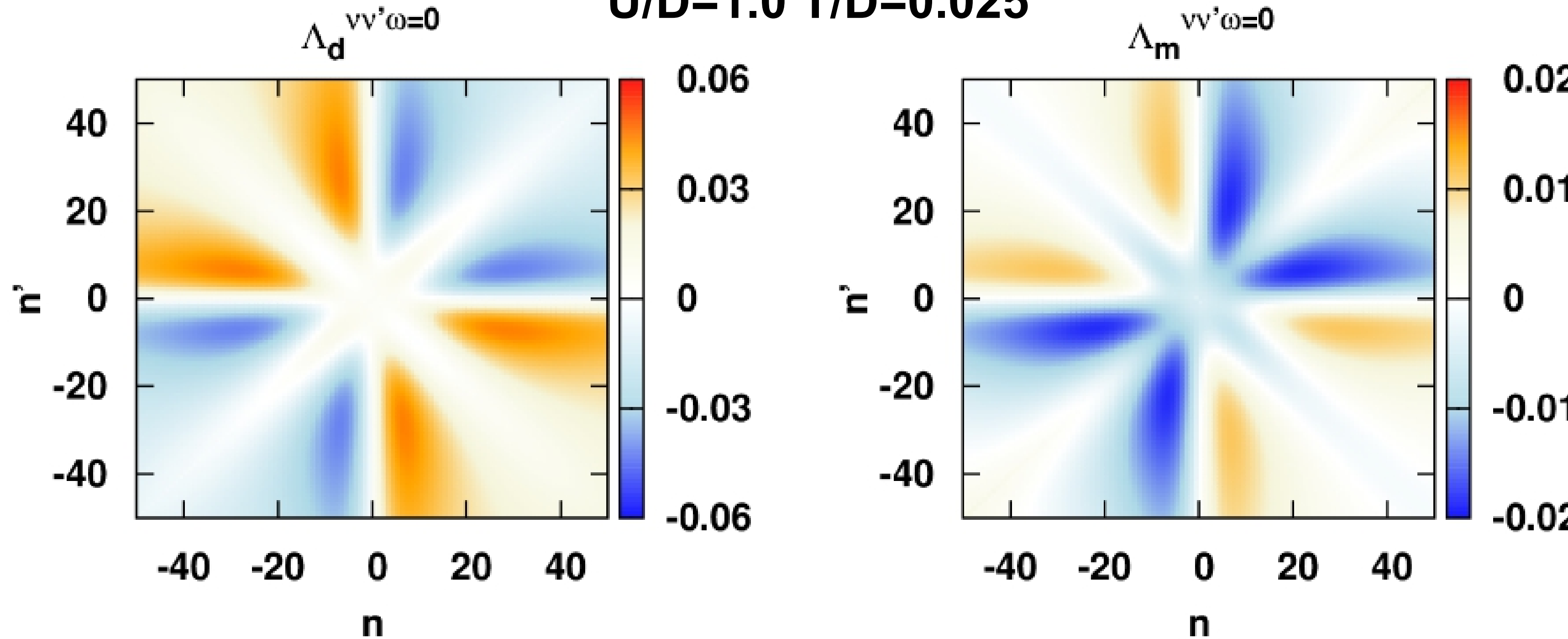
parquet approximation (PA): Λ=U

U/D=1.0 T/D=0.05



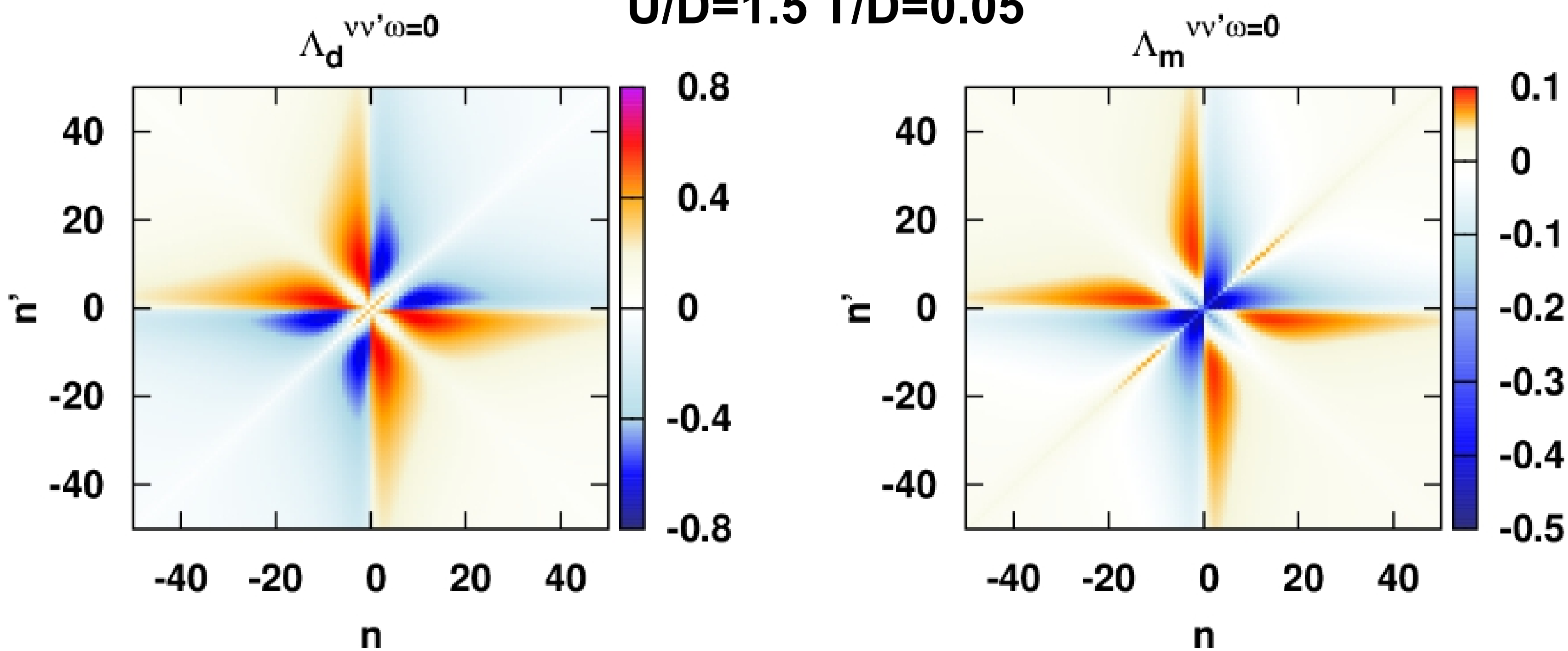
fully irreducible vertex

U/D=1.0 T/D=0.025



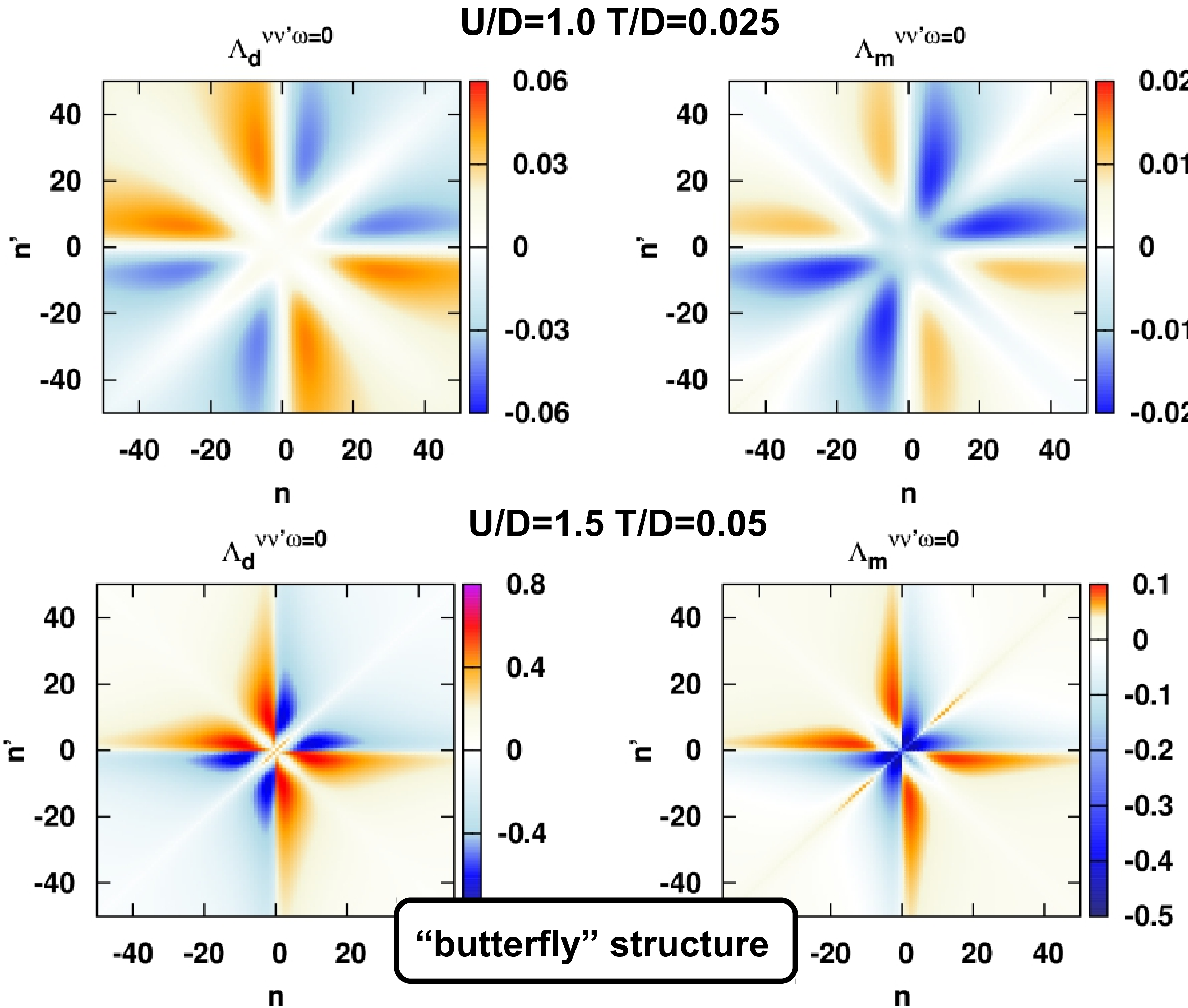
Nc=6

U/D=1.5 T/D=0.05



Nc=8

fully irreducible vertex

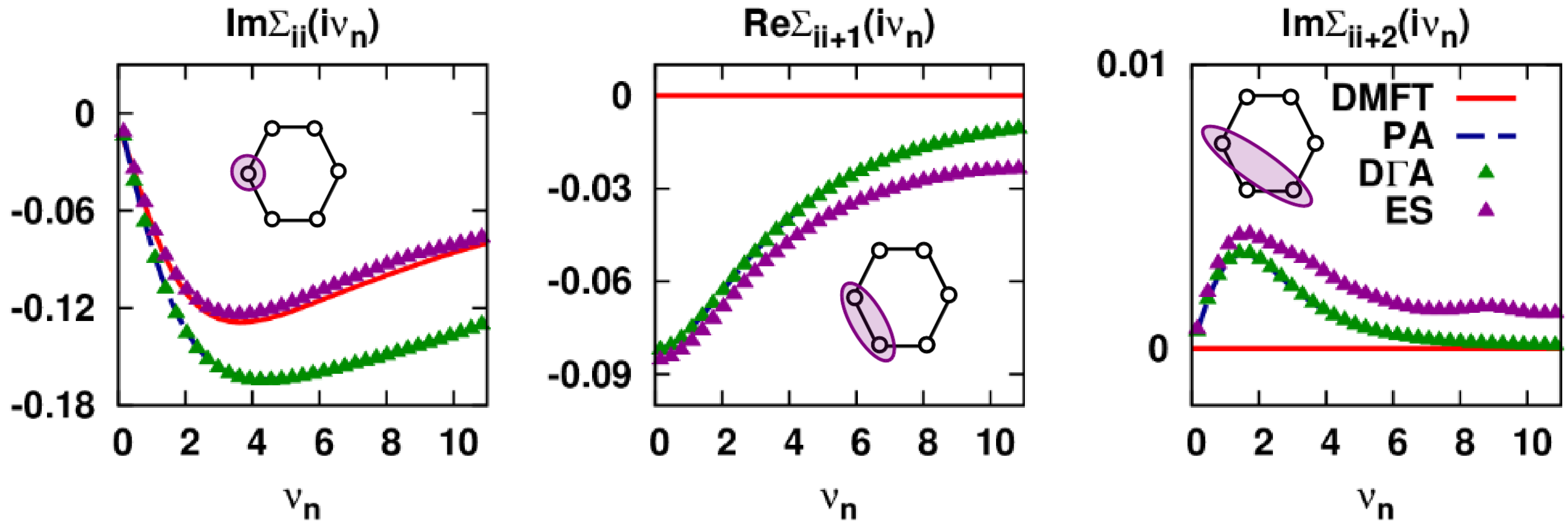


Nc=6

Nc=8

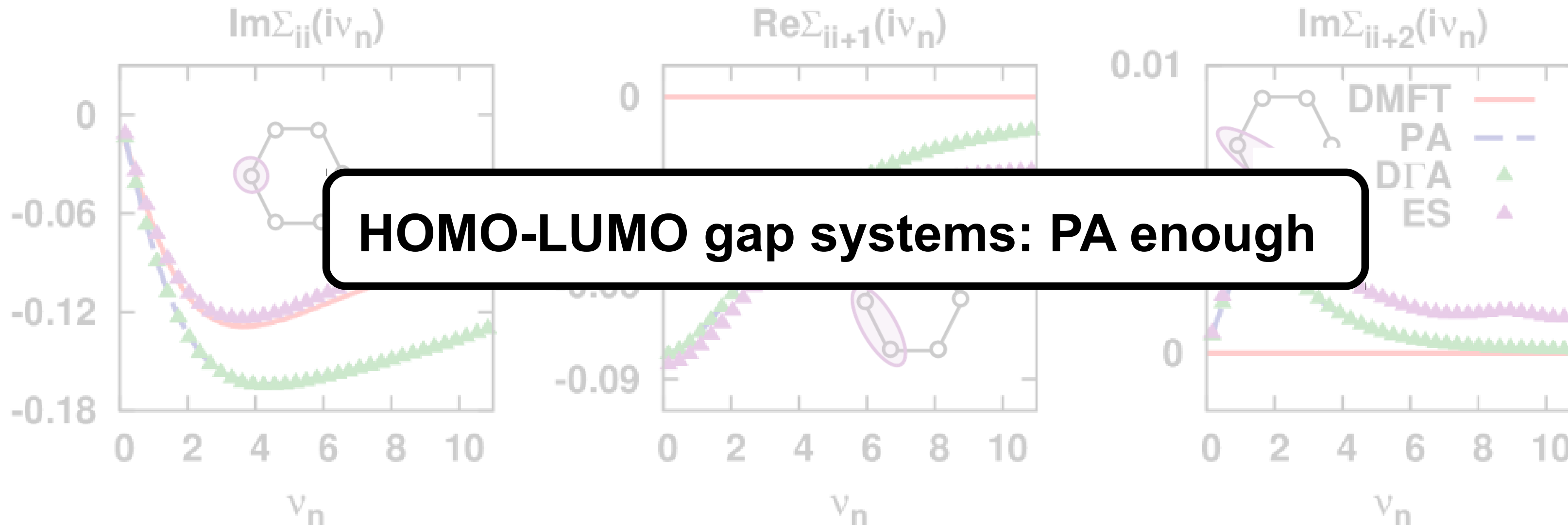
“butterfly” structure

U/D=1.0 T/D=0.025



Nc=6

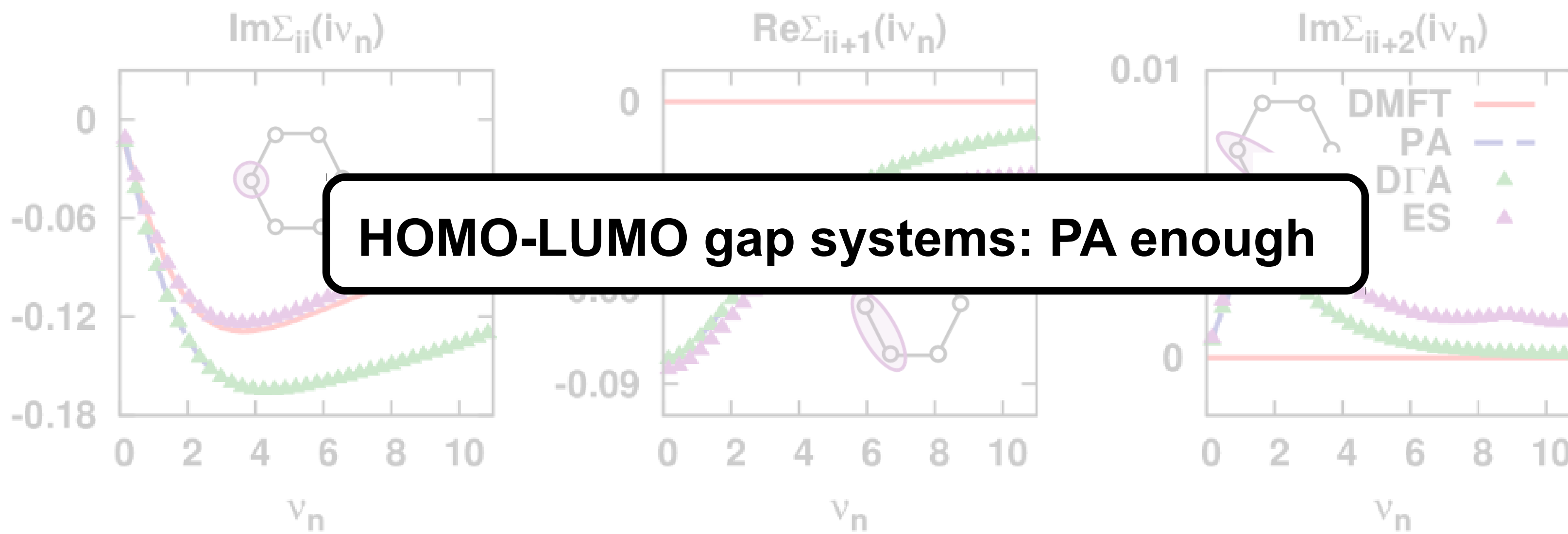
U/D=1.0 T/D=0.025



HOMO-LUMO gap systems: PA enough

Nc=6

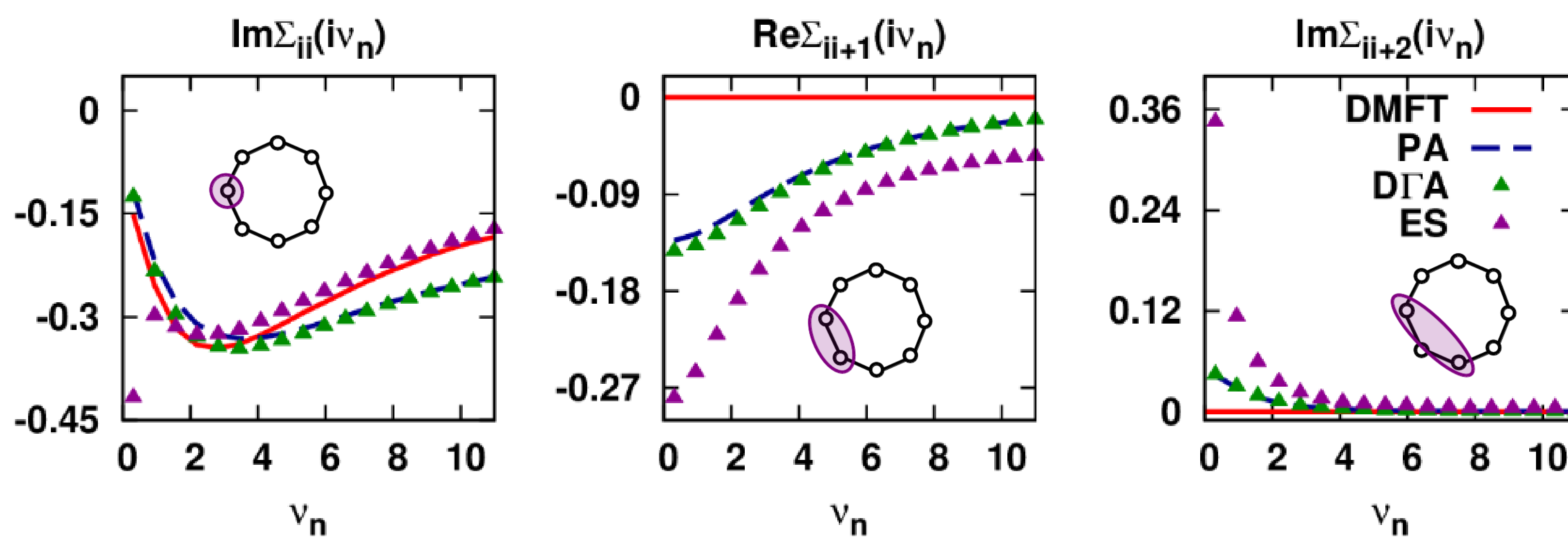
U/D=1.0 T/D=0.025



HOMO-LUMO gap systems: PA enough

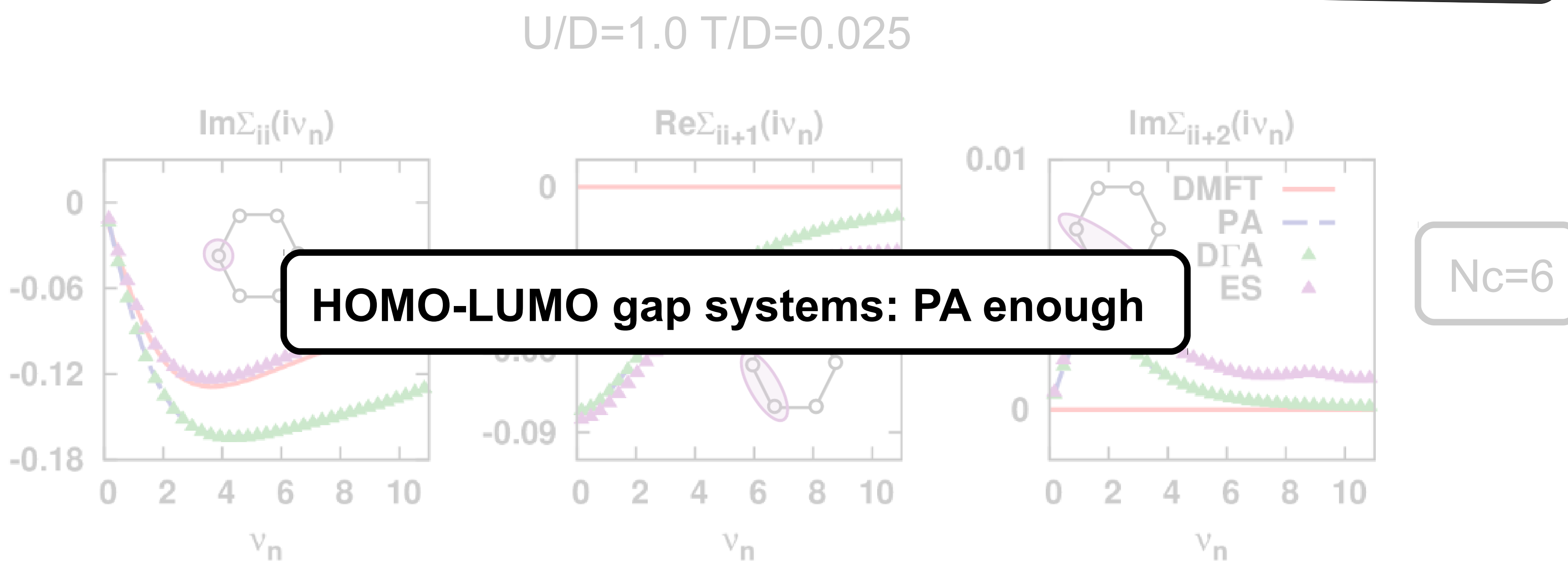
Nc=6

U/D=1.5 T/D=0.05

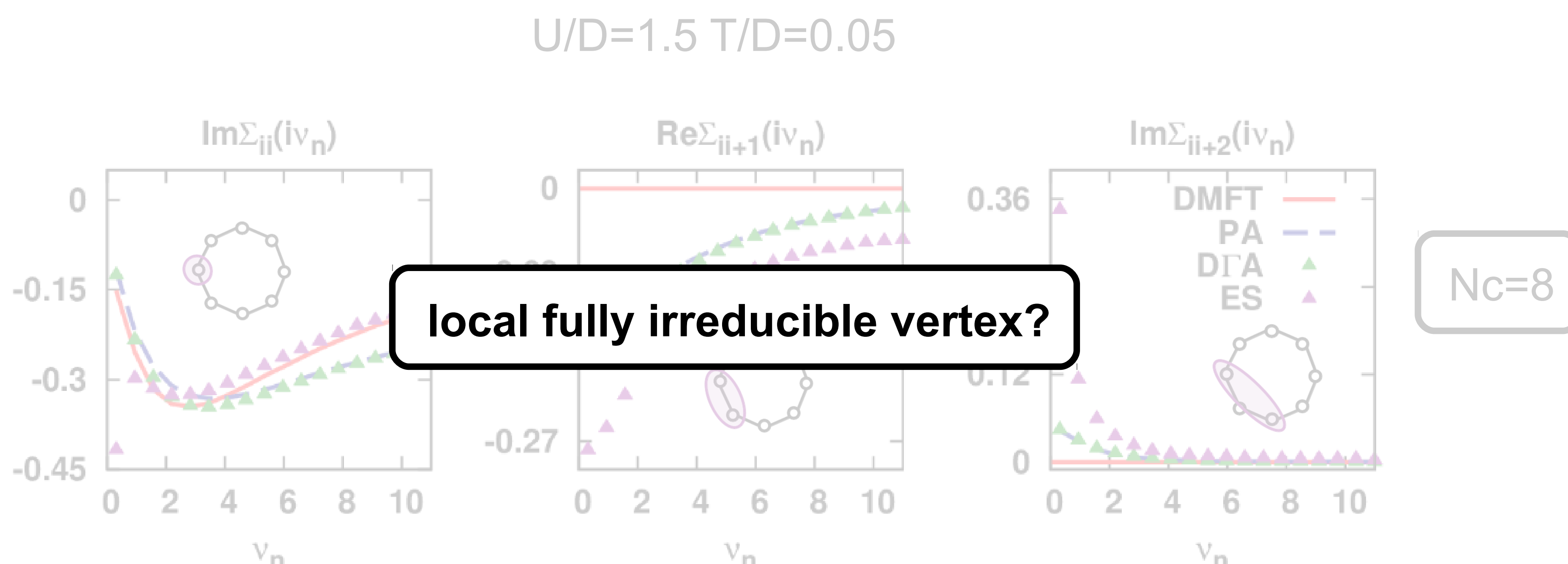


Nc=8

nano-DΓA @ work



HOMO-LUMO gap systems: PA enough



local fully irreducible vertex?

self-energy asymptotics

enforcing symmetries

- crossing symmetry ⇨ allows to solve parquet beyond weak coupling
- time-reversal symmetry
- point-group symmetries

Tam *et al.*, PRE **87** (2013)

vertex parametrization/asymptotics

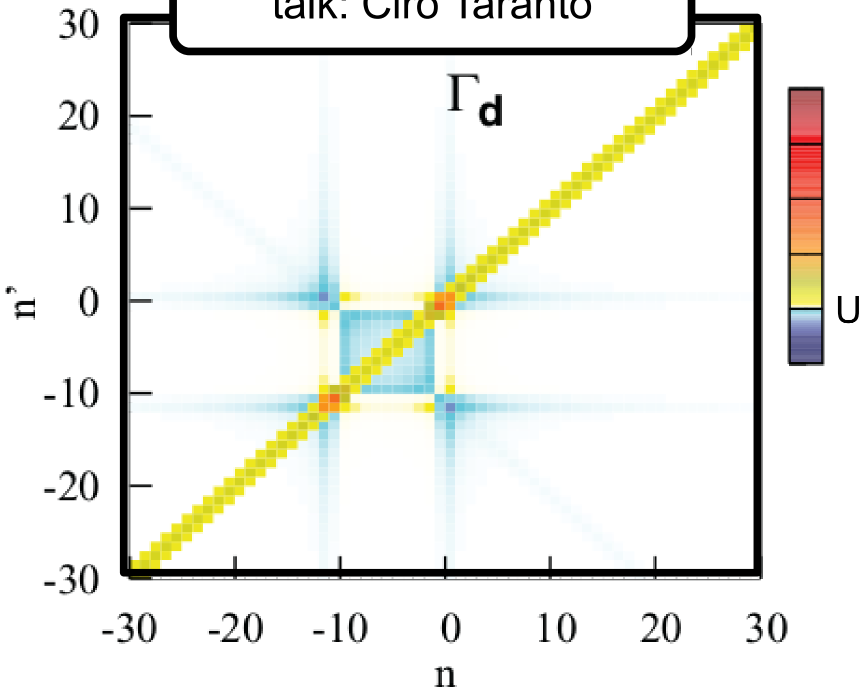
- numerical low-energy-features
- semi-analytical asymptotics

Karrasch et al., JPCM **20** (2008)

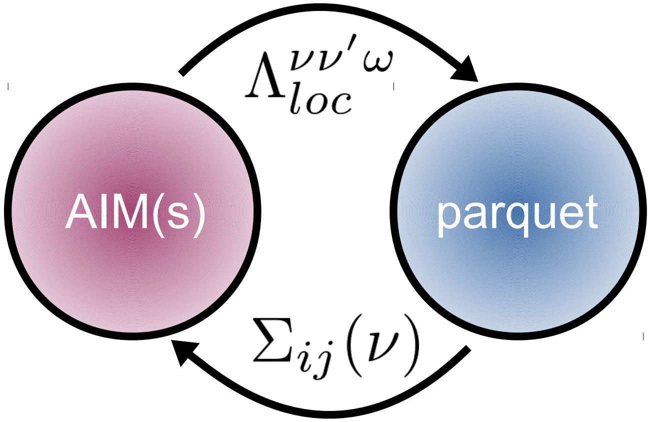
Kuneŝ PRB **83** (2011)

Rohringer, Valli, & Toschi PRB **86** (2012)

talk: Ciro Taranto



fully self-consistent nano-DΓA

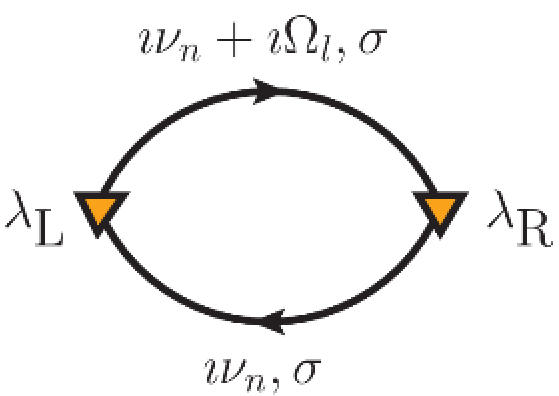


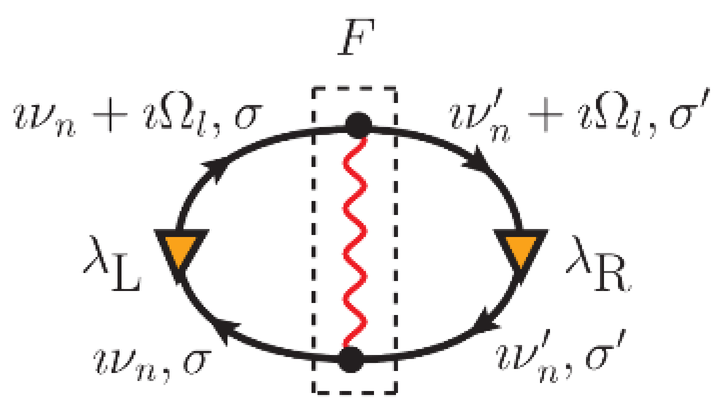
exact vertex function: k-structure

locality of fully irreducible vertex: holds in 1D or 0D systems?

vertex corrections to response functions

(e.g., electronic & thermal transport)





collaborators

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Giorgio Sangiovanni

Olle Gunnarsson

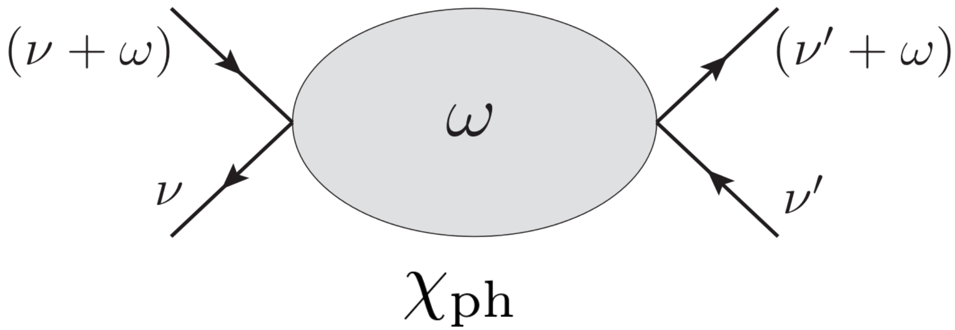
parquet solver developing

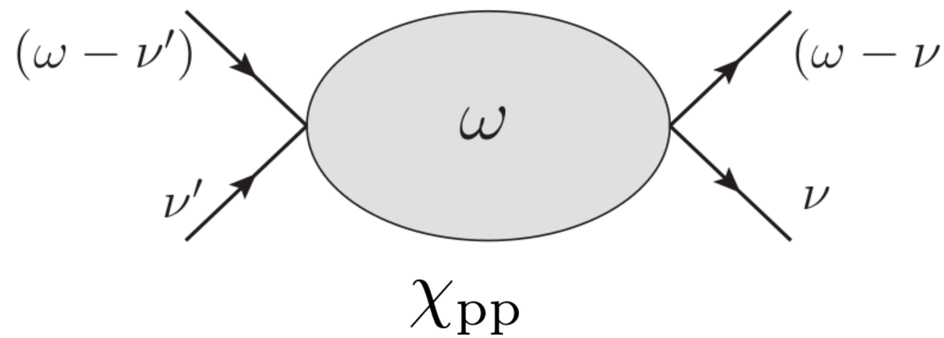
www.phys.lsu.edu/~syang/parquet

Thank you for your attention!

see additional slides below

particle-hole (ph) & particle-particle (pp) channels





SU(2) + crossing symmetry: re-definining channels

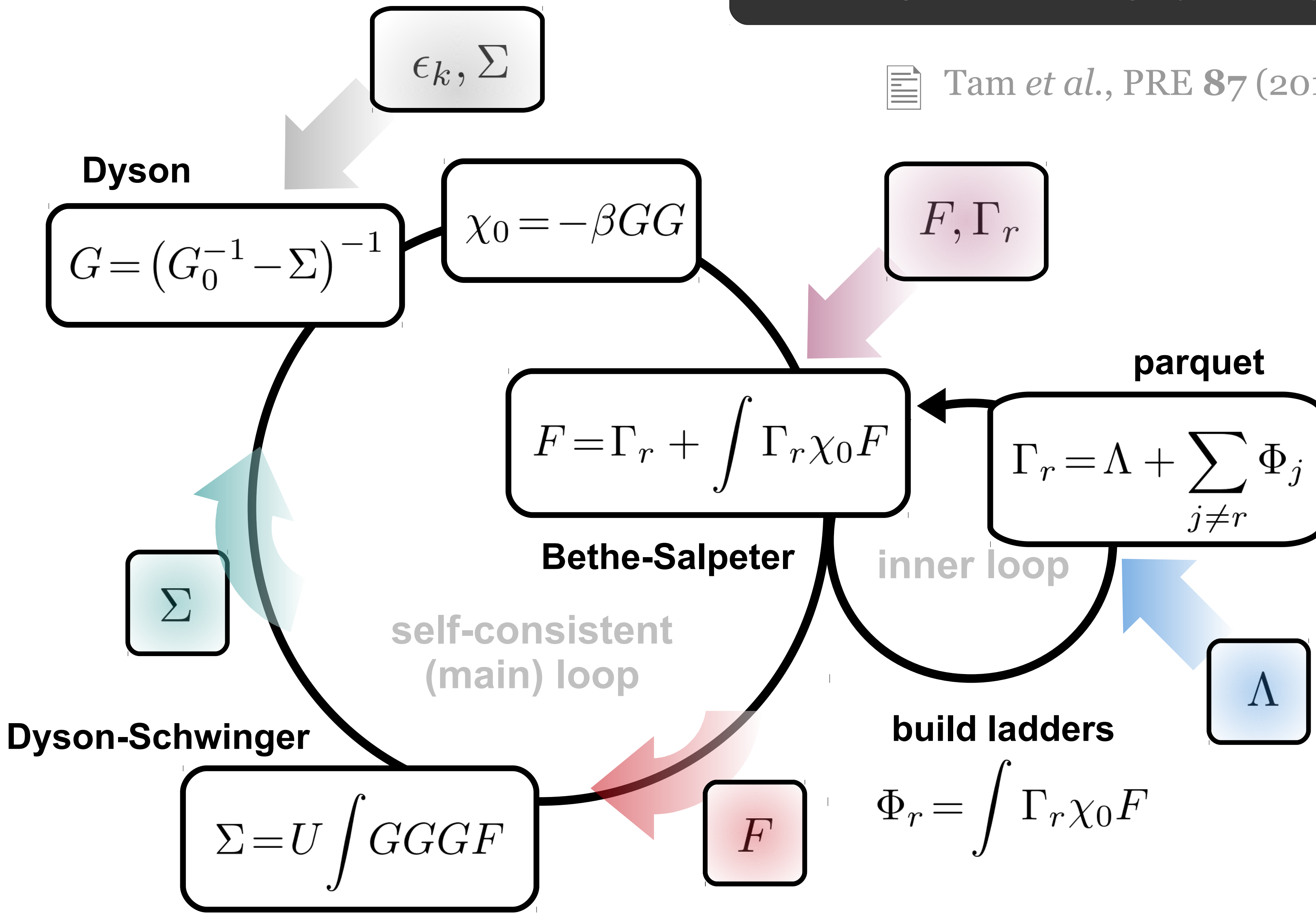
$$(\uparrow\uparrow, \uparrow\downarrow, \overline{\uparrow\downarrow}) \times (ph, \overline{ph}, pp) \rightarrow (\text{d,m,s,t})$$

$$\Gamma_d^{\nu\nu'\omega} = \Gamma_{\text{ph},\uparrow\uparrow}^{\nu\nu'\omega} + \Gamma_{\text{ph},\uparrow\downarrow}^{\nu\nu'\omega}$$
$$\Gamma_m^{\nu\nu'\omega} = \Gamma_{\text{ph},\uparrow\uparrow}^{\nu\nu'\omega} - \Gamma_{\text{ph},\uparrow\downarrow}^{\nu\nu'\omega}$$
$$\Gamma_s^{\nu\nu'\omega} = -\Gamma_{\text{pp},\uparrow\uparrow}^{\nu\nu'\omega} + 2\Gamma_{\text{pp},\uparrow\downarrow}^{\nu\nu'\omega}$$
$$\Gamma_t^{\nu\nu'\omega} = \Gamma_{\text{pp},\uparrow\uparrow}^{\nu\nu'\omega}$$

Rohringer, Valli, & Toschi PRB **86** (2012)

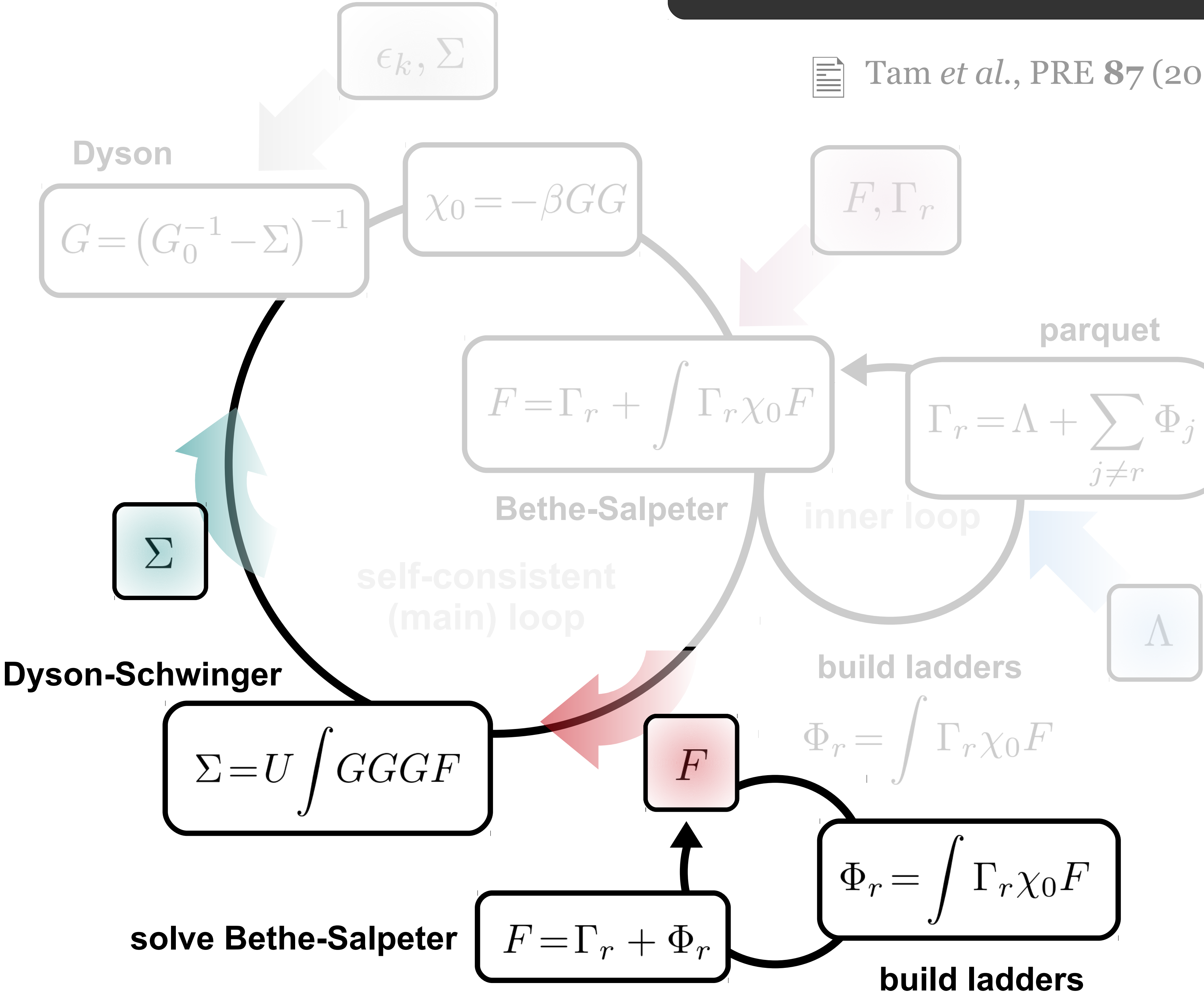
restoring the crossing symmetry

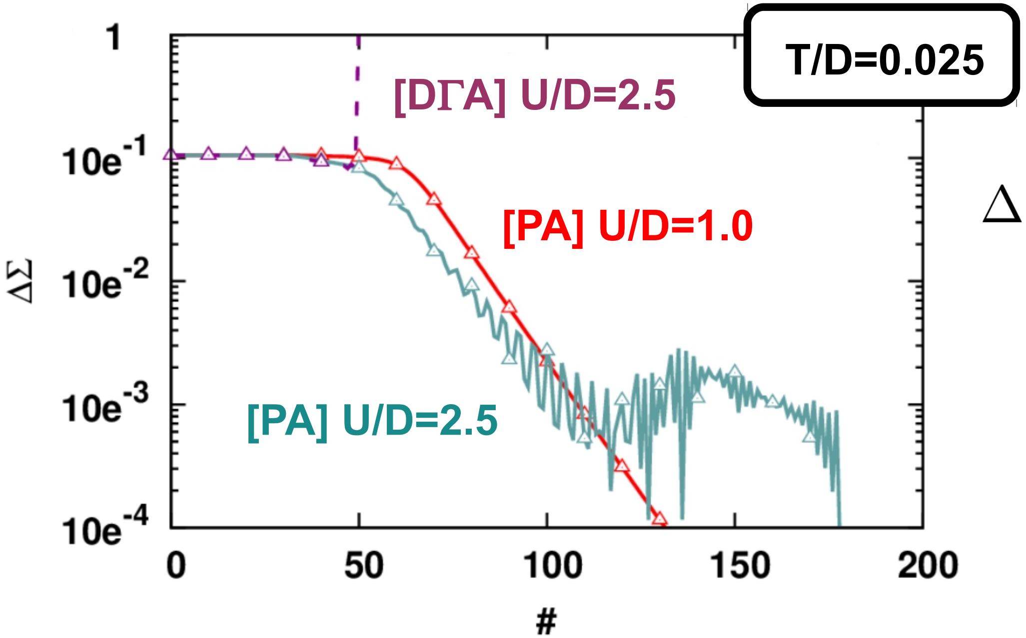
Tam *et al.*, PRE **87** (2013)



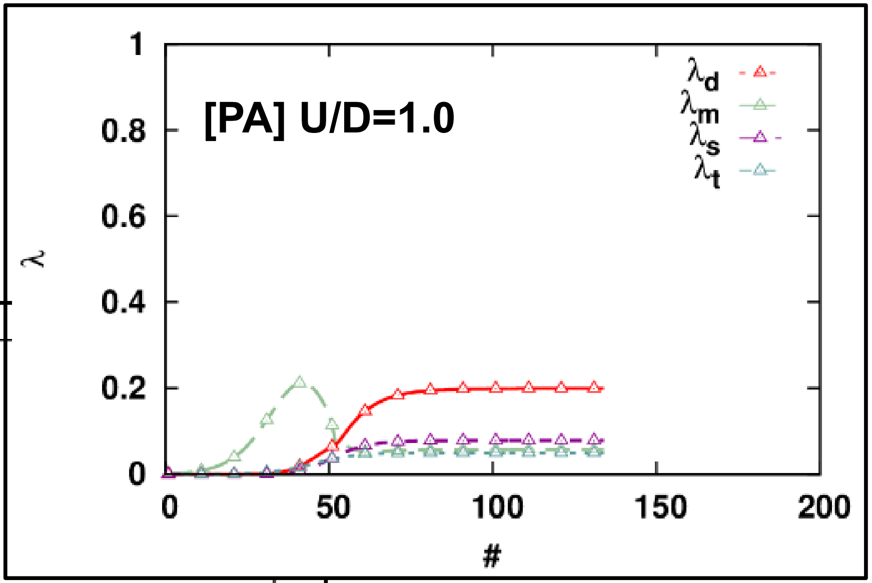
restoring the crossing symmetry

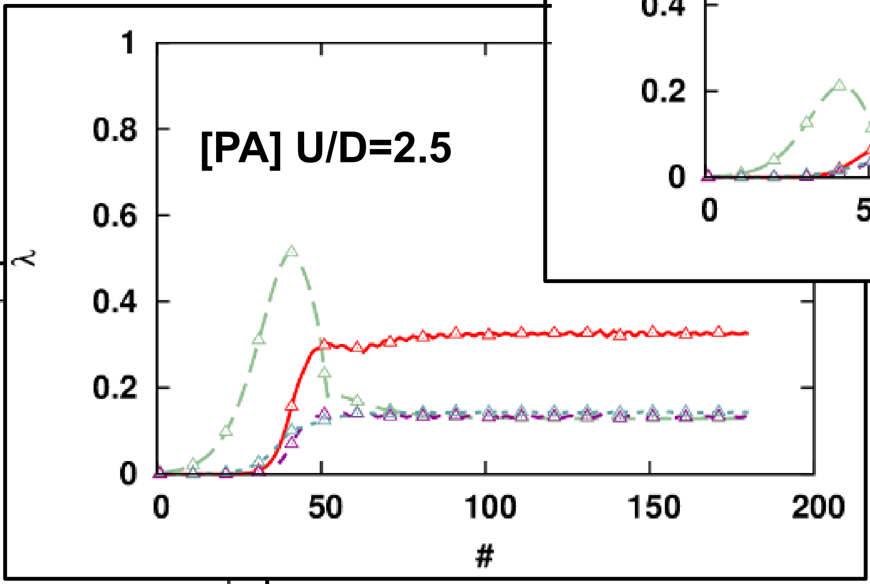
Tam *et al.*, PRE **87** (2013)

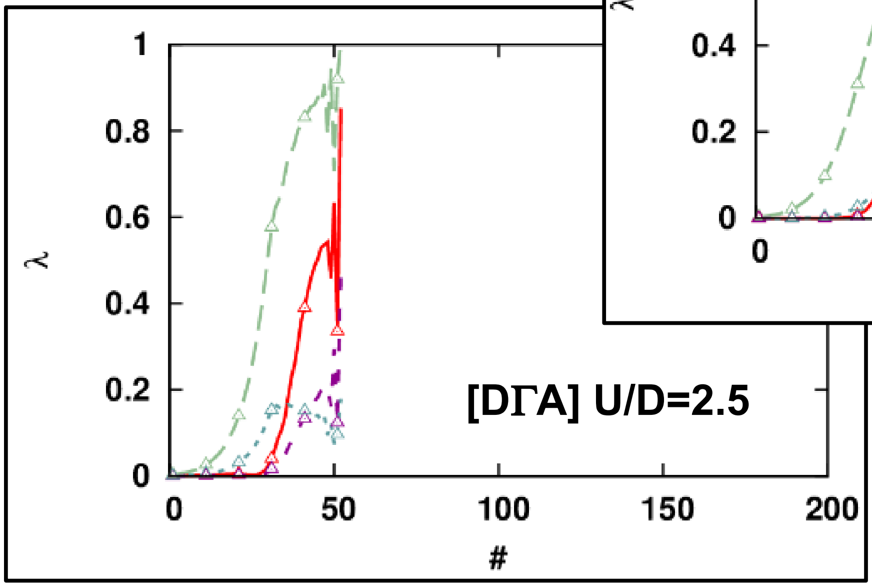




$$\Delta\Sigma = \frac{\sum_{kn} \Sigma_k^i(\imath\nu_n) - \Sigma_k^{i-1}(\imath\nu_n)}{\sum_{kn} \Sigma_k^i(\imath\nu_n) + \Sigma_k^{i-1}(\imath\nu_n)}$$





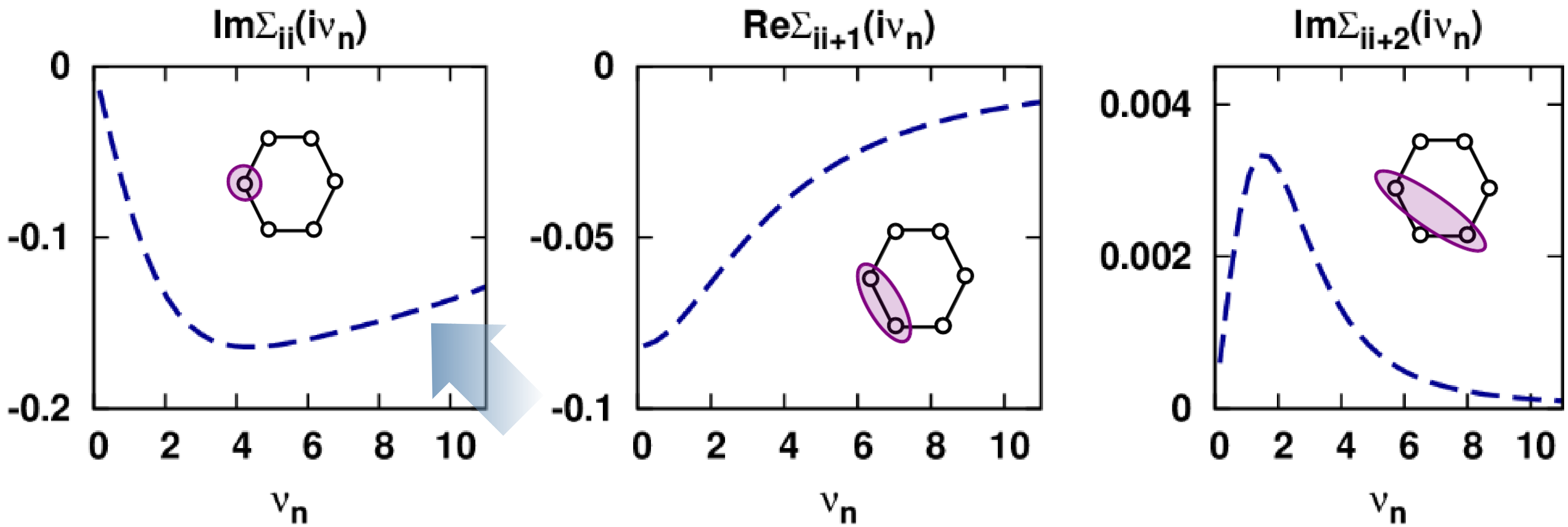


leading eigenvalue

$$\max_{\lambda_r} \mid \lambda_r \Phi_r = \alpha_r \Gamma_r G G \Phi_r$$

$\Sigma(\nu)$: asymptotic behavior in parquet

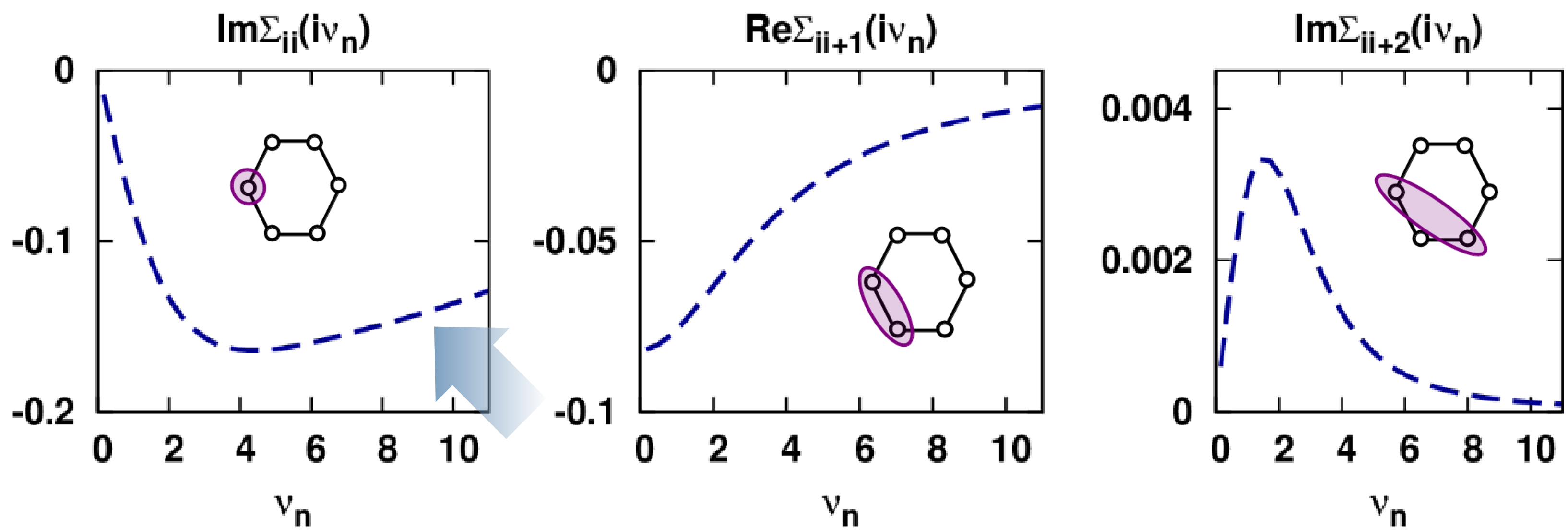
U/D=1.0   T/D=0.025



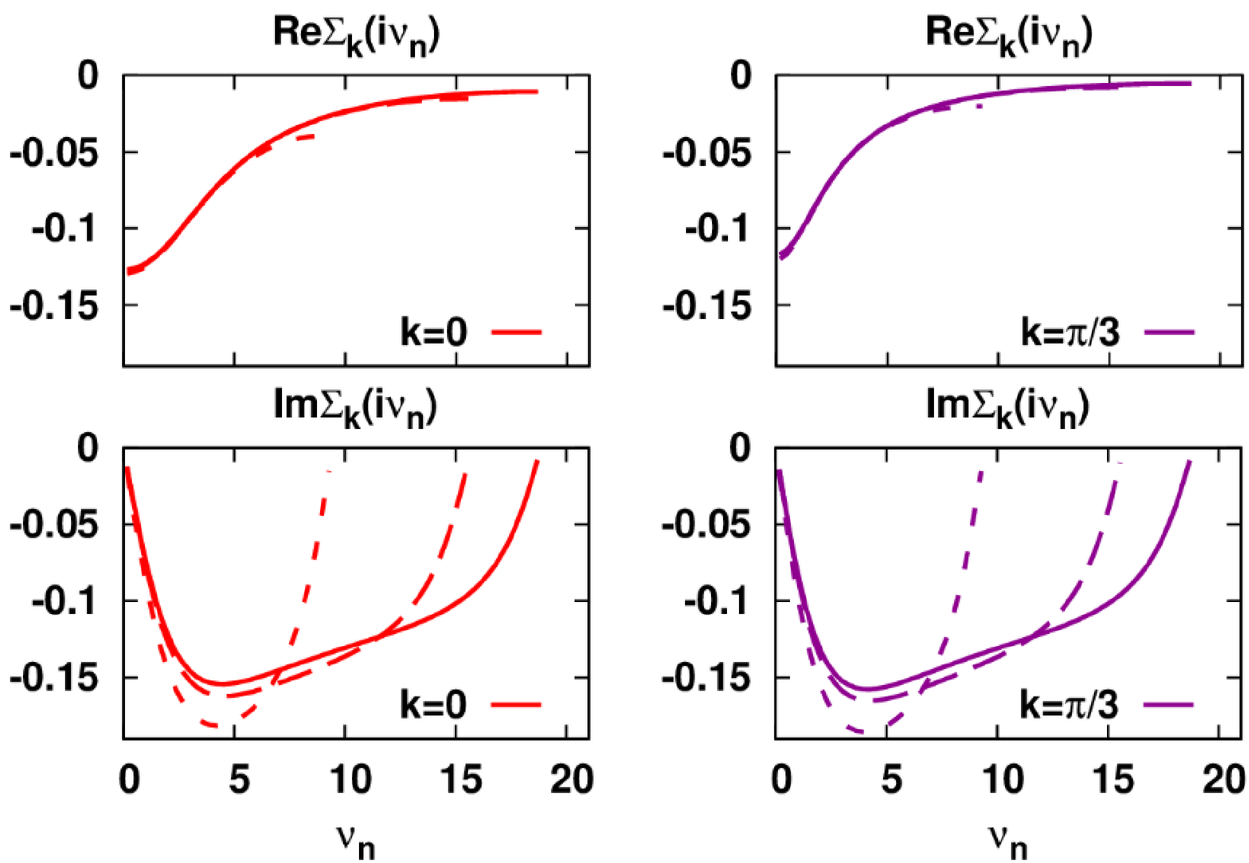
nf=100

Σ(ν): asymptotic behavior in parquet

U/D=1.0   T/D=0.025



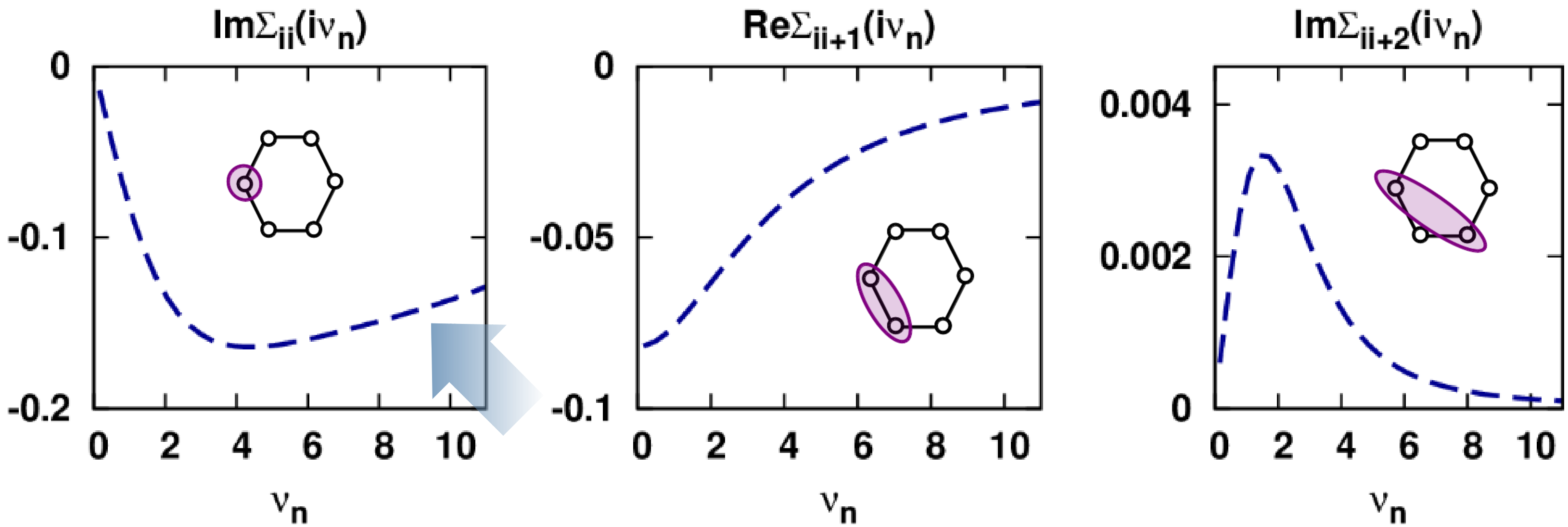
nf=100



nf=60,100,120

$\Sigma(\nu)$: asymptotic behavior in parquet

U/D=1.0   T/D=0.025



nf=100

$1/\imath\nu_n$ expansion of the self-energy

$$\Sigma_{ij\sigma}(\imath\nu_n) = \sum_{m=1}^{\infty} \left(\frac{1}{\imath\nu_n}\right)^m c_{ij\sigma}^{(m)}$$

lowest order coefficients

$$c_{ij\sigma}^{(1)} = U\langle n_{i\bar{\sigma}}\rangle\delta_{ij}$$

$$c_{ij\sigma}^{(2)} = U^2\langle n_{i\bar{\sigma}})\rangle(1-\langle n_{i\bar{\sigma}})\delta_{ij}$$

local! ⇒ $\Sigma_{i\neq j\sigma}(\imath\nu_n) \approx \left(\frac{1}{\imath\nu_n}\right)^3 c_{i\neq j\sigma}^{(3)}$