Electronic Structure – a view from finite-temperature field theory

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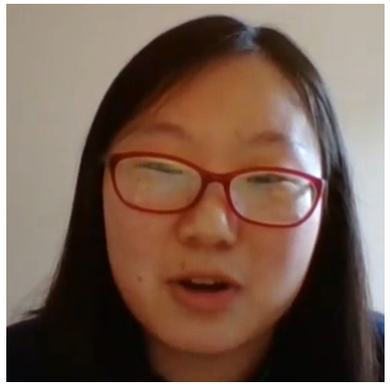
SIMONS FOUNDATION

United States – Israel Binational Science Foundation

Group Members



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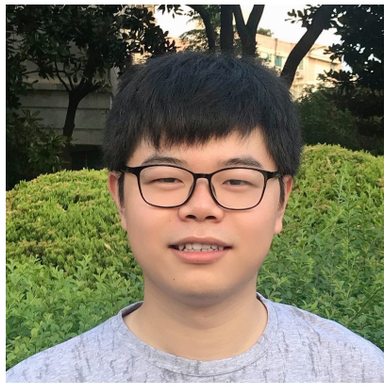
Lei Zhang



Yang Yu

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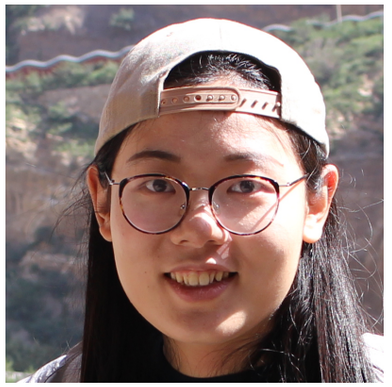

Runxue Yu

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Sergei Iskakov

Close collaborator: group of D. Zgid

Realistic Computation – What *can* we do?

Computational tools for electronic structure / condensed matter / quantum chemistry have been developed since the beginning of computers.

Density functional theory is the workhorse of electronic structure theory.

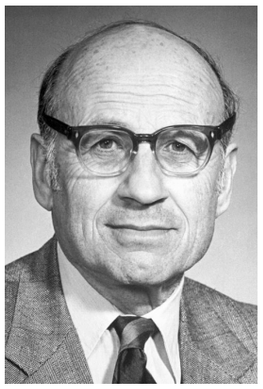
The following aspects typically work well / reliably:

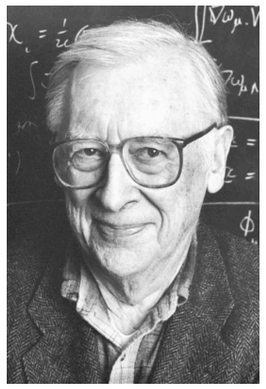
- Ground state lattice structure, phonons

- Solids with light elements

- Fermi surfaces, band structures (occupied part) of simple solids

Numerous open source software packages are readily available and easy to use. Calculations are straightforward (laptop). No deep theory/computation knowledge needed.





The Nobel Prize in Chemistry 1998: Walter Kohn "for his development of the density-functional theory" and John A. Pople "for his development of computational methods in quantum chemistry"

Computation – What would we *like* to do?

Physics perspective: *Need to connect to experiment & theory*

- Strong Electron Correlation (d- and f-electron materials)
- Finite temperature phenomena
- (Electronic) Phase transitions
- Simulation of optics experiment (ARPES, Raman, RIXS, conductivities, SHG…)
- Simulation of transport experiment
- Simulation of time-dependence
- Theory insight

Applied Math Perspective: *Need to be right for the right reason*

- A priori and a posteriori error estimate
- Possibility for adaptive refinement
- Rigorous uncertainty estimate
- No free parameters

It therefore becomes desirable that approximate practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation.

Paul Dirac, Proc. R. Soc. A 123 (1929), 714

Theoretical framework

Consider an electron…

…propagating through a solid.

Theoretical framework

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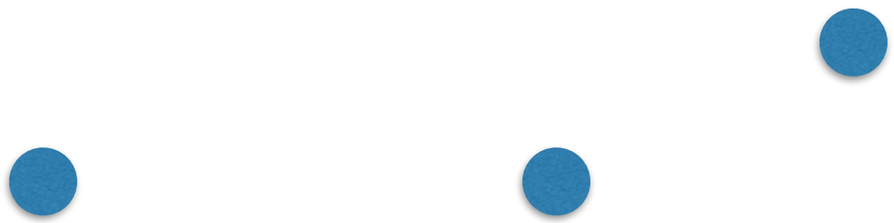
…propagating through a solid.

Theoretical framework

Consider an electron…

…when it encounters another electron, it gets deflected according to the Coulomb law…

…propagating through a solid.



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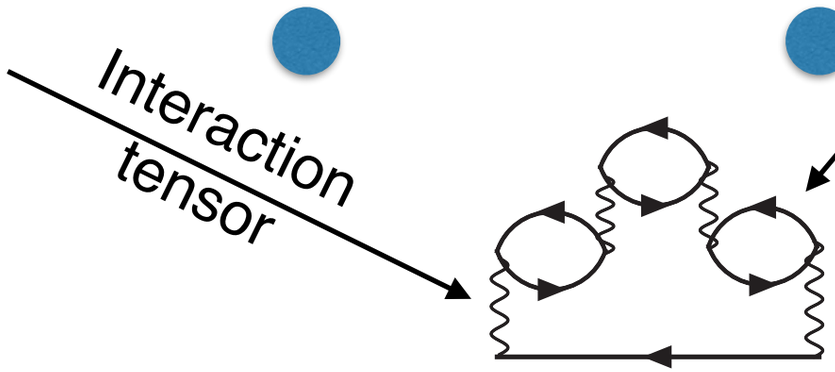

…propagating through a solid.

…and if there are few of these interaction processes, and they are simple enough, we can enumerate them one-by-one and draw pictures.

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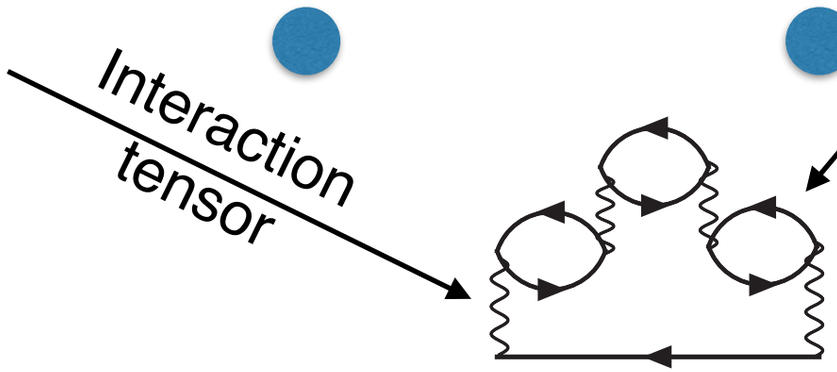

…propagating through a solid.

…and if there are few of these interaction processes, and they are simple enough, we can enumerate them one-by-one and draw pictures.

…but much of the formalism remains valid and directly connects to experiment: the 'propagators' are measured in ARPES experiment.

…when there are many electrons and complex 'entangled' processes, these simple pictures are not sufficient…

All of this is straightforward on paper! Diagrammatic perturbation theory of the 1960s…

Diagrammatic theories

Variants of theories:

- Bare low-order expansions (MP2, etc)
- Partial summation methods (RPA/‘GW’, ladders): Additionally capture certain fluctuations
- Conserving (Φ-/Ψ-derivable) expansions: Additionally satisfy certain conservation laws, thermodynamic consistency, and starting-point independence through self-consistency
- Embedding formulations: Additionally add non-perturbative contributions: route to strong correlations

Variants of formulation:

- Real-frequency formulation (typically zero-T)
- Matsubara formulation (nonzero-T)
- Real-time / Keldysh / Non-equilibrium formulation:

…and there is much more: Expansions in hopping, expansion in non-local interactions and correlations, expansion in external perturbations, …

Additional non-diagrammatic approximations: quasi-particles, …

Design decisions: ab-initio strong correlation theory, weak coupling part

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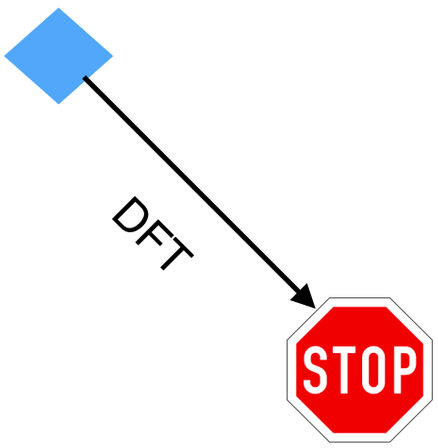

Interface to strong correlation:
Double counting, interactions, downfolding

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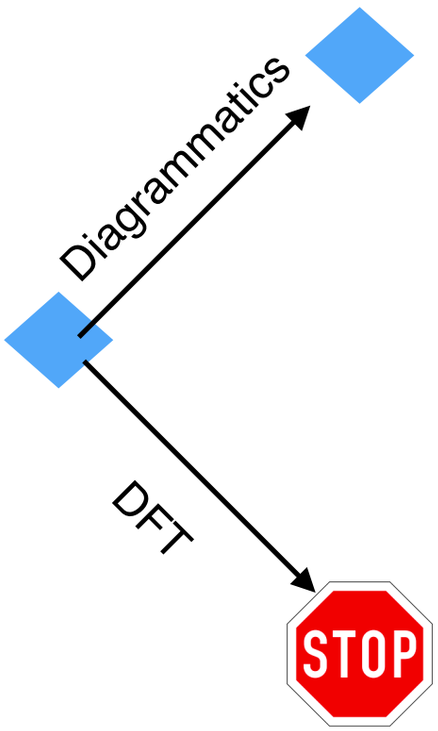

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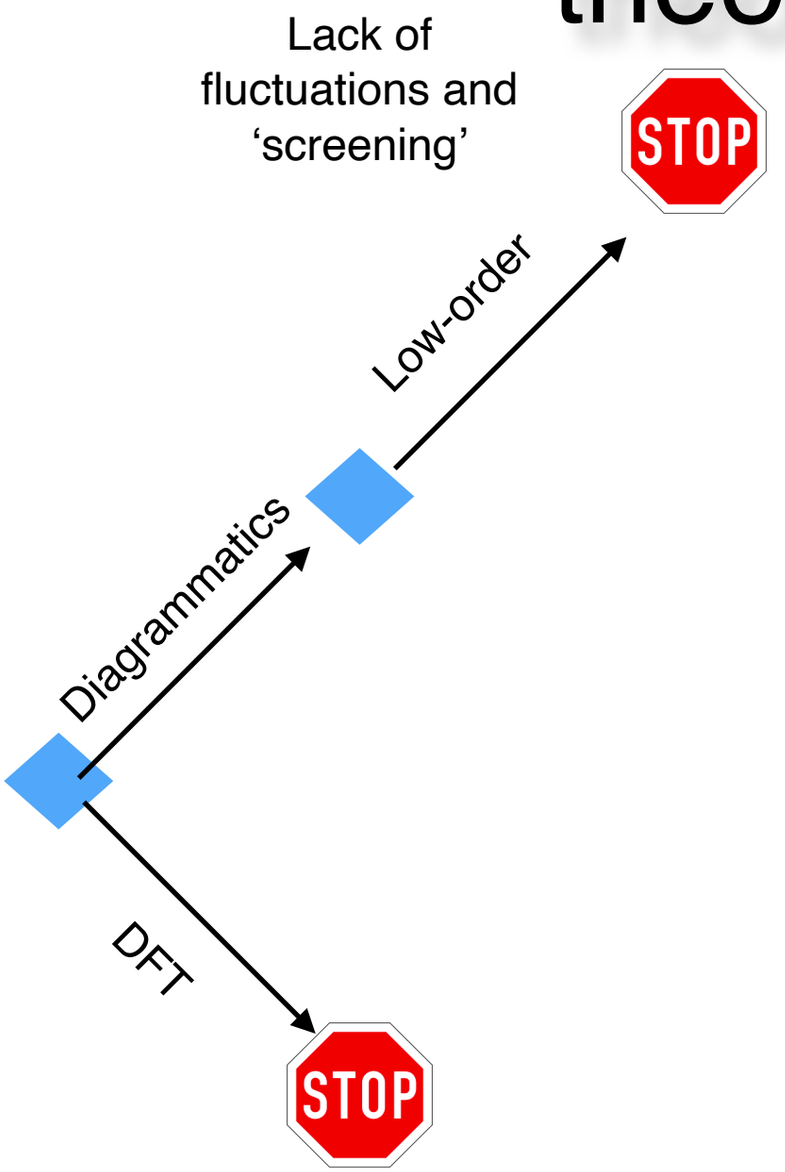

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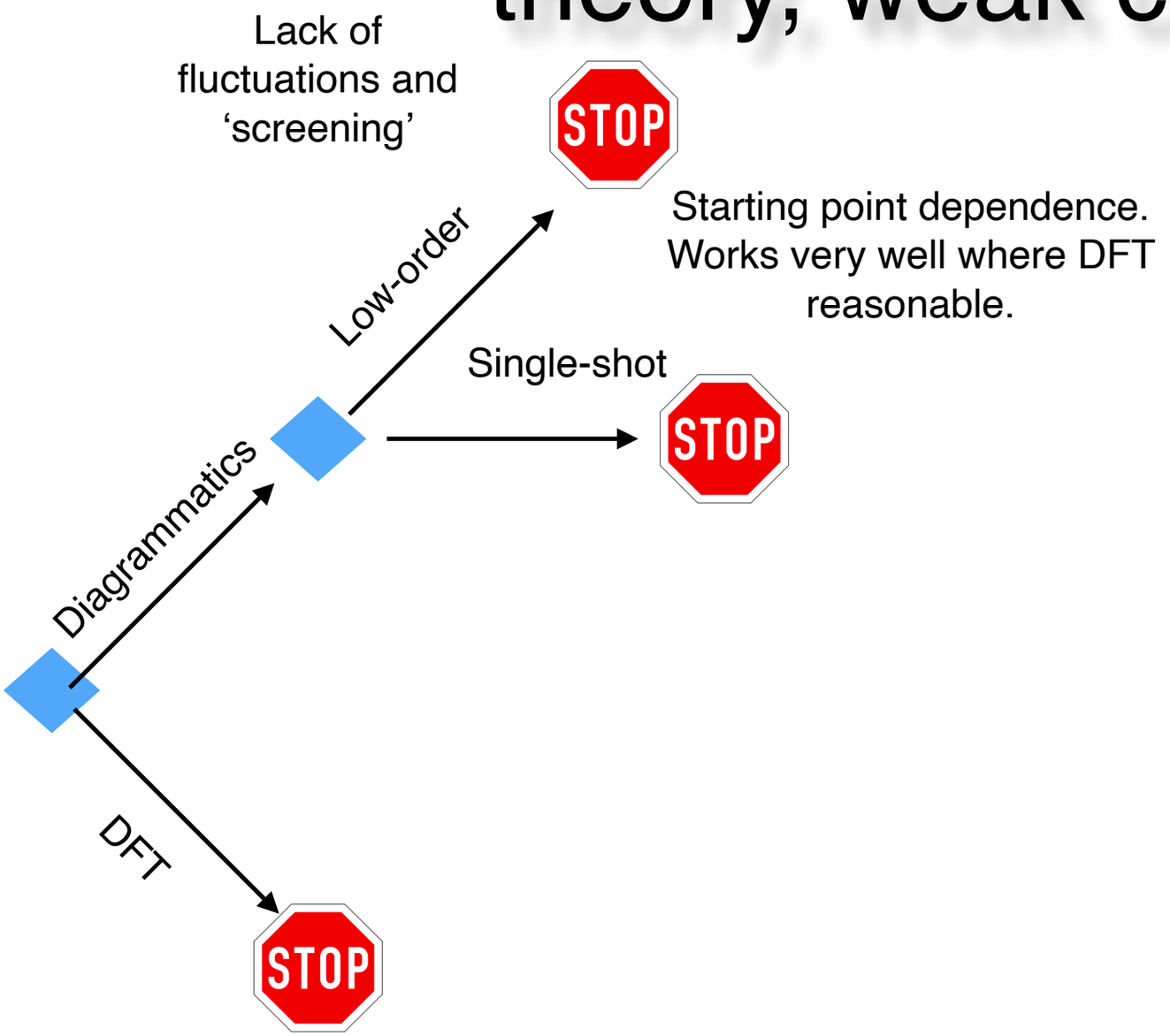

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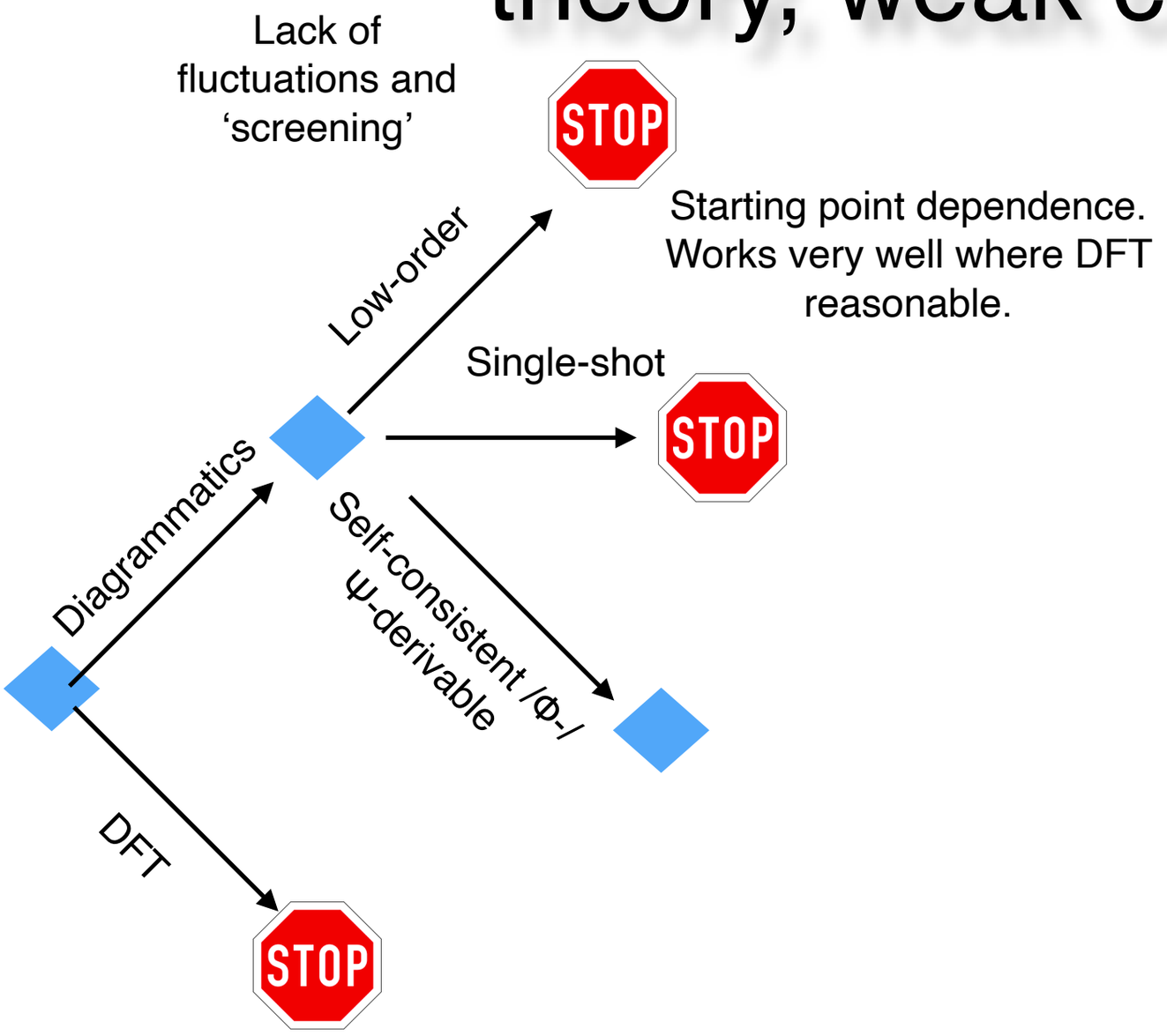

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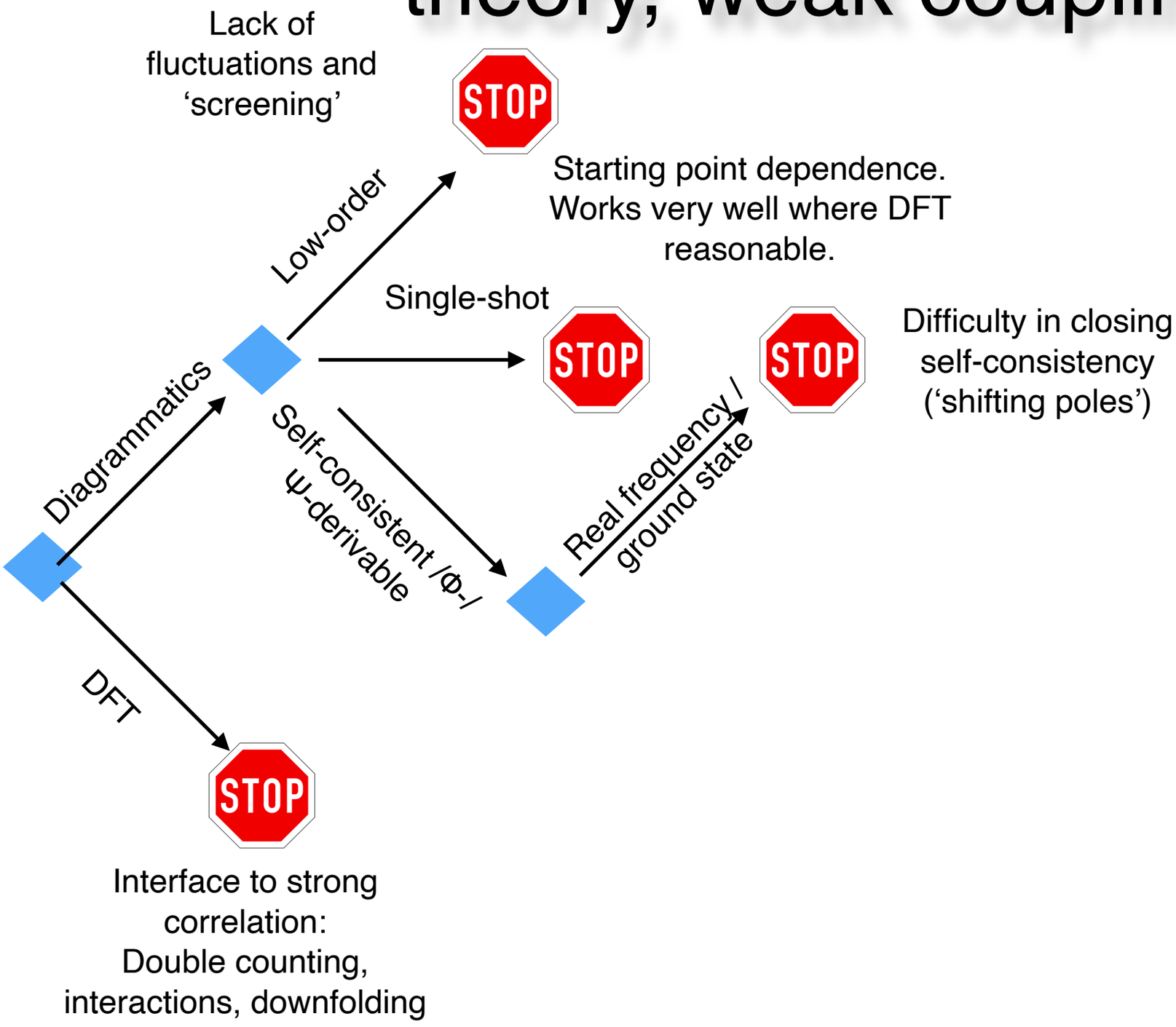

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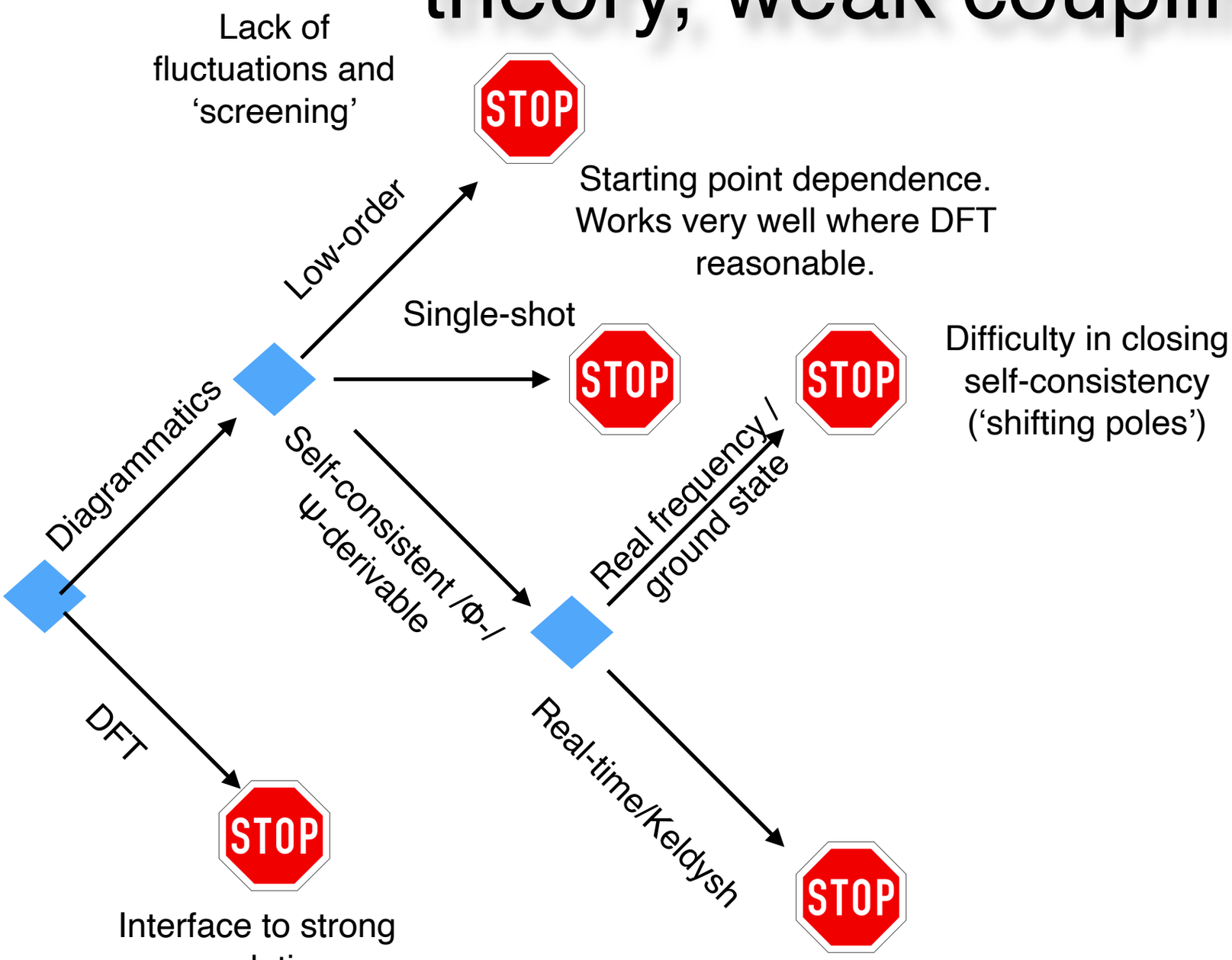

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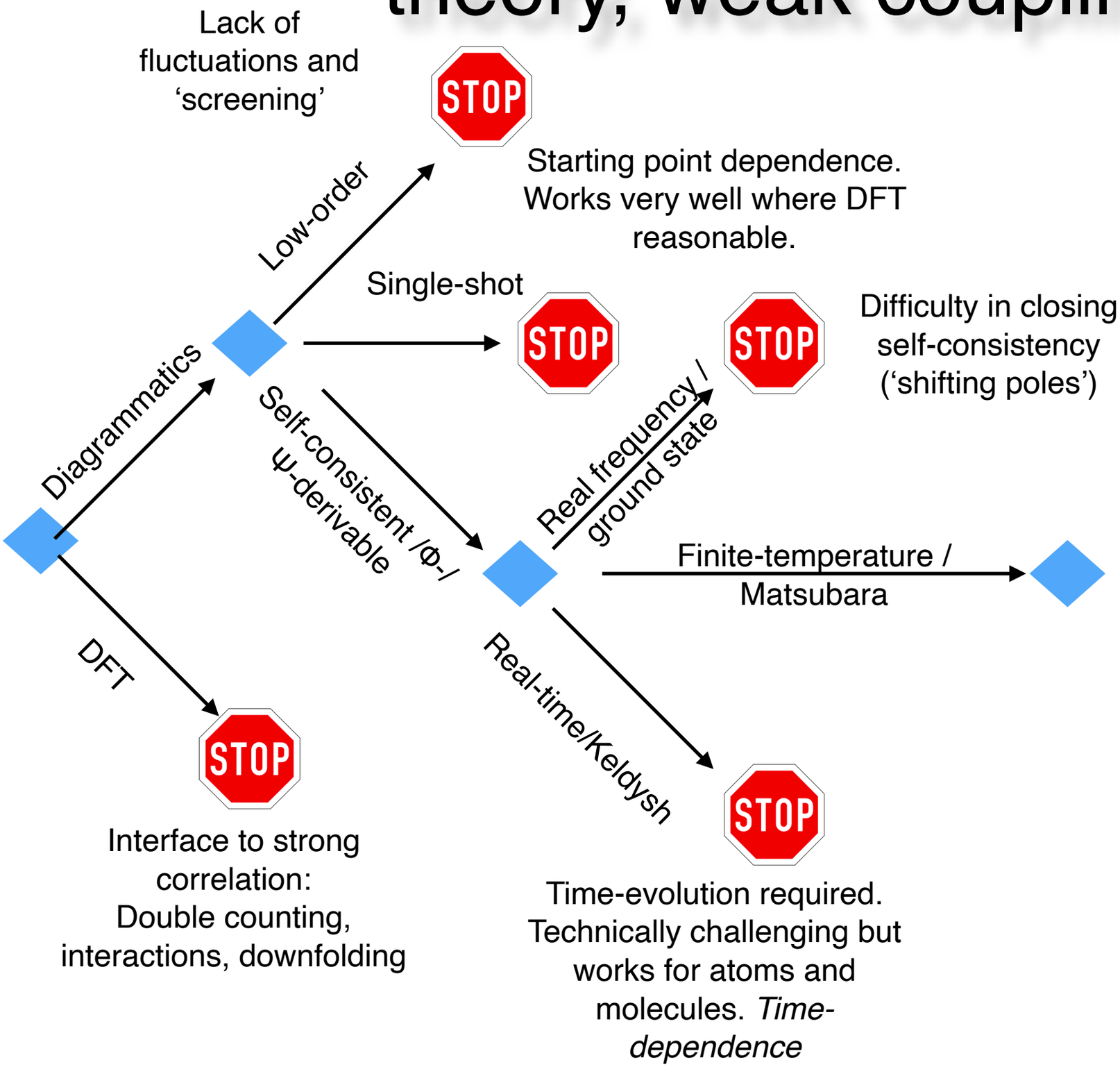

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Lack of fluctuations and 'screening'

STOP

Low-order

Starting point dependence. Works very well where DFT reasonable.

Single-shot

STOP

STOP

Difficulty in closing self-consistency ('shifting poles')

Diagrammatics

Self-consistent /Φ-/ Ψ-derivable

Real frequency / ground state

Energetics and thermodynamics

Consistency of thermodynamic equations (path independence), **Conservation** of number, energy, momentum, and angular momentum

Finite-temperature / Matsubara

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Interface to strong correlation: Double counting, interactions, downfolding

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Time-evolution required. Technically challenging but works for atoms and molecules. *Time-dependence*

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Same formulation as powerful impurity solvers. Embedding construction straightforward

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Response functions and gaps

Analytic continuation

Gaps, single-and two-particle spectral functions washed out

Laying the foundations – weak coupling

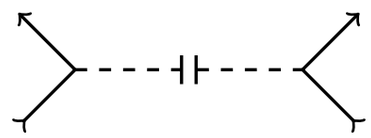
Remarkably non-trivial for a realistic solid!

- Finite temperature formalism: Billions of Matsubara frequencies needed to resolve bare energy scales and temperature

- Size of interaction tensor:
  - 120 orbitals per unit cell,
    minimal 4x4x4 grid would use 405 TB of storage.

$$V_{ijkl} = \iint d^3r d^3r' \frac{\phi_i^*(r)\phi_j(r)\phi_k^*(r')\phi_l(r)}{|r - r'|}$$

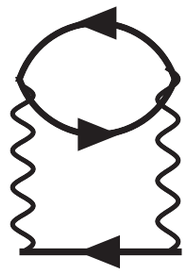
- Computational effort for even the simplest analytic theories far beyond current capabilities



- Remarkable behind-the-scenes technical / numerical developments over the last years in basis functions, compression methods, adaptive methods to enable low-order diagram calculations in solids *without additional approximations*.

Yeh, Iskakov, Zgid, Gull, arXiv:2206.07660

Supersizing weak coupling expansions

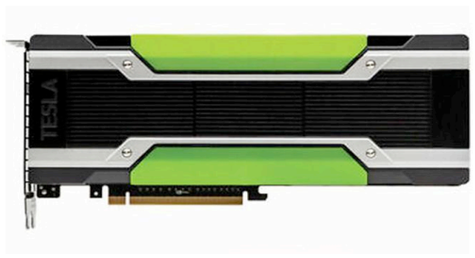


Express diagram contraction as a sequence of blocked dense matrix multiplications

Major peculiarity: O(N3) operations, O(N2) data.
Large arithmetic intensity, little sensitivity to latency.

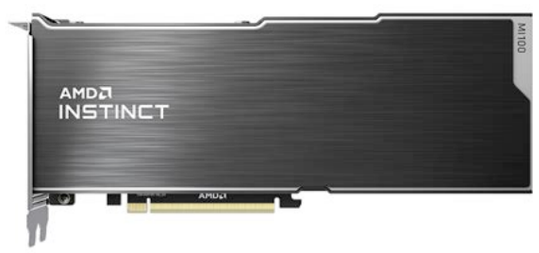
Graphics cards (GPUs) are extremely efficient at running dense matrix multiplications!

High-end CPU:
~1 TFlops/s, ~4K$



Prev-gen GPU:
~6 TFlops/s

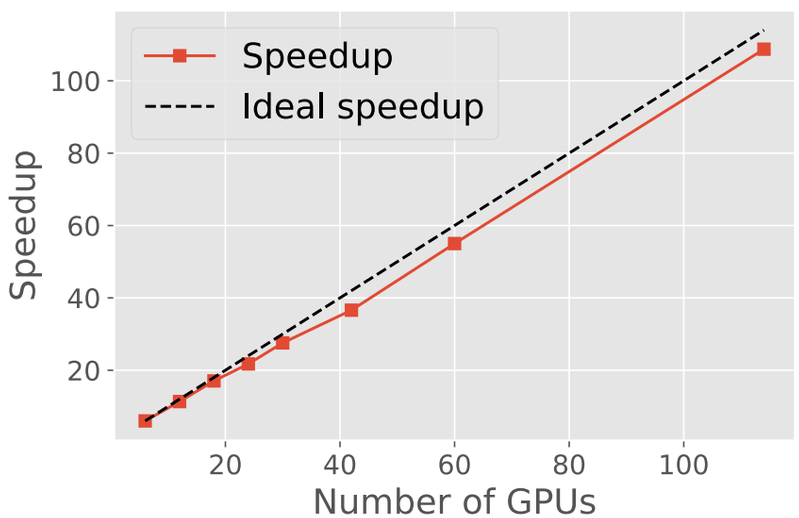
Current-gen GPU:
~20 TFlops/s

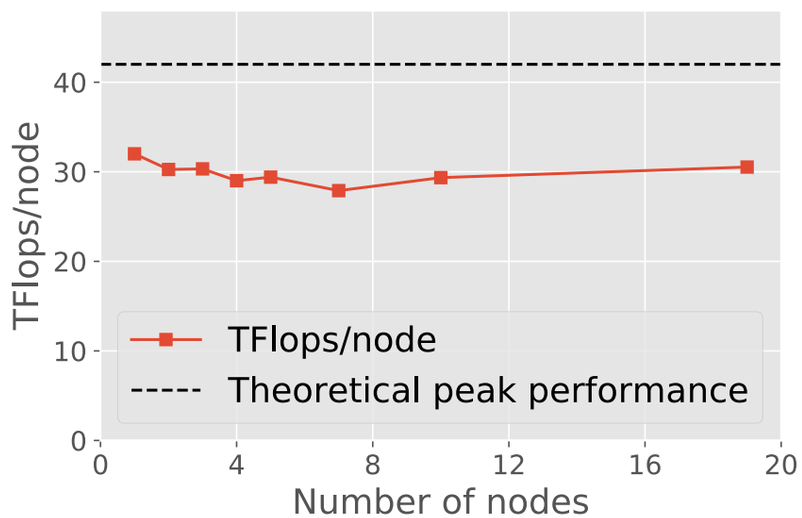


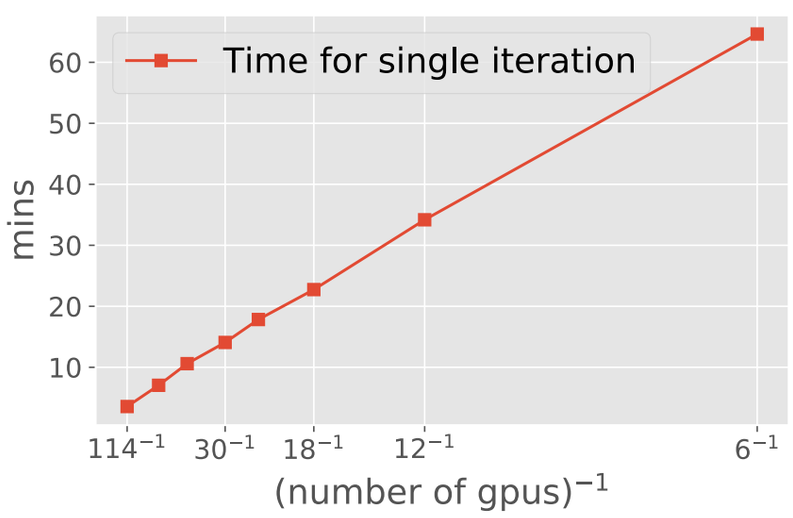
Next-gen GPU:
~100 TFlops/s, ~10K$

C-N Yeh, S. Iskakov, D. Zgid, E. Gull, to be submitted

Weak coupling on supercomputers





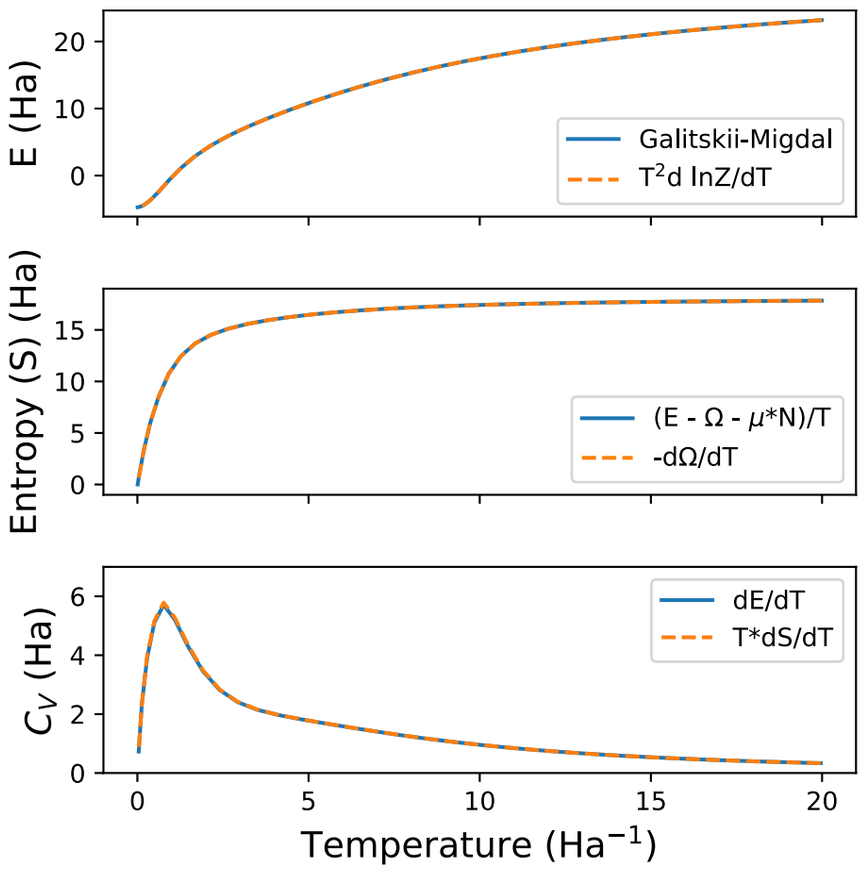


Antiferromagnetic MnO (6x6x6) within [fully self-consistent] GW on summit at ORNL

Runs at ~75% theoretical peak FLOP sustained, scales to hundreds of GPUs.

Finite-temperature weak coupling simulations of solids with ‘kosher’ many-body diagrammatics are now possible!

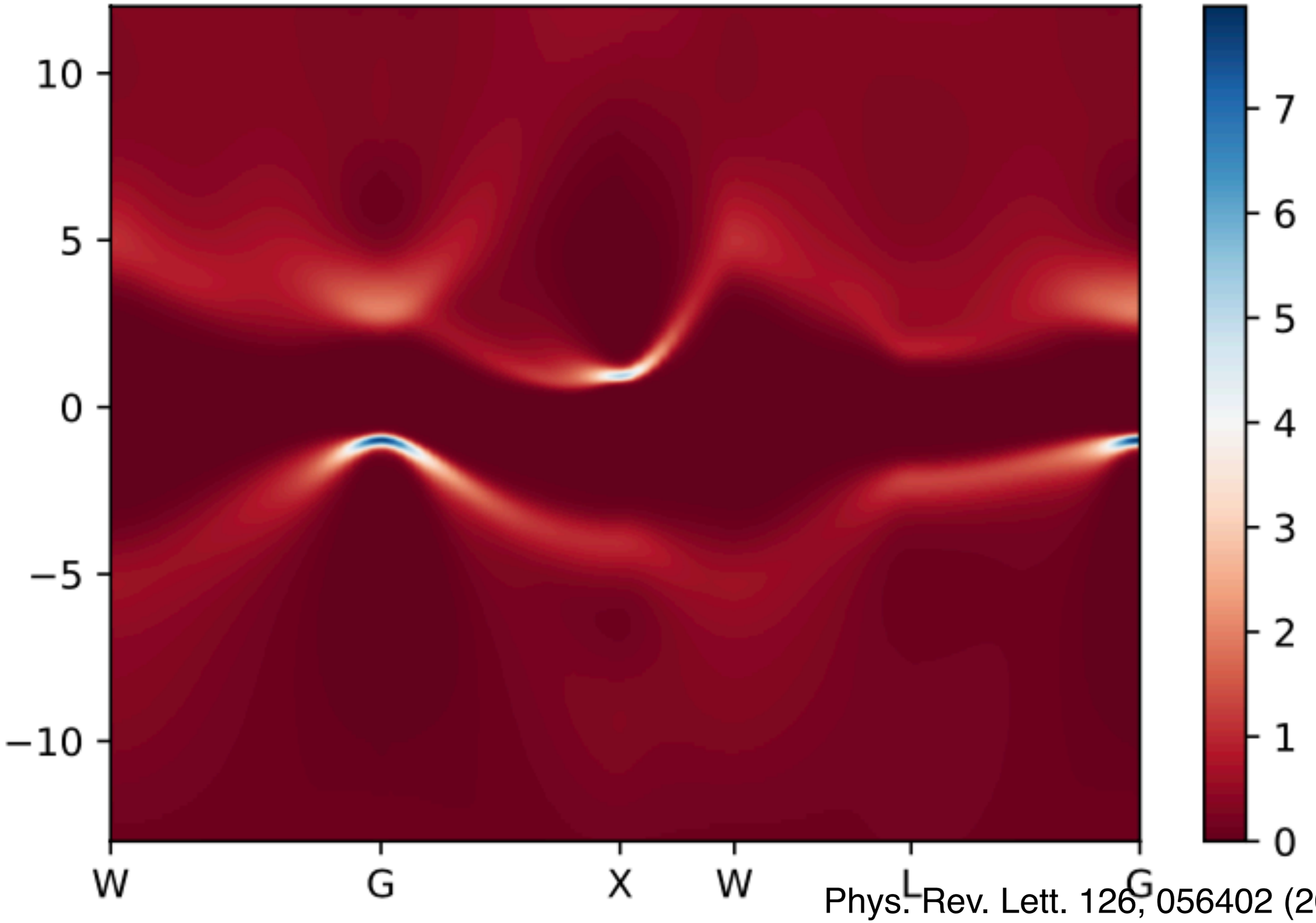
Thermodynamic consistency (…of the electronic system) validated in practice for real materials, both for self-consistent second-order PT and self-consistent GW



C-N Yeh, S. Iskakov, D. Zgid, E. Gull, (2022)

Obtaining spectral functions

Si, 6x6x6 lattice, self-consistent GW



Phys. Rev. Lett. 126, 056402 (2021)

Obtaining spectral functions

- Well-known problem with finite-temperature field theories: analytic continuation to the real axis to obtain spectral functions is ill conditioned.

$$G(i\omega_n) = -\frac{1}{\pi}\int \frac{\mathrm{Im}G(\omega)d\omega}{i\omega_n - \omega}$$

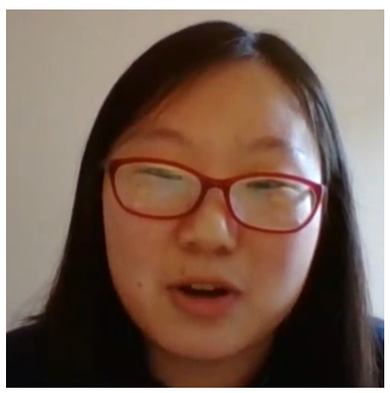
$$A(\omega) = \frac{-1}{\pi}\mathrm{Im}G(\omega)$$

$$G(i\omega_n) = K(i\omega_n, \omega)G(\omega)$$

$$\downarrow$$

$$G(\omega) = [K(i\omega_n, \omega)]^{-1}G(i\omega_n)$$

- Little progress can be made where data is noisy. However, when data is accurate, progress is possible…



Jiani Fei

- Green's functions satisfy mathematical ('Nevanlinna') properties

$$\mathrm{Im}\ G_\gamma(z) \leq 0 \quad \text{for} \quad z \in \mathbb{C}^+$$

- It is possible (and rather straightforward) to construct a numerical method that satisfies these mathematical properties
- Doing so vastly constrains the solution space and yields much better continuations.

Phys. Rev. Lett. 126, 056402 (2021)

Green's functions & Lehmann Representation

Lehmann representation

$$G_\gamma(z) = \frac{1}{Z}\sum_{m,n}\frac{|\langle m|c_\gamma^\dagger|n\rangle|^2}{z+E_n-E_m}(e^{-\beta E_n}+e^{-\beta E_m})$$

G coincides with Matsubara Green's function on imaginary axis, with retarded Green's function just above real axis. Define

$$A = \frac{1}{Z}|\langle m|c_\gamma^\dagger|n\rangle|^2(e^{-\beta E_b}+e^{-\beta E_m}) > 0$$

For

$$z = x+iy$$

$$S = \frac{A}{(x+E_n-E_m)+iy} = \frac{A(x+E_n-E_m-iy)}{(x+E_n-E_m)^2+y^2}$$

$$\mathrm{Im}S = \frac{-Ay}{(x+E_n-E_m)^2+y^2}$$

And therefore for any Green's function, independent of the system:

$$\mathrm{Im}\ G_\gamma(z) \leq 0 \quad \text{for} \quad z \in \mathbb{C}^+$$

Phys. Rev. Lett. 126, 056402 (2021)

Nevanlinna and Schur functions

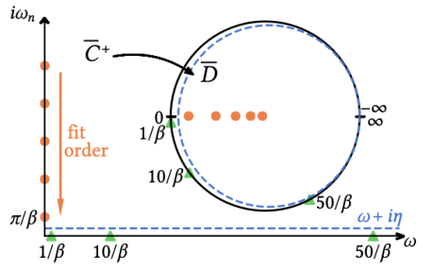
Rolf Nevanlinna

Nevanlinna functions are functions with a positive imaginary part on the upper half of the complex plane.

$\mathcal{N}G = -G$ Is a Nevanlinna function

The invertible Möbius transform h maps the upper half plane to the unit disk

$$h(z) : z \to \frac{z - i}{z + i}$$



$$\mathcal{D} = \{z : |z| < 1\};$$

$$\overline{\mathcal{D}} = \{z : |z| \leq 1\}$$

Nevanlinna functions can be mapped onto Schur functions: Schur functions map the open unit disk D to the closed unit disk D (‘contractive’ functions). Every Schur function has a continued fraction expansion that can be recursively defined.

Combine mapping to contractive functions with Schur’s continued fraction expansion to obtain an intrinsically causal expansion for Green’s functions

Phys. Rev. Lett. 126, 056402 (2021)

The Schur algorithm

Input data $f(Y_i) = C_i \quad i = 1,2,\ldots,M \qquad Y_i = i\omega_n \in \mathcal{C}^+ \text{ and } C_i \in \mathcal{C}^+$

Contractive interpolant.

$$\theta(Y_i) = \lambda_i = h(C_i) = \frac{C_i - i}{C_i + i} \quad i = 1,2,\ldots,M$$

Start the interpolation by constructing an interplant through Y_1. Express this contractive interpolant as a function that is zero at Y_1, and a constant λ_1:

We want $\theta(Y_1) = \lambda_1 \quad |\lambda_1| < 1$

Functional form $\theta(z) = \dfrac{\phi(z) + \lambda_1}{\lambda_1^* \phi(z) + 1}$ Where $\phi(z) = \dfrac{z - Y_1}{z - Y_1^*}\theta_1(z)$

Such that $\phi \in \mathrm{B} \text{ and } \phi(Y_1) = 0$

Issai Schur

J. Schur, Uber potenzreihen, die im innern des einheitskreises beschränkt sind, Journal für die reine und angewandte Mathematik **1918**, 122 (1918).

Note that $\theta_1(z)$ is now an arbitrary contractive function. Express it as a sum of a function that is λ_2 at Y_2 and an arbitrary contractive function. Express that one as the sum of a function that is λ_3 at Y_3 and an arbitrary contractive function, iterate and repeat for all interpolation points.

This will result in an expression for **all possible interpolants** in terms of a remaining arbitrary Schur/Nevanlinna function. We will use this freedom later.

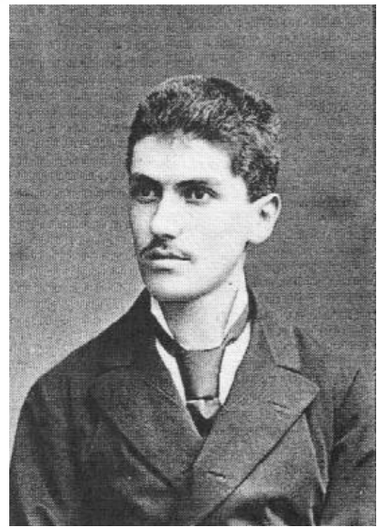
Phys. Rev. Lett. 126, 056402 (2021)

The Pick criterion: existence of interpolants

If $g(x_i) = y_i \quad (x_i \in \mathcal{D}, y_i \in \overline{\mathcal{D}})$

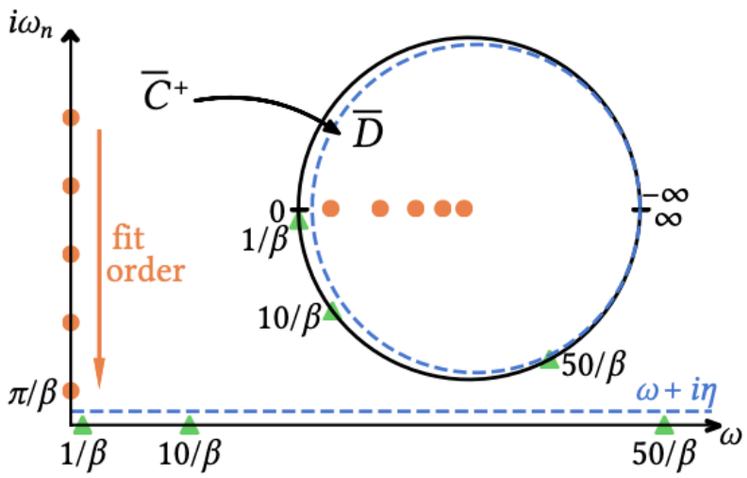
Then a Schur interpolant to g can be found iff the Pick matrix is positive semi-definite. It has a unique solution if furthermore the Pick matrix is singular.

$$P_{ij} = \left[\frac{1 - y_i y_j^*}{1 - x_i x_j^*} \right]$$



Georg A. Pick

G. Pick, Über die beschränkungen analytischer funktionen, welche durch vorgegebene funktionswerte bewirkt werden- ber die beschränkungen analytischer funktionen, welche durch vorgegebene funktionswerte bewirkt werden, Math. Ann. **78**, 270 (1917).



Provides a straightforward check on any input data. Transform the data to the unit circle, evaluate Pick matrix, check if it has negative eigenvalues. If it does, there WILL NOT be a positive spectral function.

Interesting observation: Monte Carlo data never fulfills this criterion. GW data only if very well converged and not too many interpolation points. Synthetic benchmark data shows very high precision at high frequency needed to make it work. Sign of the very constrained nature of Nevanlinna/Schur function space.

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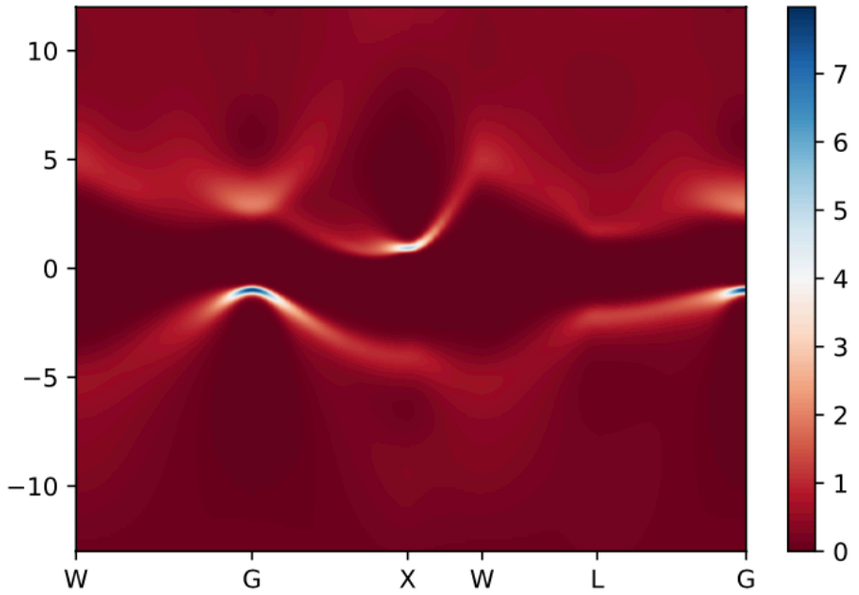
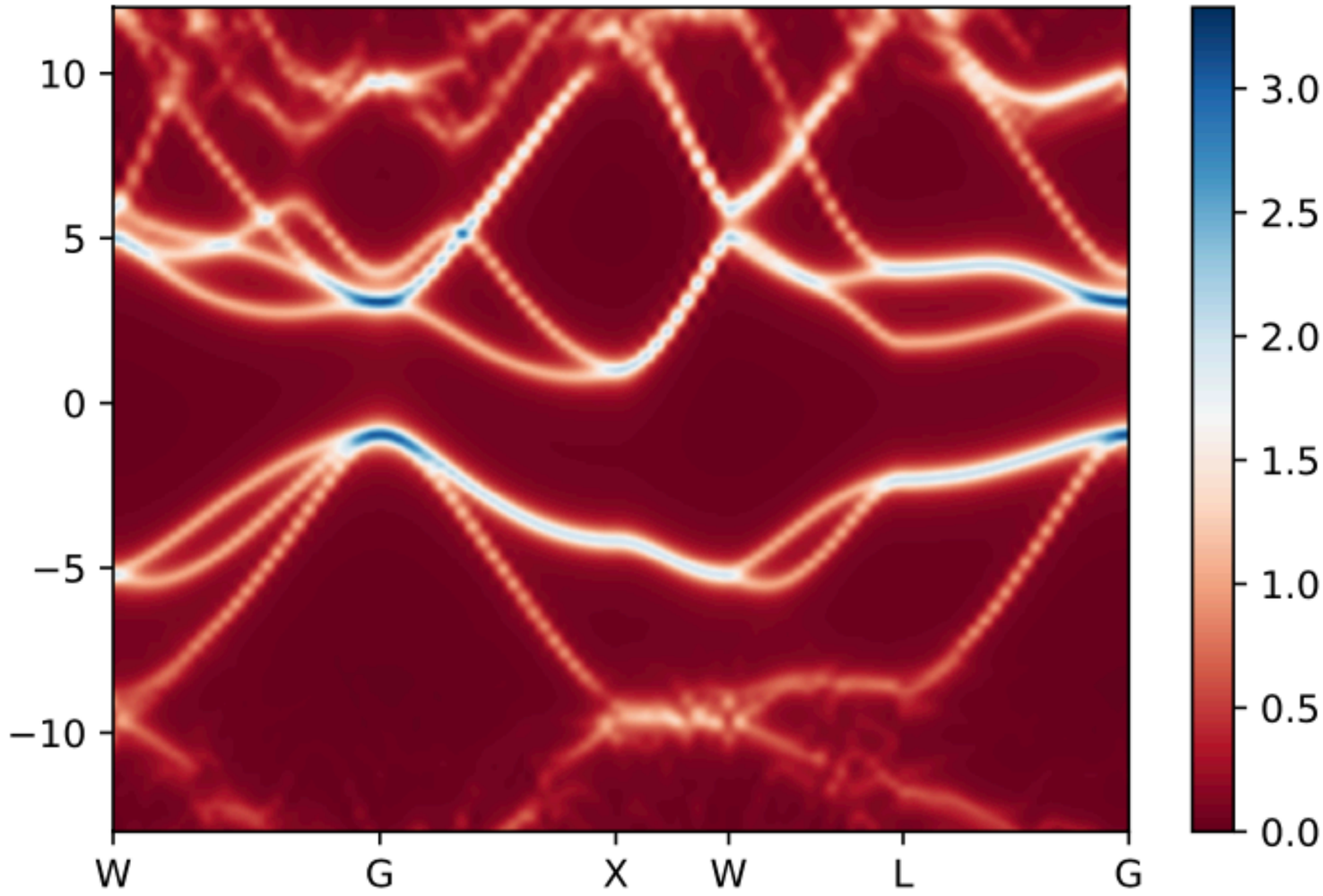
Old Technology vs Nevanlinna



Maxent, orbital- and k-resolved

Band structure is visible, individual bands can be separated

Both continuations operating on same input data!



Nevanlinna, orbital- and k-resolved

Fully self-consistent GW of Si, no quasiparticle or similar approximations, analytic continuation of fully interacting Green's function.

Analytic continuation

Phys. Rev. Lett. 126, 056402 (2021)

Matrix-valued Carathéodory generalization

The Carathéodory class of matrix-valued analytic functions in the unit disk (or: upper half plane) is defined as

$$C = \{M(z) : M(z) + M^\dagger(z) \geq 0 \quad \forall\ |z| < 1\}$$

Note that $$M(z) + M^\dagger(z) \geq 0 \ \leftrightarrow\ \mathrm{Re}\{x^\dagger M(z) x \geq 0\}$$

i.e. the real part of M is positive semidefinite.

$-iG^<(\omega)$ is Carathéodory:

Κωνσταντίνος Καραθεοδωρή

C. Carathéodory, Über den variabilitätsbereich der koeffizienten von potenzreihen, die gegebene werte nicht annehmen, Math. Ann. **64**, 95 (1907).

$$G^<_{ij}(\omega) = 2\pi i \sum_{mn} \frac{e^{-\beta E_n}}{Z} \langle n|c_j^\dagger|m\rangle\langle m|c_i|n\rangle \delta(\omega - E_n + E_m)$$

Insert x, do the Math:

$$\langle x| - iG^<(\omega)|x\rangle = 2\pi \sum_{mn} \frac{e^{-\beta E_n}}{Z} |\langle m| \sum_i c_i x_i^*|n\rangle|^2 \delta(\omega - E_n + E_m)$$

Phys. Rev. B 104, 165111(2021)

The Hubbard Dimer

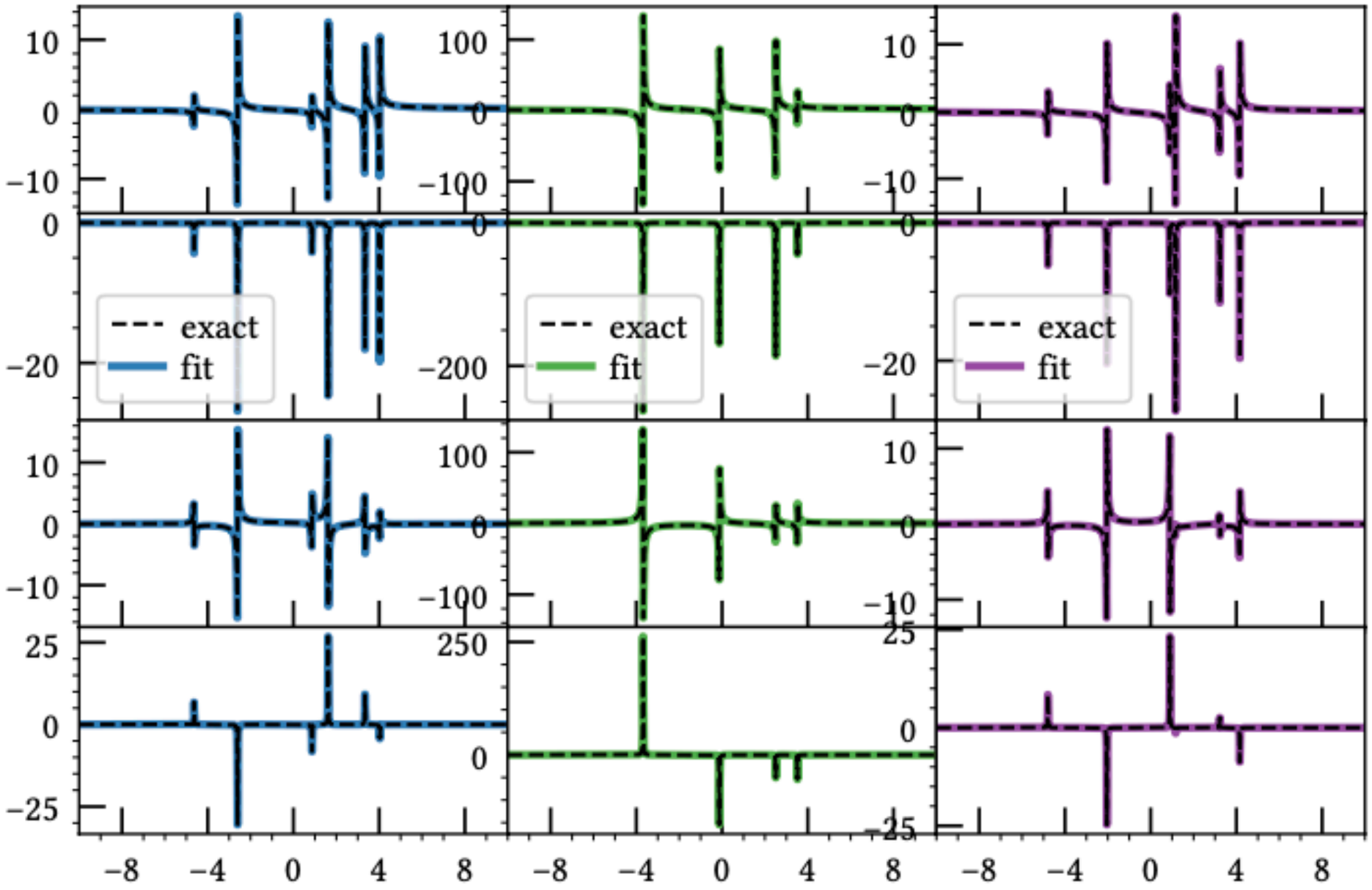


Figure 2.2: The Hubbard dimer model interpolation, 2 random non-zero entries [rows are real(1), imaginary(1), real(2), imaginary(2) part] of the 4×4 matrices, on the real axis. Using input data with 10^{-7} standard deviation Gaussian noise. Left are Green's functions, middle are self-energies, right are cumulants. The exact data comes from analytic model formula. Fitted data comes from continuing matrices from the imaginary axis, which we get from analytic formula and disturbed with noise.

Phys. Rev. B 104, 165111(2021)

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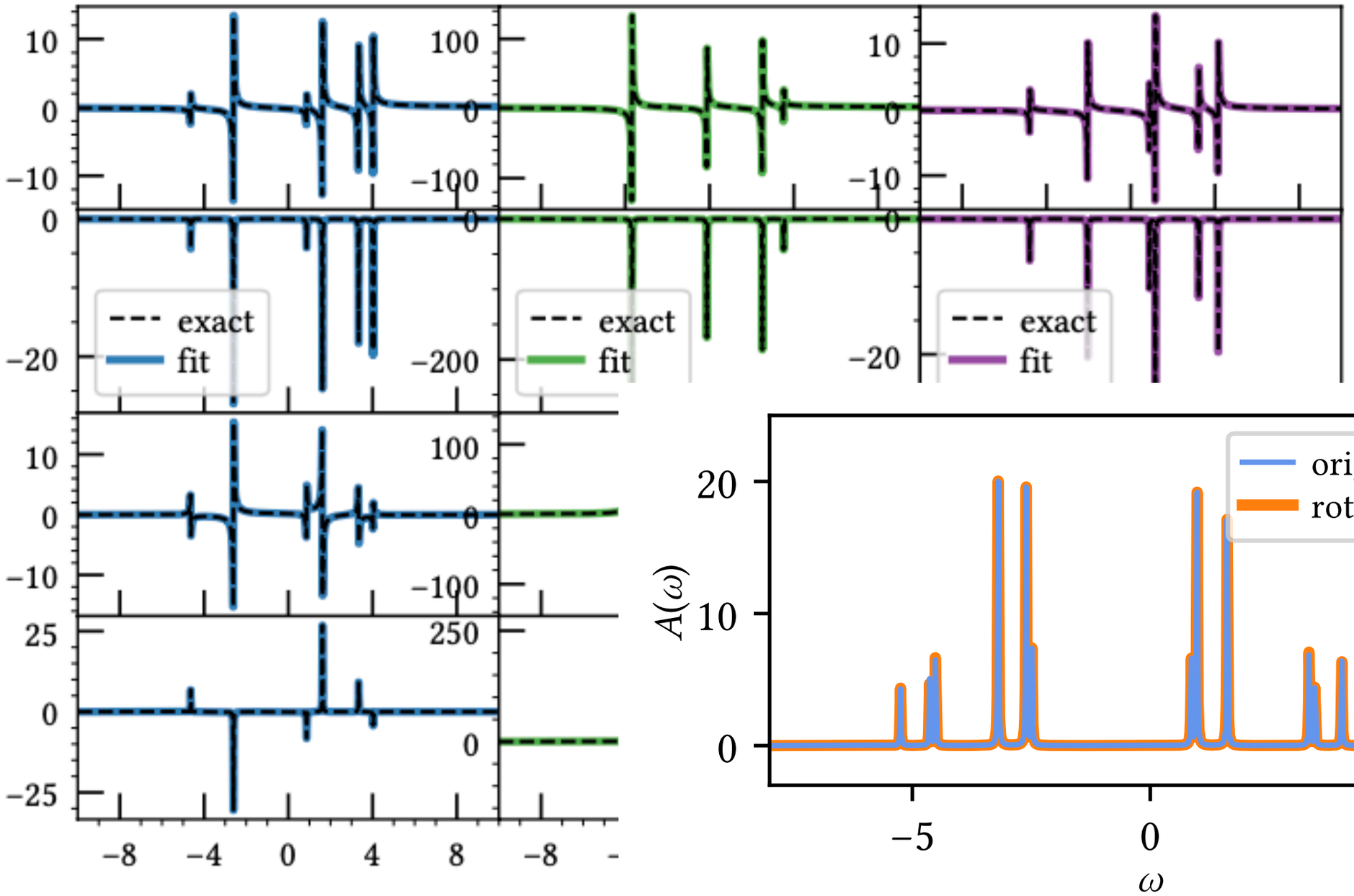

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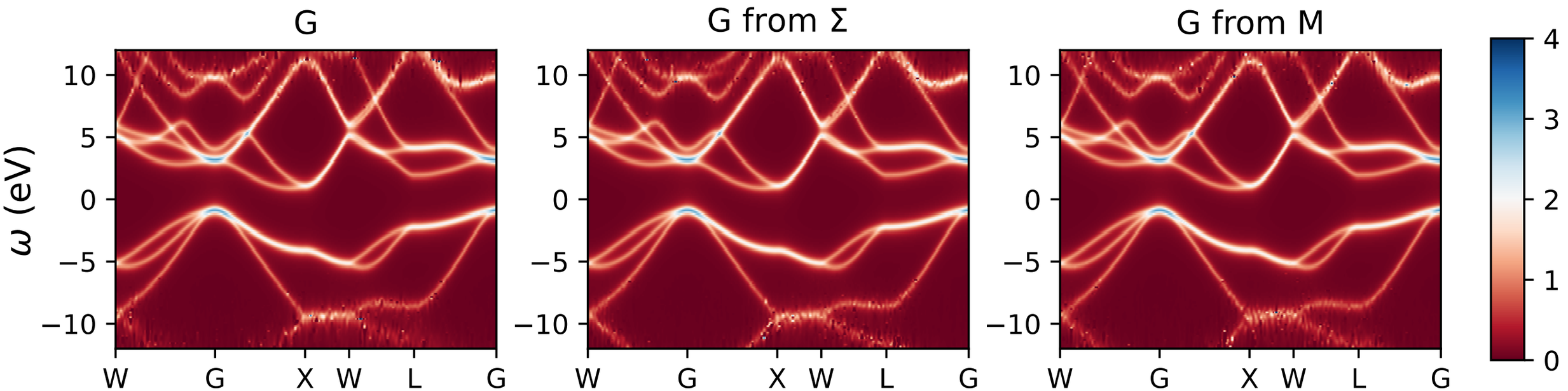
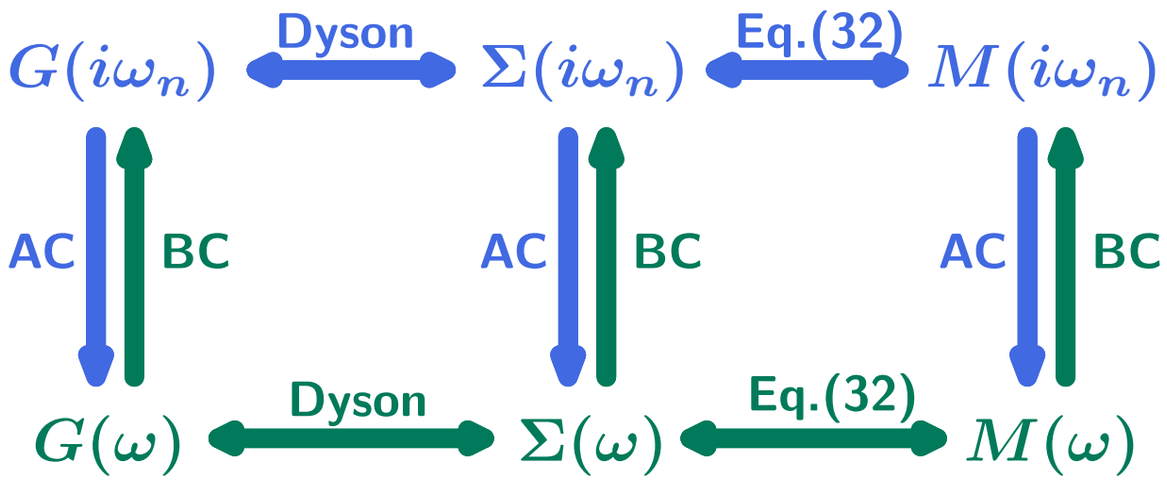
Phys. Rev. B 104, 165111(2021)

Dyson commutes with continuation

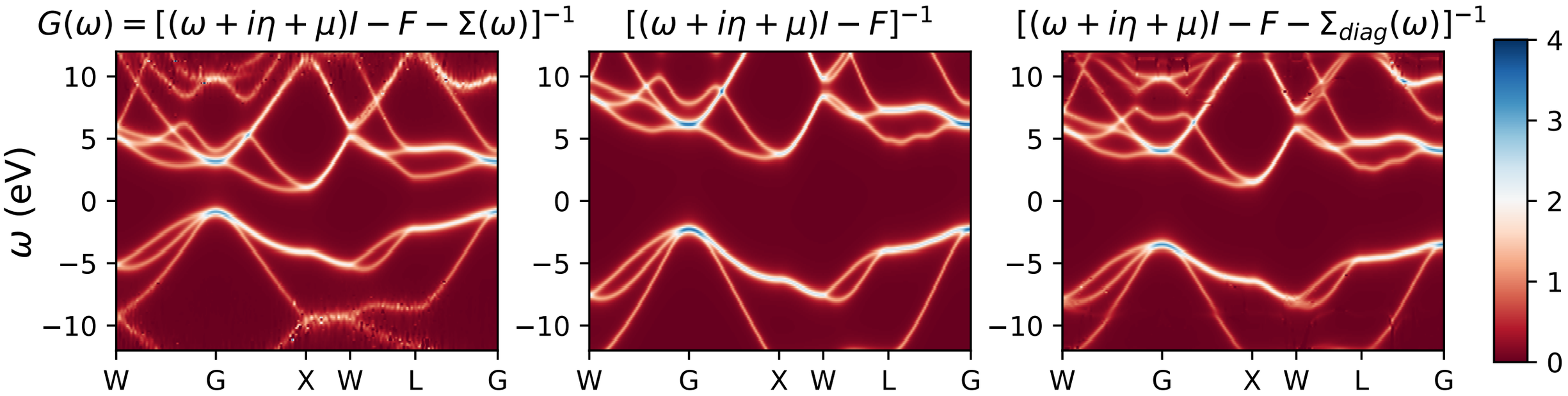


Band structure of crystalline Silicon (26 orbitals per unit cell), simulation in Gaussian orbitals with fully self-consistent GW; matrix valued continuation followed by Dyson equation.



Phys. Rev. B 104, 165111(2021)

Careful with approximations!



Truncation of the dynamical part of the self-energy (just the Fock matrix of the fully interacting system); or truncation of the self-energy to just diagonal parts.

‘Diagonal’ approximations to the self-energy have a huge effect on the band structure.

Careful, this is what is usually done in LDA+DMFT-type calculations… and compensated for by an appropriate choice of U, double counting, and downfolding.

Phys. Rev. B 104, 165111(2021)

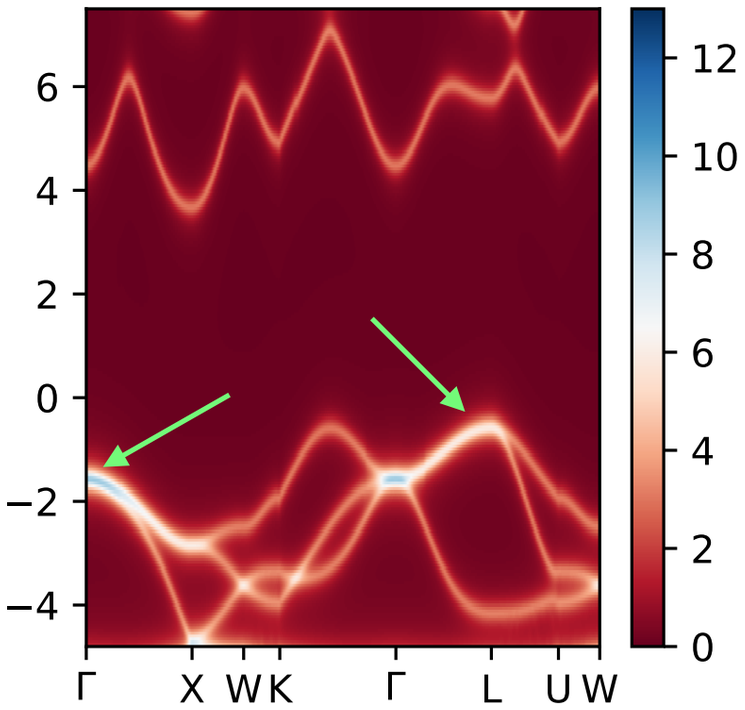
Adaptation to relativistics (SOC)

- Spin-orbit coupling emerges ab-initio from the solution of the Dirac, rather than the Schrödinger equation. Electrons and Positrons!

- Recent development in quantum chemistry: exact two-component relativistic approach. In the x2c1e approximation, the two-body integrals remain non-relativistic

  - Neglects relativistic corrections to the two-body integrals

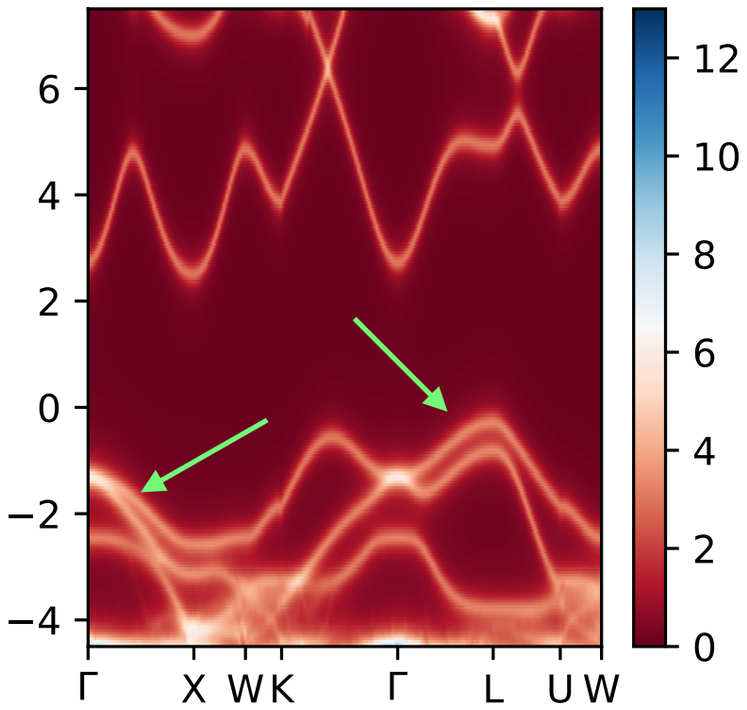
  - Diagrammatic structure remains unchanged, interaction vertices remain unchanged, bare propagators pick up relativistic contributions. (Identical impurity solvers!)



Non-relativistic

Example: self-consistent GW simulation of AgI in the x2c1e approximation

Parameter free ab-initio spin-orbit coupling splits bands



Relativistic x2c1e

C-N Yeh, *A.* Shee, Q. Sun, E. Gull, D. Zgid, Phys. Rev. B 106, 085121 (2022)

Design decisions: beyond weak coupling

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Remains at weak coupling

Add a ‘vertex correction’

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Add fluctuation channels

Technically doable but not systematically improving results, captures electronic phase transitions

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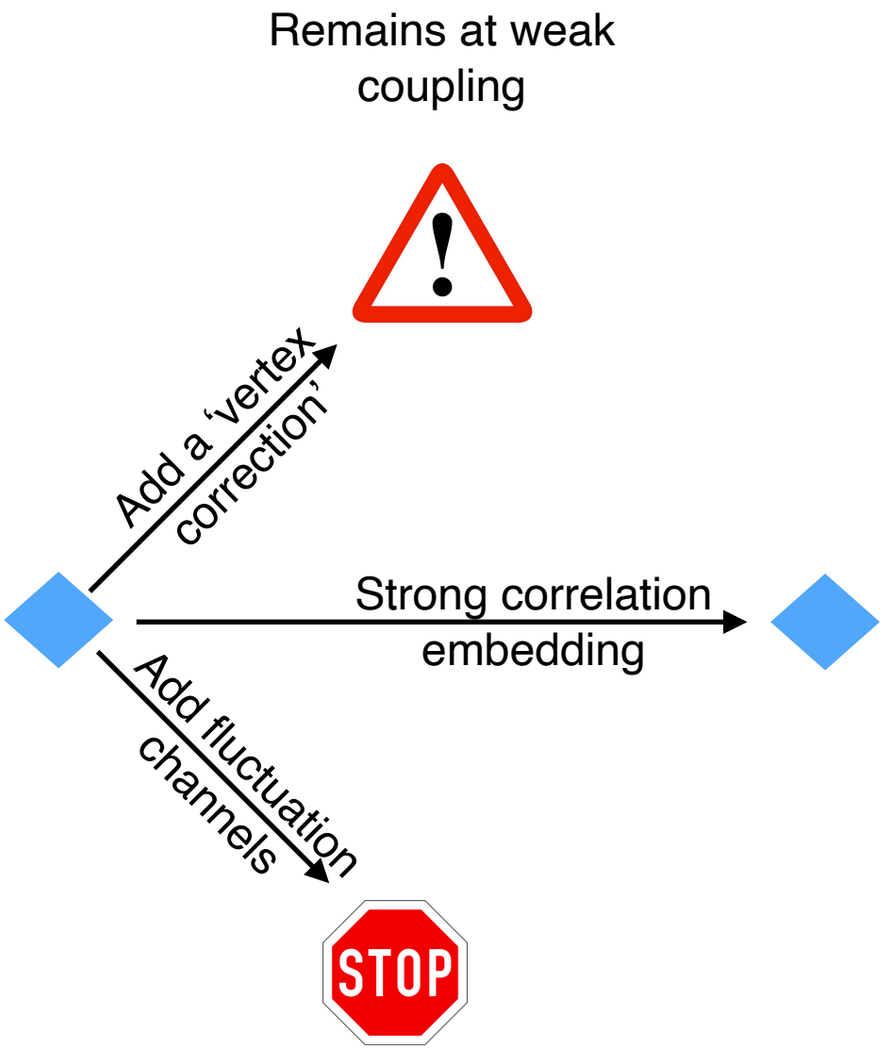

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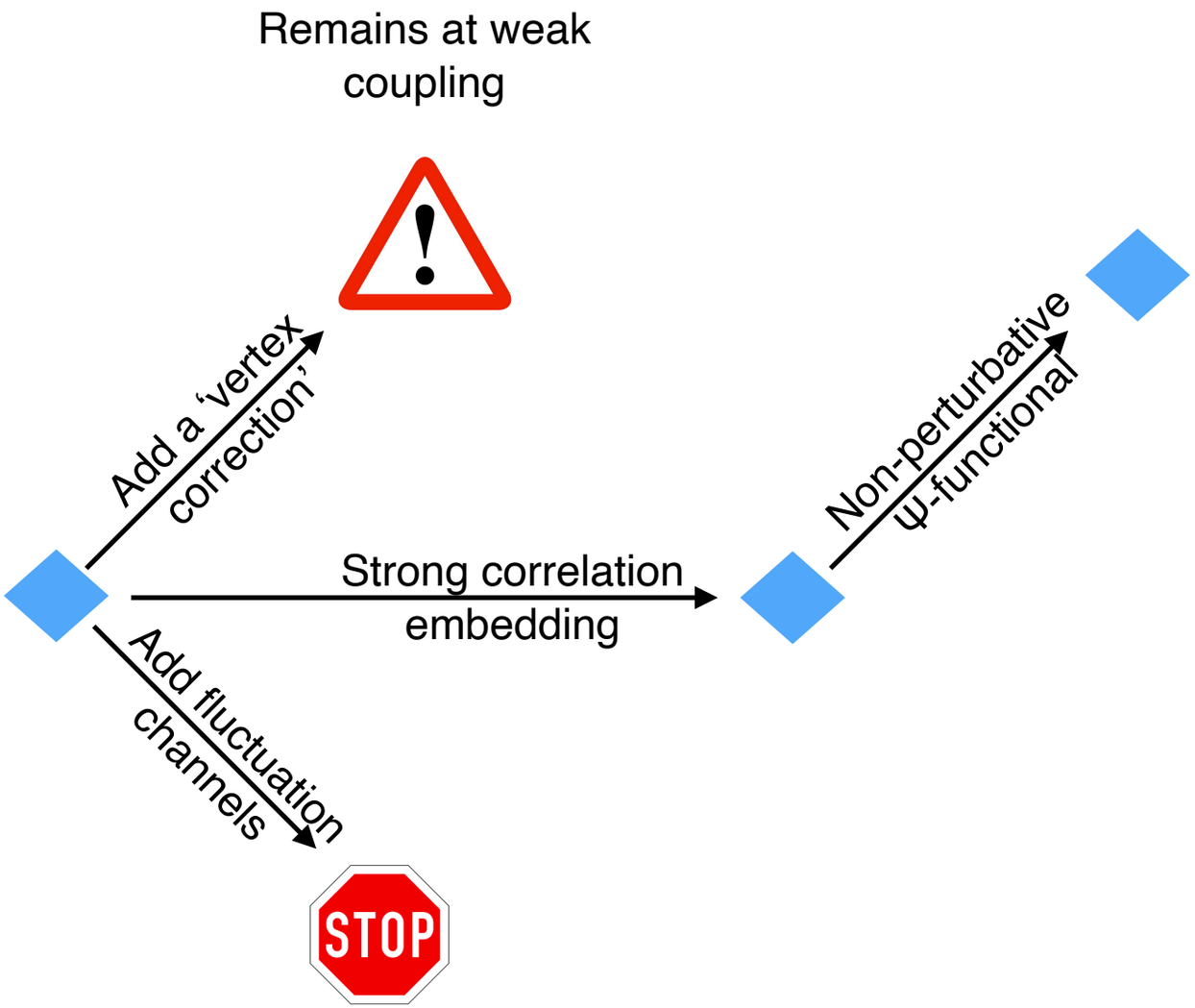

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Strong correlation embedding

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Non-perturbative Ψ-functional

No truncation of hybridization and interactions

No impurity solvers for retarded interactions and retarded propagators

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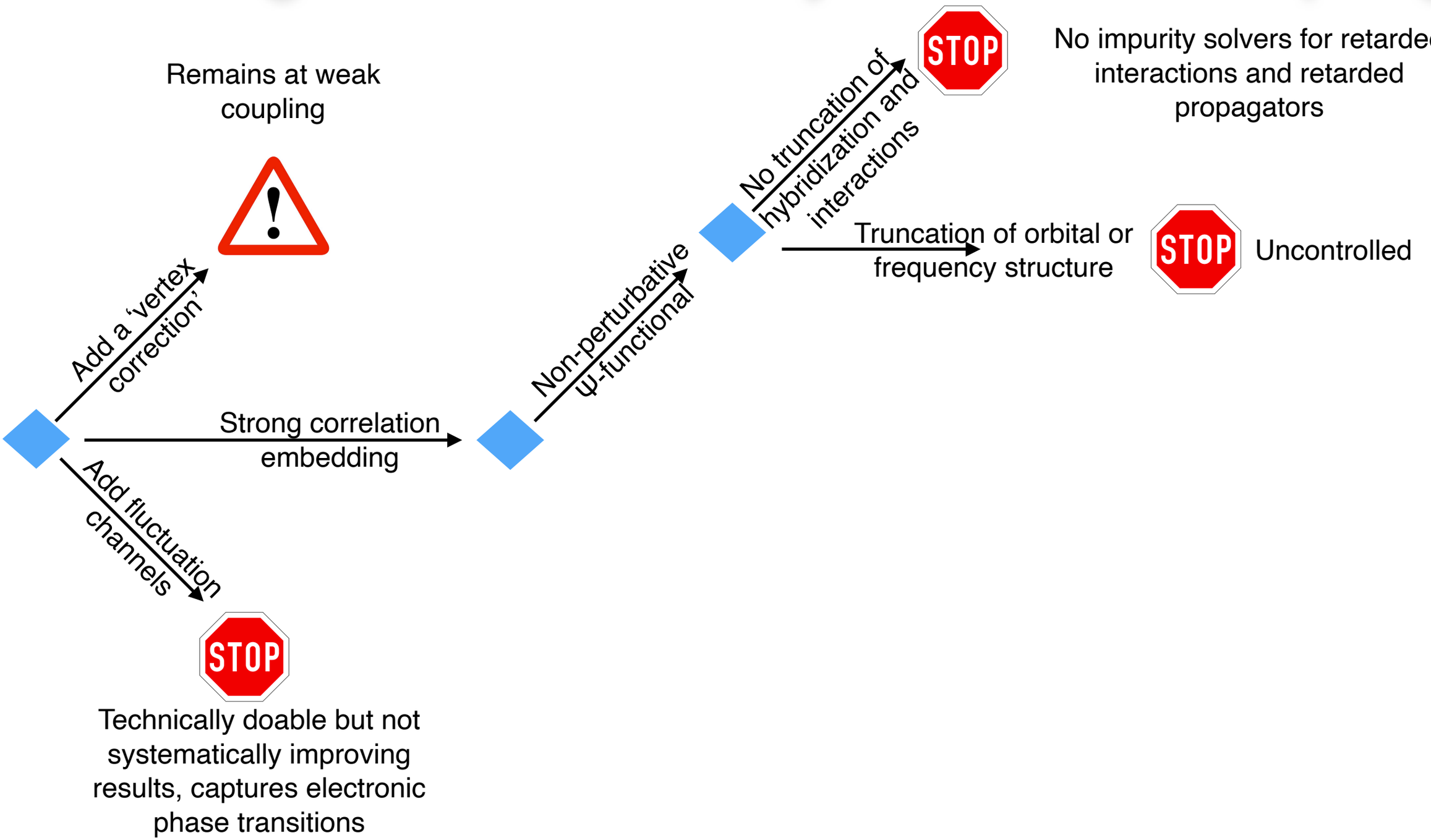

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Truncation of orbital or frequency structure

Uncontrolled

Impurity problems with large energy scales (bare energy and temperature)

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Uncontrolled

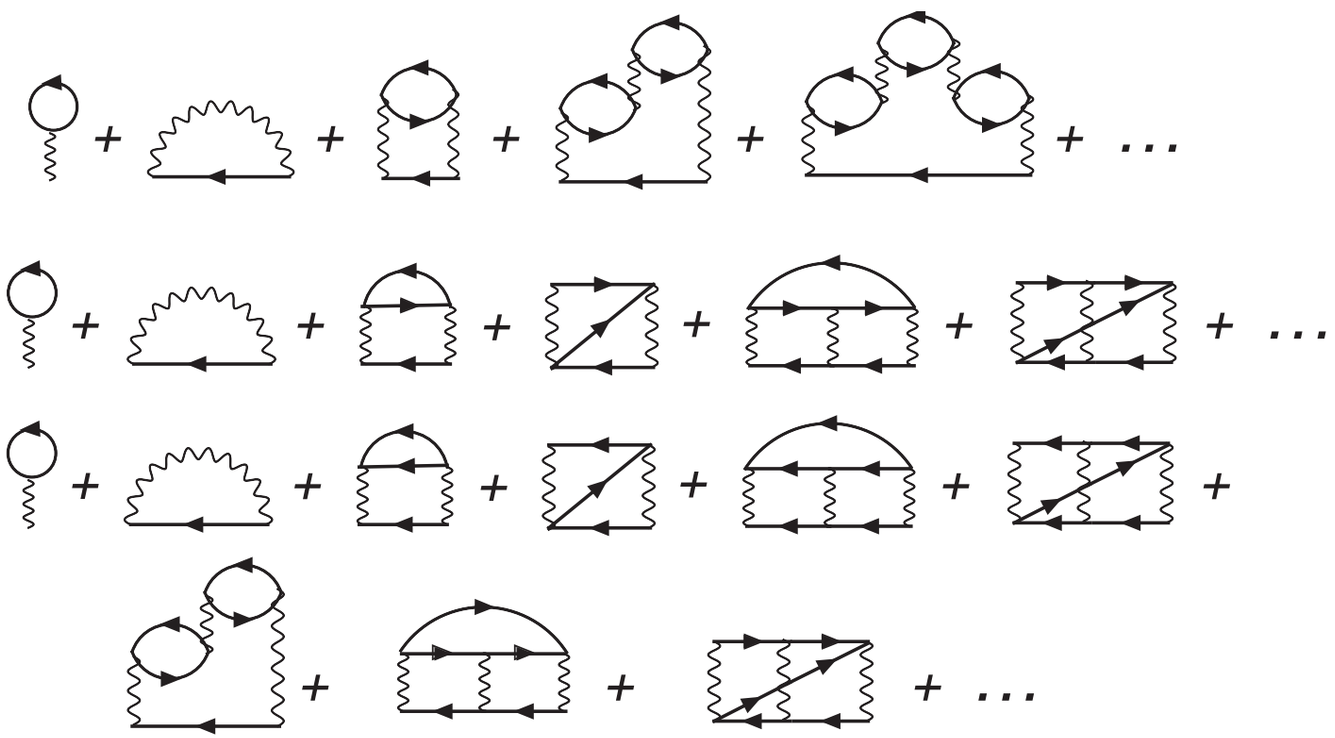
Impurity problems with large energy scales (bare energy and temperature)

Adaptive grids and numerical mathematics

Adding correlations – the fluctuation route

- Largest source of error: lack of correlation from higher-order diagrammatics.
- Add a vertex?
  - Yes, but only in model systems. Generalized susceptibilities have 3 momenta and 3 frequencies, much too large to compute / store!
- How far can one get with diagrammatic expressions dependent on a single frequency?

- Infinite number of diagrams so we can have phase transitions! One frequency implies one geometric series. Obvious candidates: density, magnetic, and superconducting diagram series.



- GW
- T-matrix (magnetic and superconducting ladders)
- All together: Flex

Bickers, Scalapino & White, PRL 62, 961 (1989)

Application to 3d Hubbard

- Simplest model with fermionic phase transition where exact result is known (QMC)
  How well do these methods work? U=6t (outside weak coupling)

Iskakov & Gull, Phys Rev B 105, 045109 (2022)

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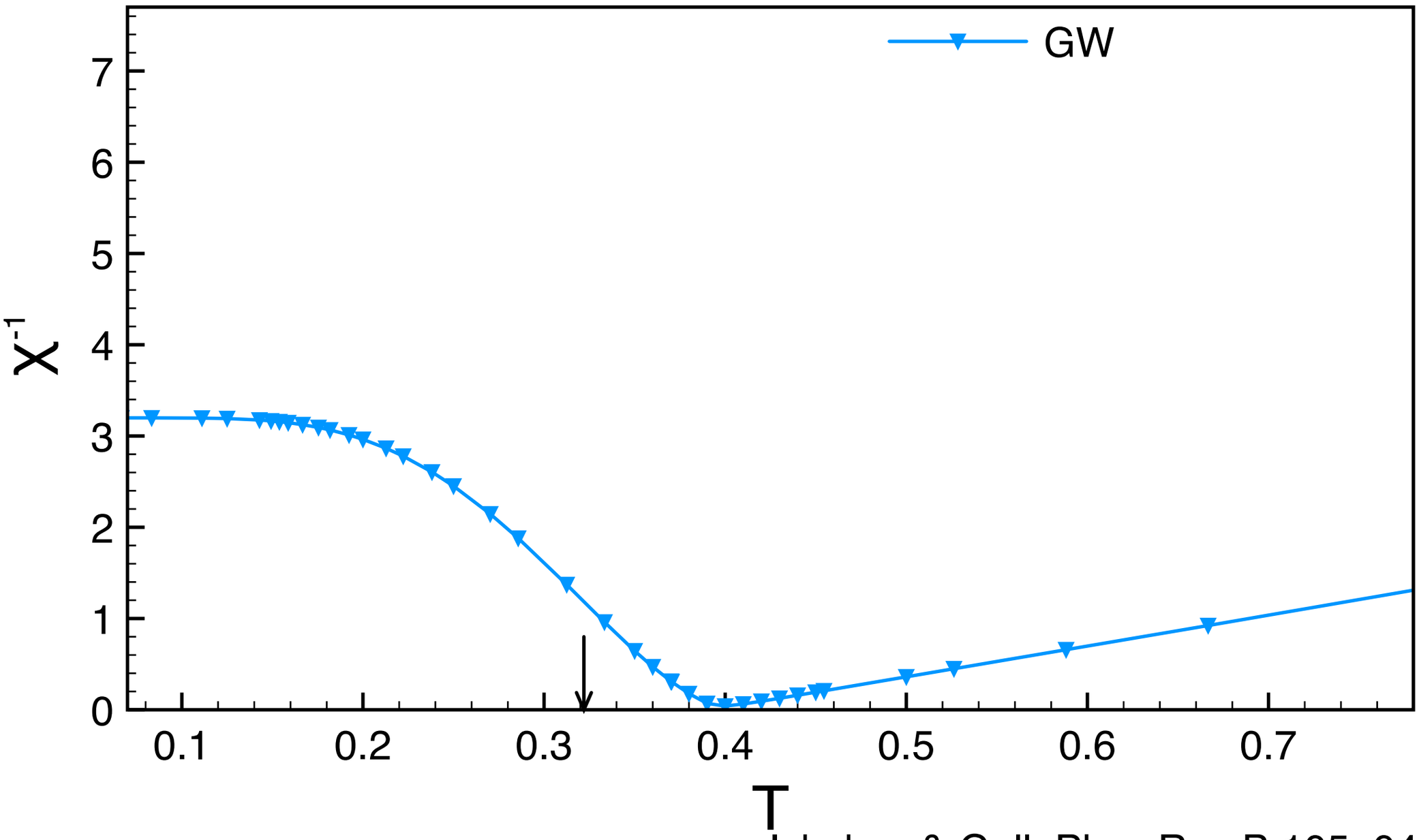

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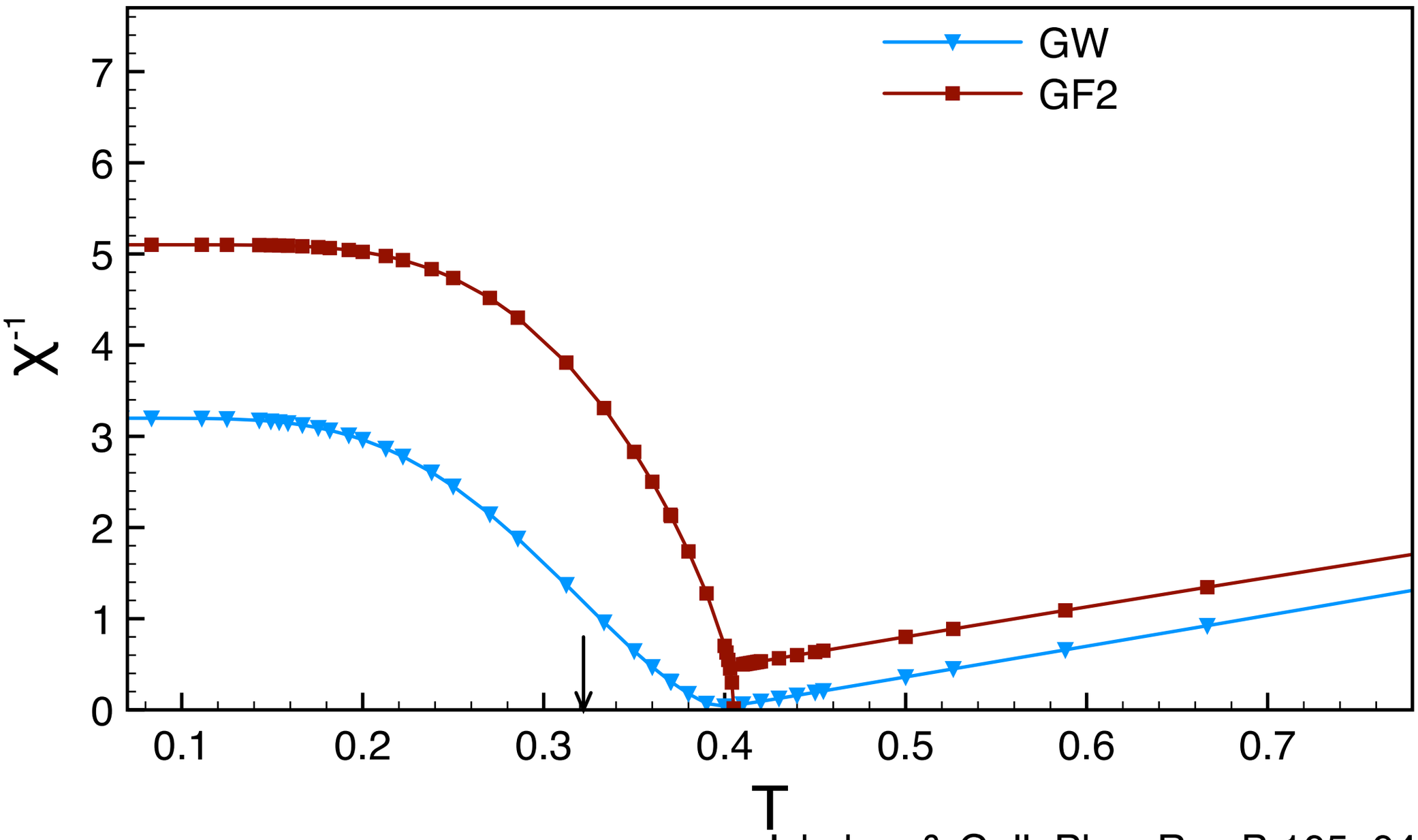

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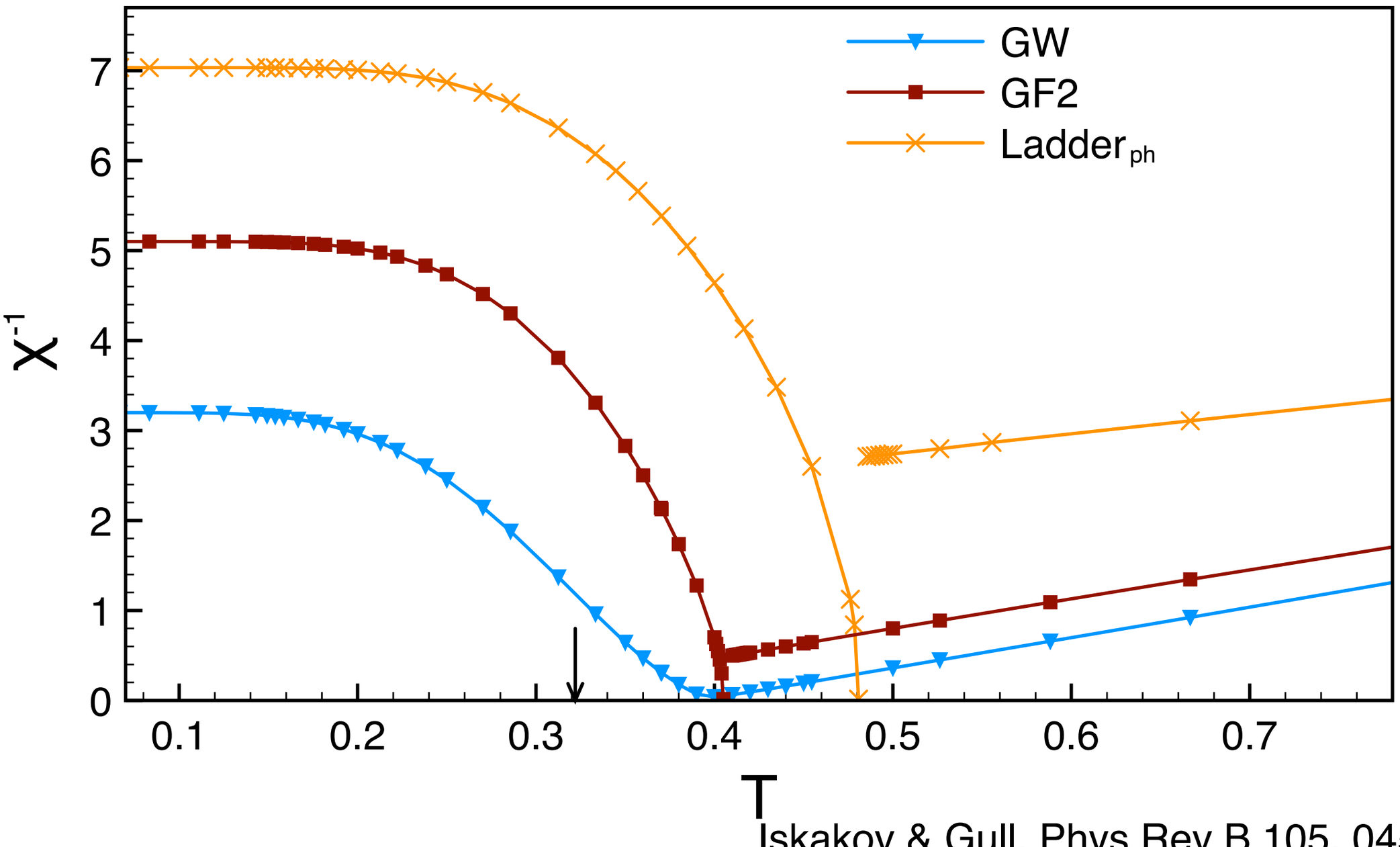

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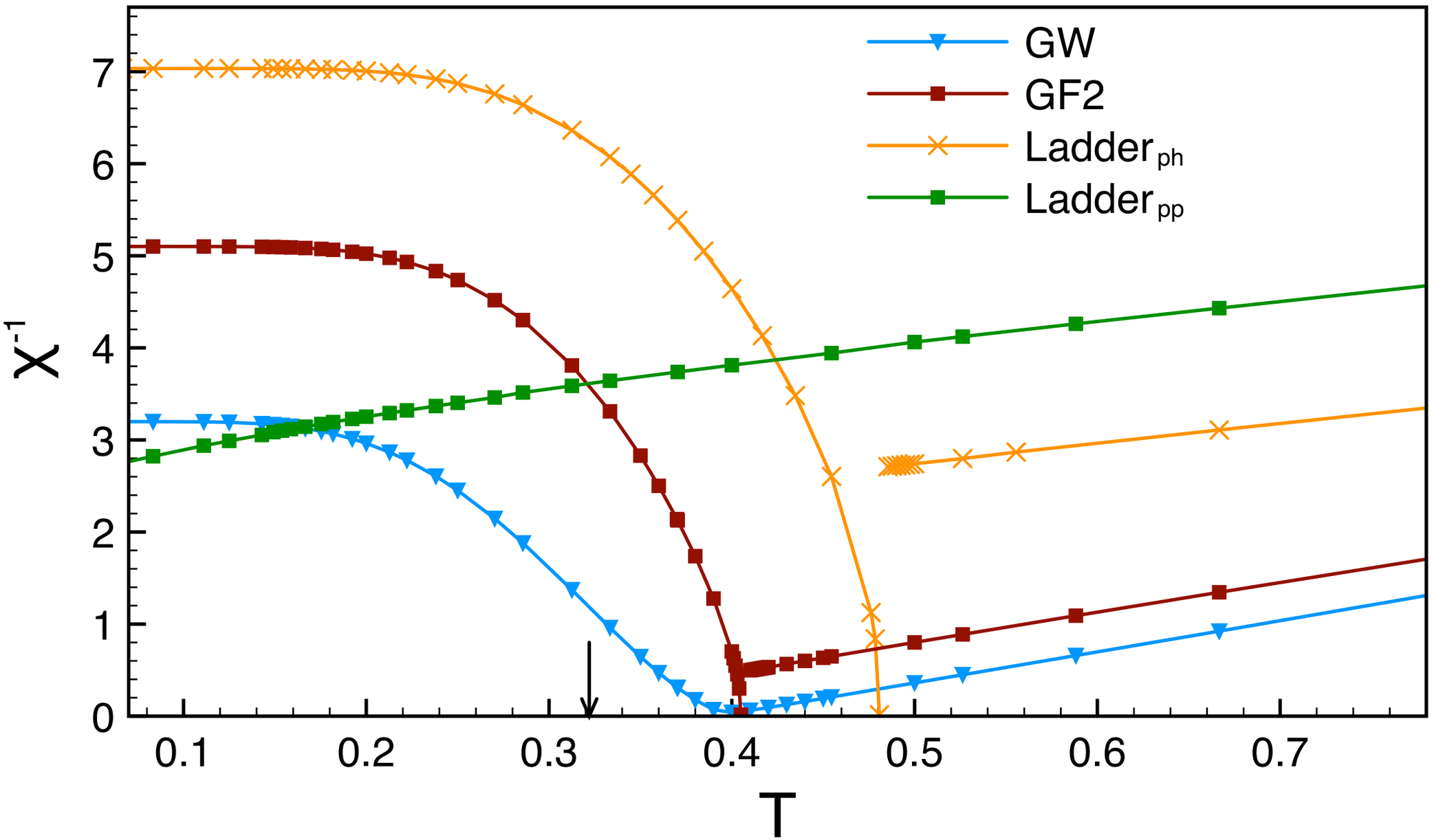

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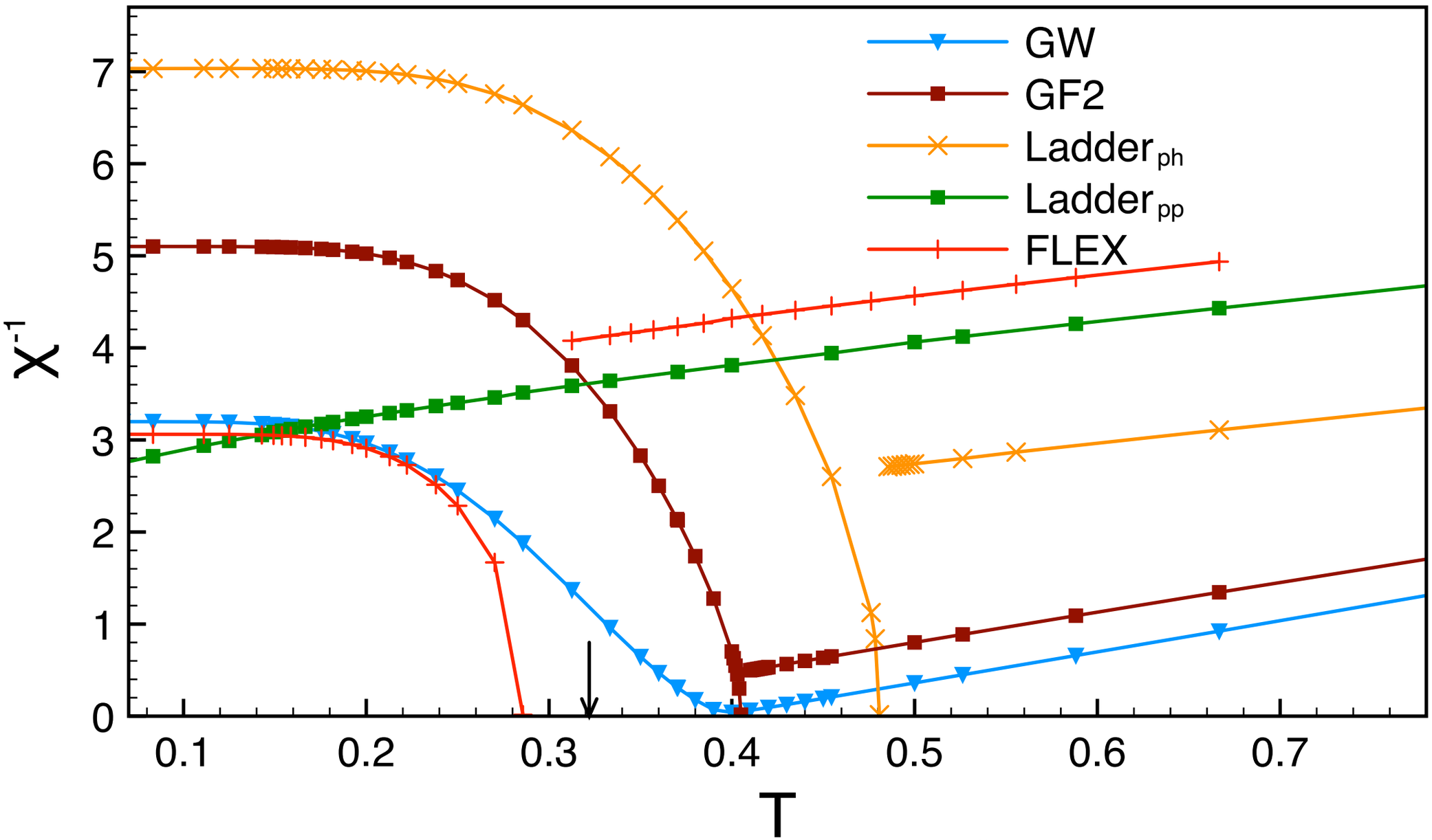
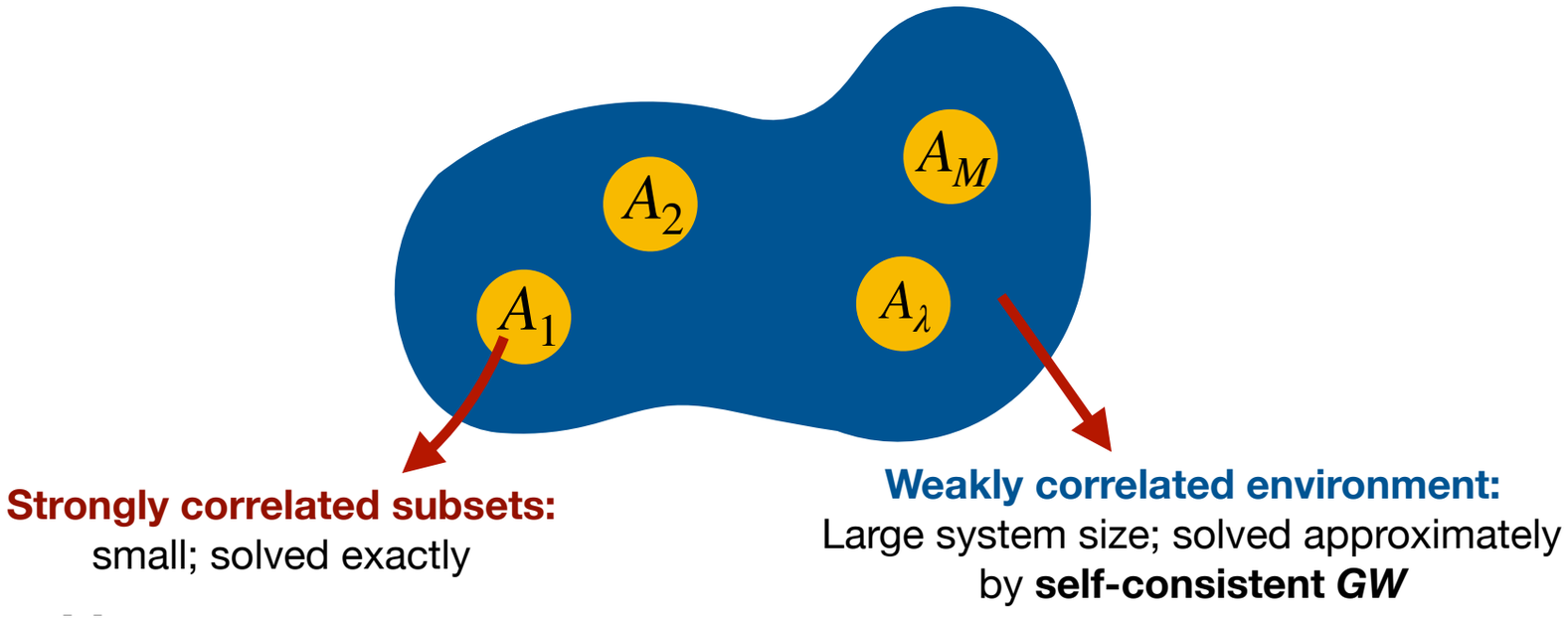


Iskakov & Gull, Phys Rev B 105, 045109 (2022)

Quantum Embedding

- Self-energy embedding theory



- Split system into weakly coupled background and strongly coupled orbitals
- Solve background approximately within weak coupling method
- Solve correlated orbitals exactly with impurity construction
- Use self-consistent diagrammatic theories for feedback between weakly coupled space and strongly coupled spaces

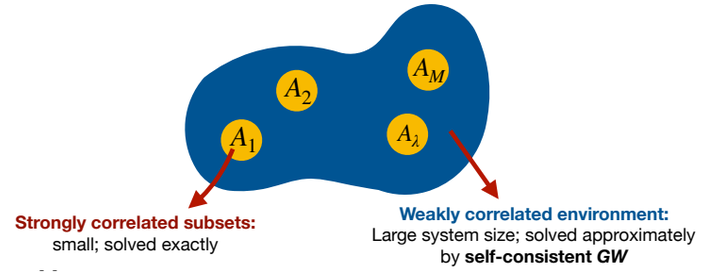
- For models with local interactions and mean field background solution simplifies to weak coupling+dynamical mean field when the correlated subspace is local

Zgid & Gull, New J. Phys. 19, 023047 (2017)

Self-energy embedding theory

- Non-perturbative solution of the correlated subspace: solution of a quantum impurity model. Fast and numerically exact QMC impurity solvers exist.

- Coupling to the environment appears as retardation effects (‘hybridization function’). Bare interactions



- Methodology can be viewed as an approximation to the Luttinger-Ward functional of the crystal, containing all diagrams with all indices in the correlated subspace, as well as all diagrams of the weak coupling method

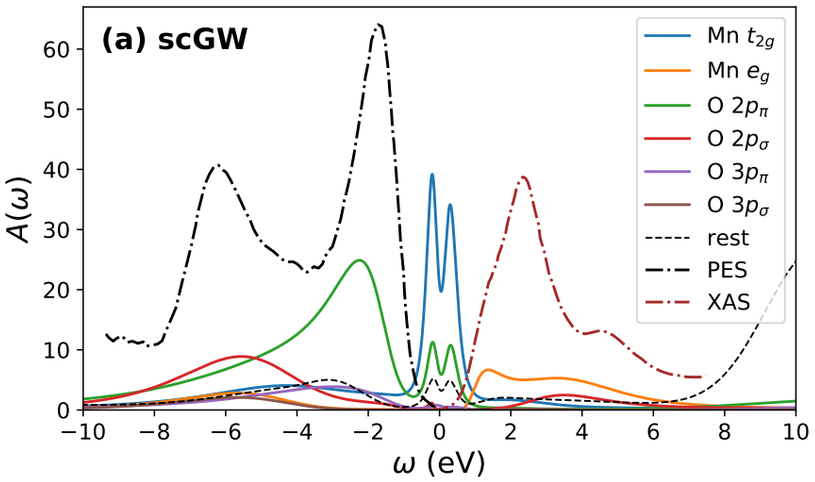
- Approach the exact limit by:
  - Ramping up the number of diagrams in the weak coupling background
  - Enlarging the correlated subspace of the impurity orbitals

- Fully ab-initio: only choice of basis and choice of correlated subspace required as external input

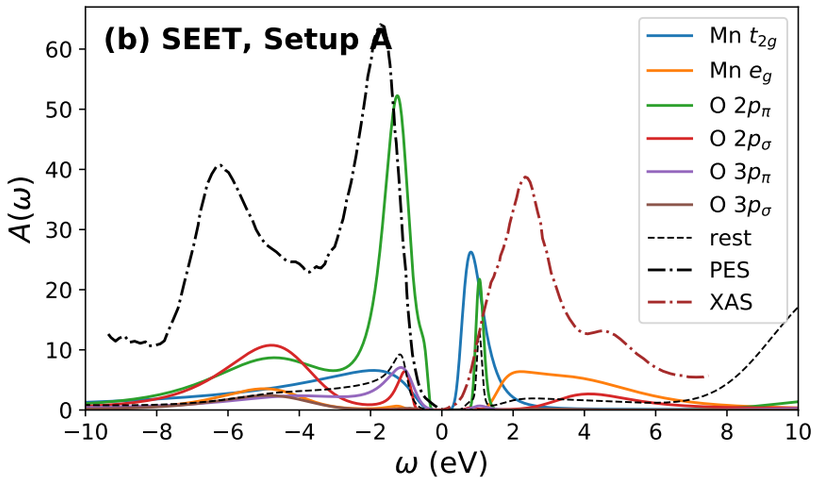
Zgid & Gull, New J. Phys. 19, 023047 (2017)

Example: correlated insulator $SrMnO_3$

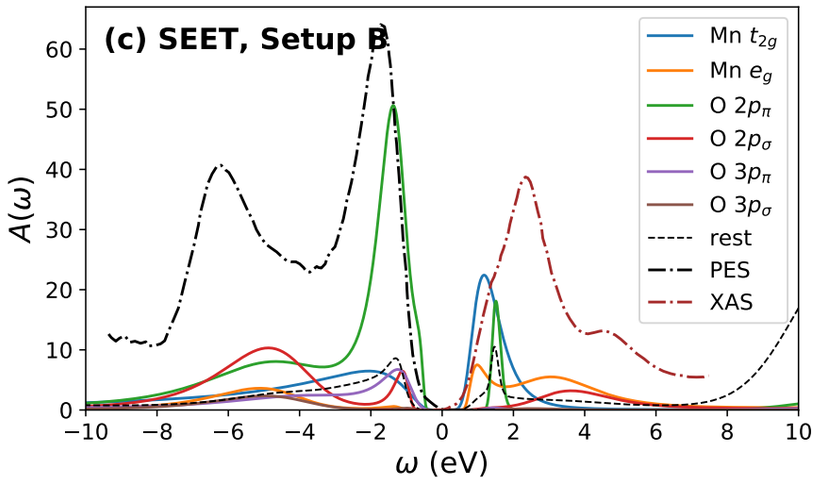
Incorrect Metallic state



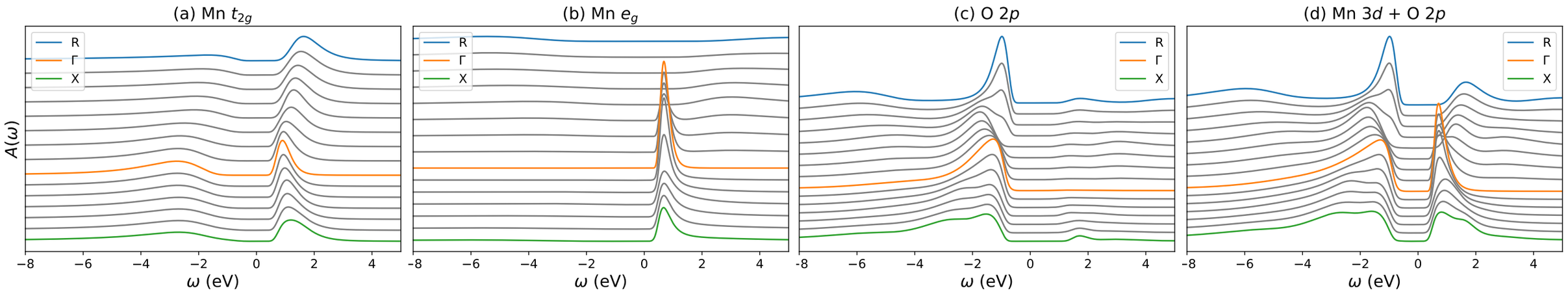
Gap opening due to strong local correlation from Mn t_{2g}



Local correlation from Mn e_g causes qualitative and quantitative changes



charge transfer insulator

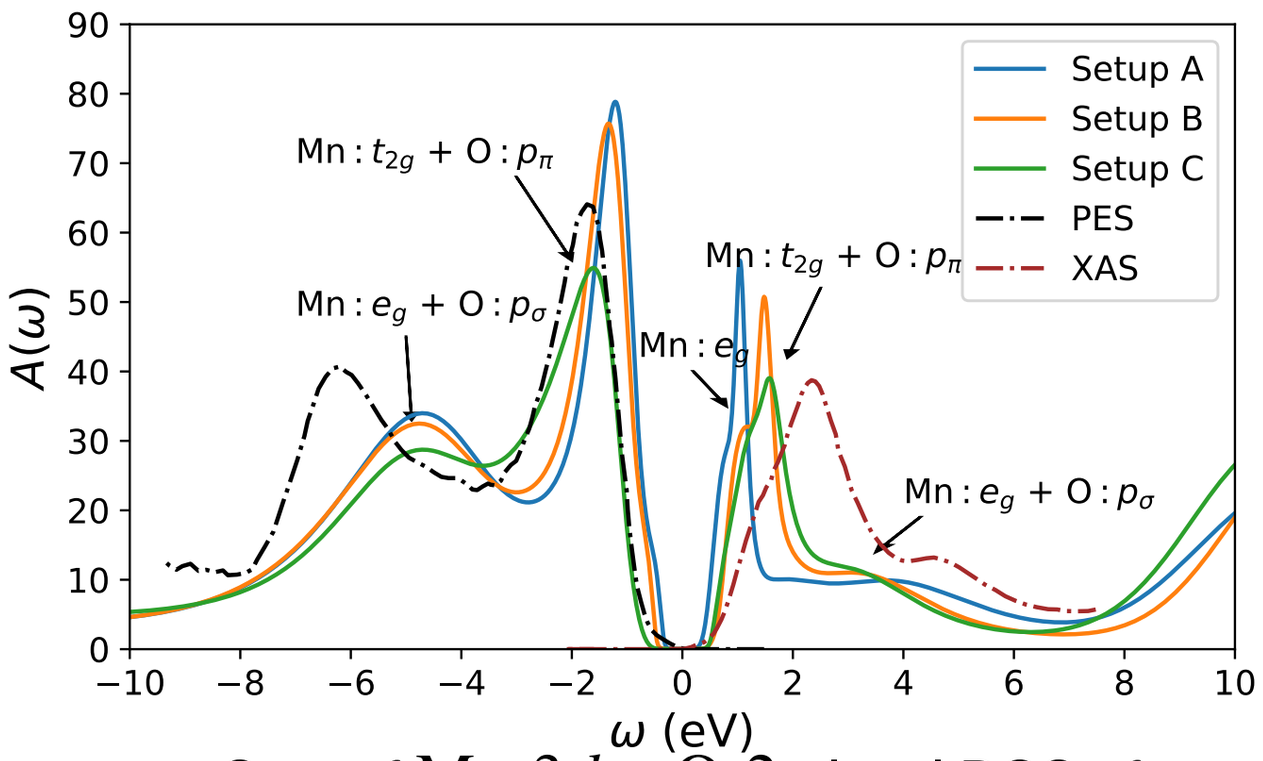


Cubic paramagnetic perovskite. Nominal 3 electrons in Mn 3d

Name	Number of impurity	Descriptions
A	1	Mn t_{2g}
B	2	Mn t_{2g}; Mn e_g
C	4	Mn t_{2g}; Mn e_g; O p_π; O p_σ

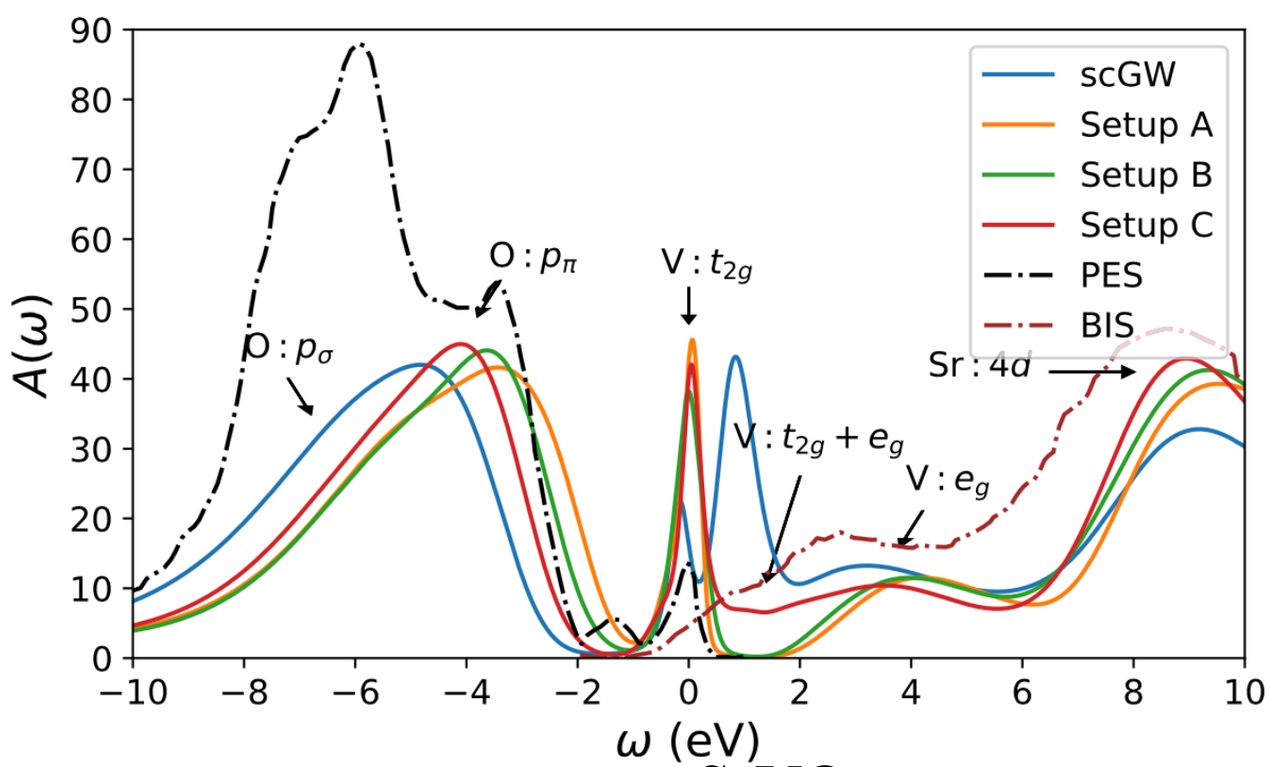
C.N. Yeh, S. Iskakov, D. Zgid and E. Gull, Phys. Rev. B 103, 195149 (2021)

Example: correlated metal $SrVO_3$



Sum of Mn $3d$ + O $2p$ local DOS of $SrMnO_3$ from SEET

Name	Number of impurity	Descriptions
A	1	Mn t_{2g}
B	2	Mn t_{2g}; Mn e_g
C	4	Mn t_{2g}; Mn e_g; O p_π; O p_σ



Total local DOS of $SrVO_3$ from SEET

Name	Imp	Description
A	1	V t_{2g}
B	3	V t_{2g}; O p_π; O p_σ
C	4	V t_{2g}; V e_g; O p_π; O p_σ

Embedding formulated in terms of bare interaction, screening generated by embedding

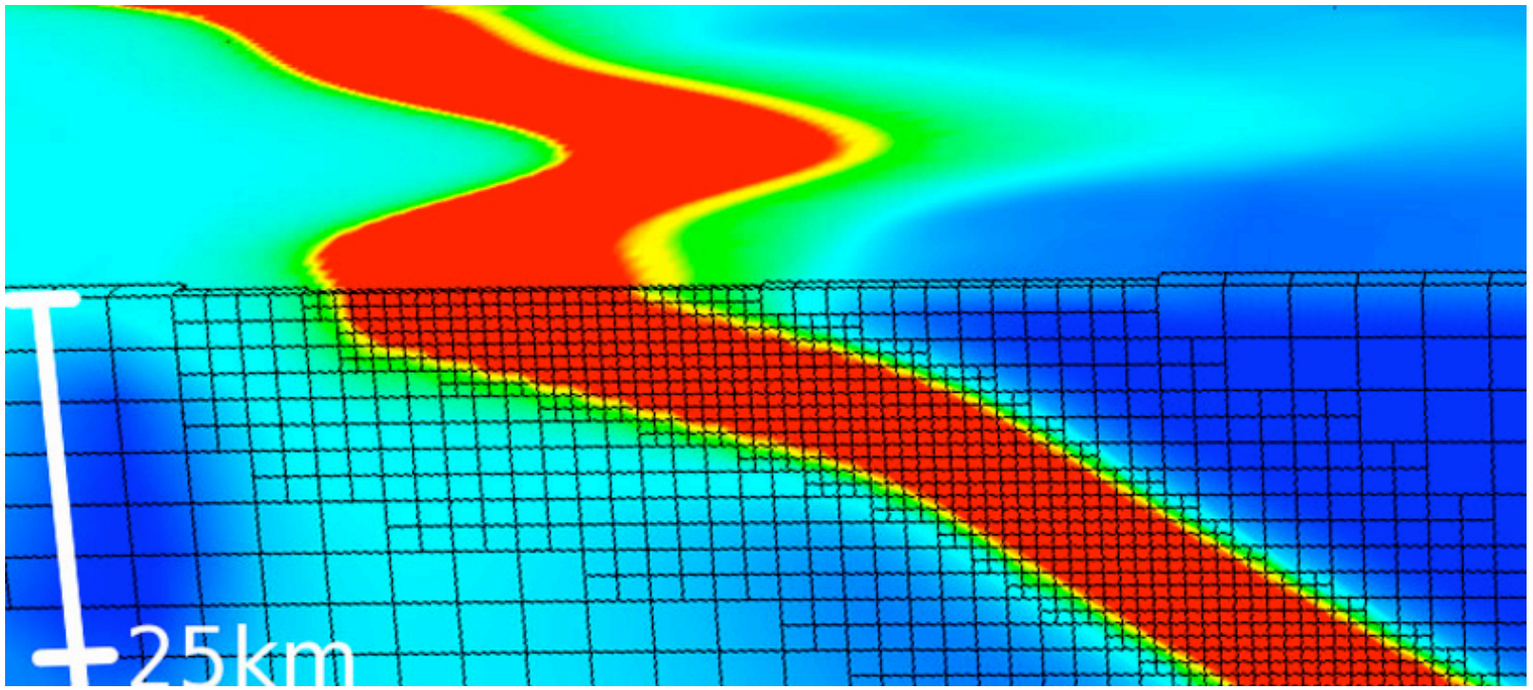
C.N. Yeh, S. Iskakov, D. Zgid and E. Gull, Phys. Rev. B 103, 195149 (2021)

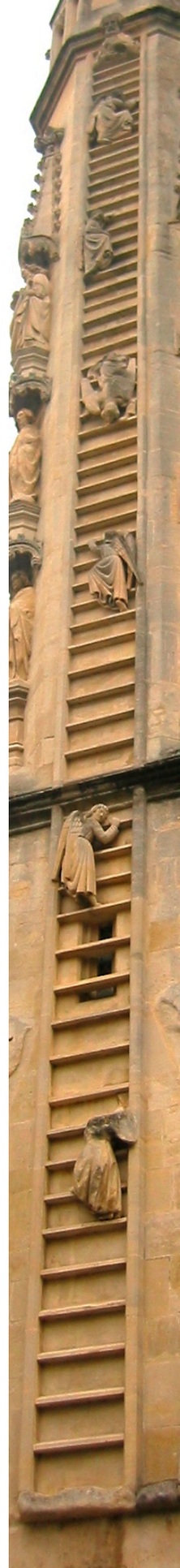
Hierarchy for strong correlations

- Idea/dream of a convergent hierarchy. Every additional step adds complexity but gets closer to the exact results

- Numerical analysis concept: Solutions provide both an a priori and an a posteriori error estimate.

- Paired with an adaptive refinement scheme, solutions can be tuned from cheap-but-fast to expensive-but-accurate. Prime example: FEM





Angels climbing
Jacob's ladder
Bath Abbey, UK

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- Our hierarchies?

System size

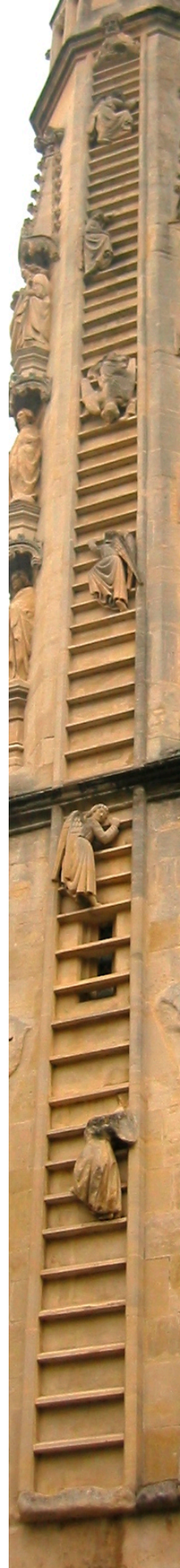
Diagram types

Basis set

Diagram order

Entanglement

Embedding subspace



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Where do we stand?

- Solid foundation to perform many-body theory *ab initio,* without adjustable parameters, reproducibly and accurately.

- Leading error is the weak coupling approximation. All other approximations (finite size, basis set, DFT/starting-point dependence) eliminated or under control.

- We can now do ab-initio relativity & excitations!

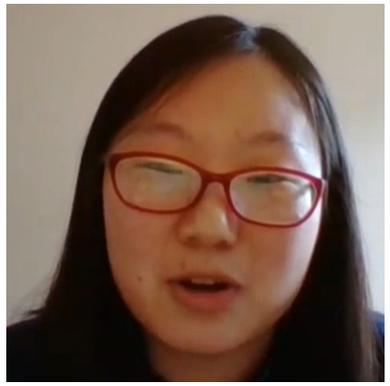
- Increasing diagram order is not a way forward, even though it is technically doable.

- Embedding works very well, both in models and in realistic systems. Technically expensive, toolkit improves rapidly. Provides non-perturbative component.

- Experimental connections at our fingertips

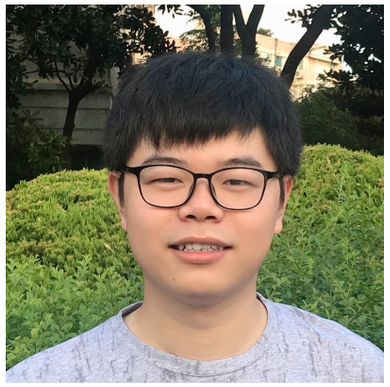
- Still a long way to go to routine parameter-free ab-initio calculations for strong correlations!

Group Members



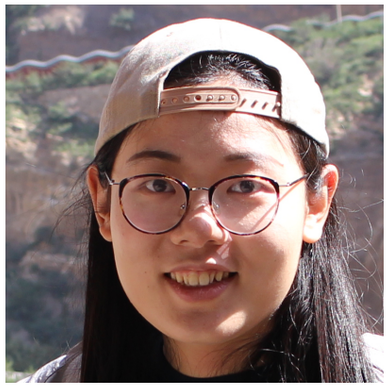
Jiani Fei

Lei Zhang



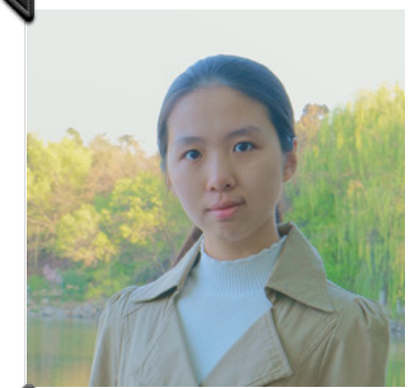
Yang Yu

Thomas Blommel

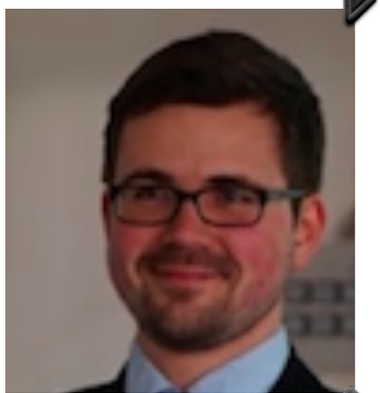


Runxue Yu

Chia-Nan Yeh



Xinyang Dong



André Erpenbeck

Sergei Iskakov

Close collaborators: group of D. Zgid