Sparse sampling approach to efficient *ab initio* and many-body calculations at finite temperature

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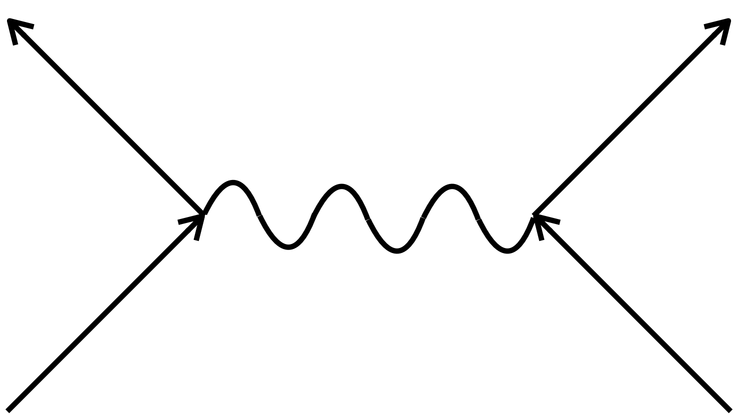
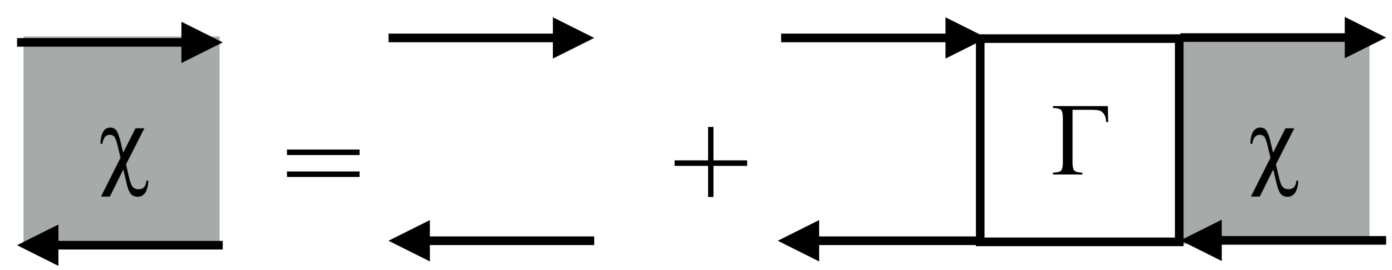
J-Physics：多極子伝導系の物理
J-Physics : Physics of Conductive Multipole Systems 領域番号：2704

Green's function in physics

Building block of perturbative methods, quantum Monte Carlo, response calculations

GW, RPA, FLEX, Migdal-Eliashberg theory, quantum embedding theories

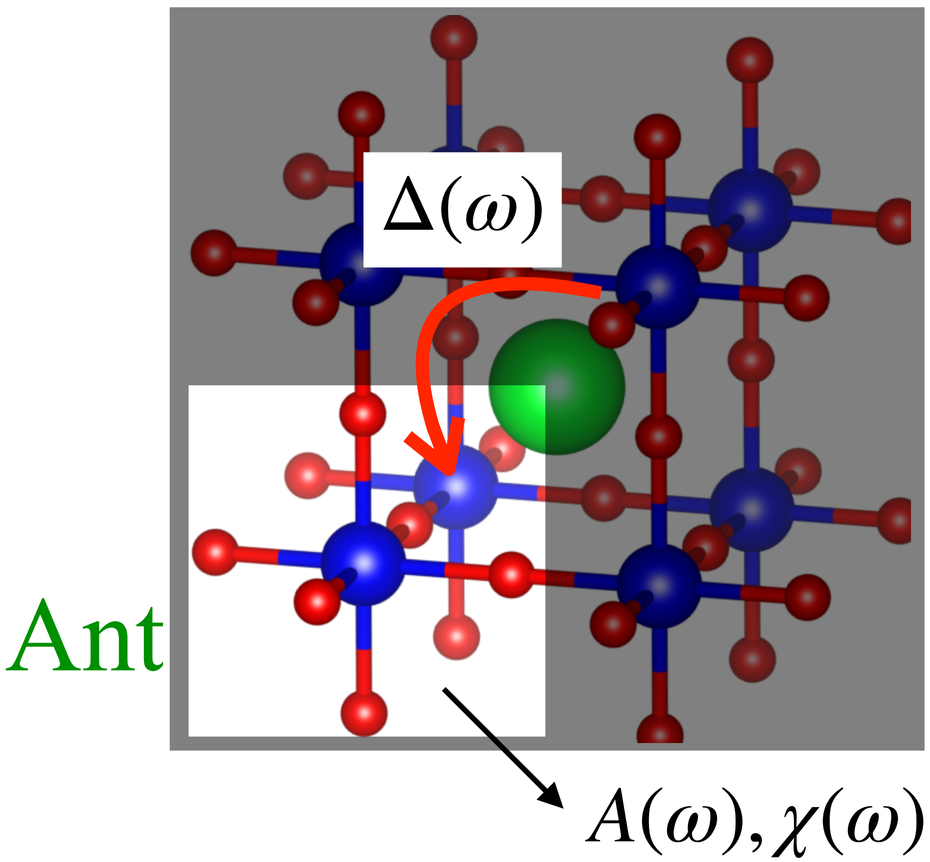




Quantum embedding theories

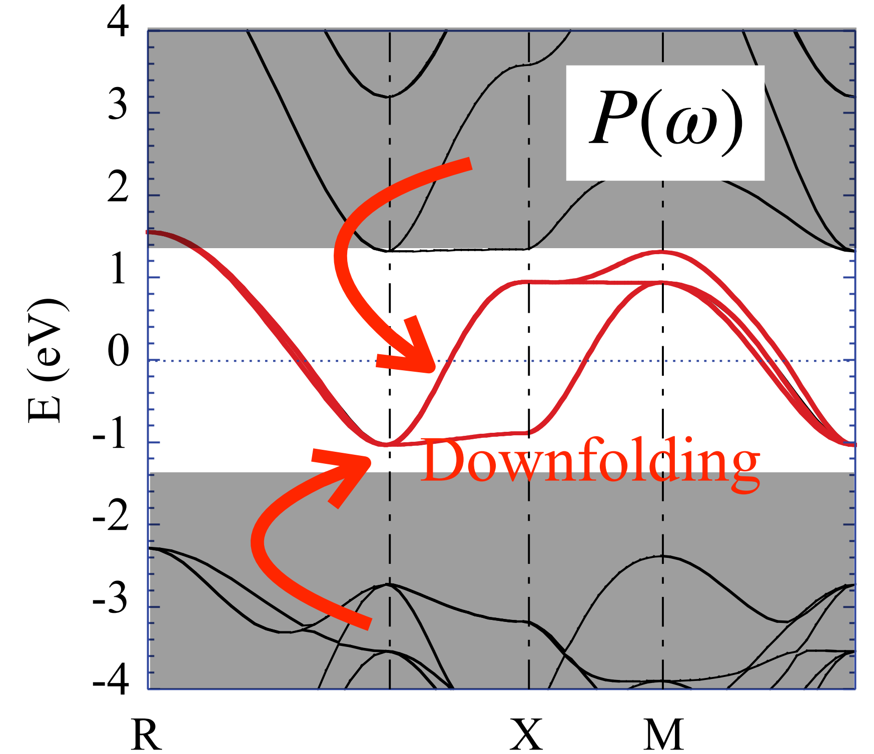
Accurate treatment of strong correlation in active space

Embedding in space



Dynamical mean-field theory, dynamical vertex approximation *etc*.

Embedding in energy

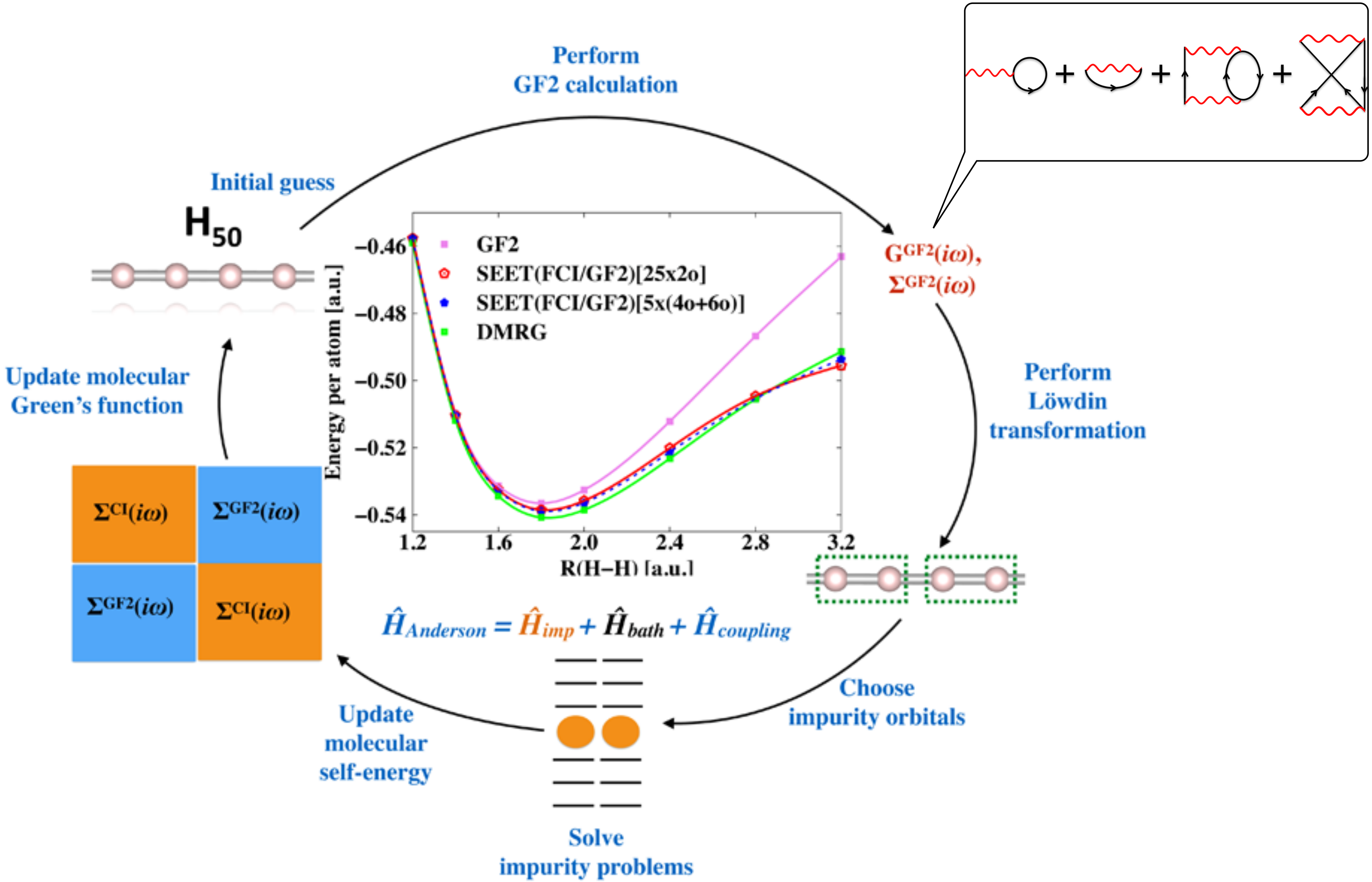


M. Imada and T. Miyake (2010)

Constrained RPA *etc*.

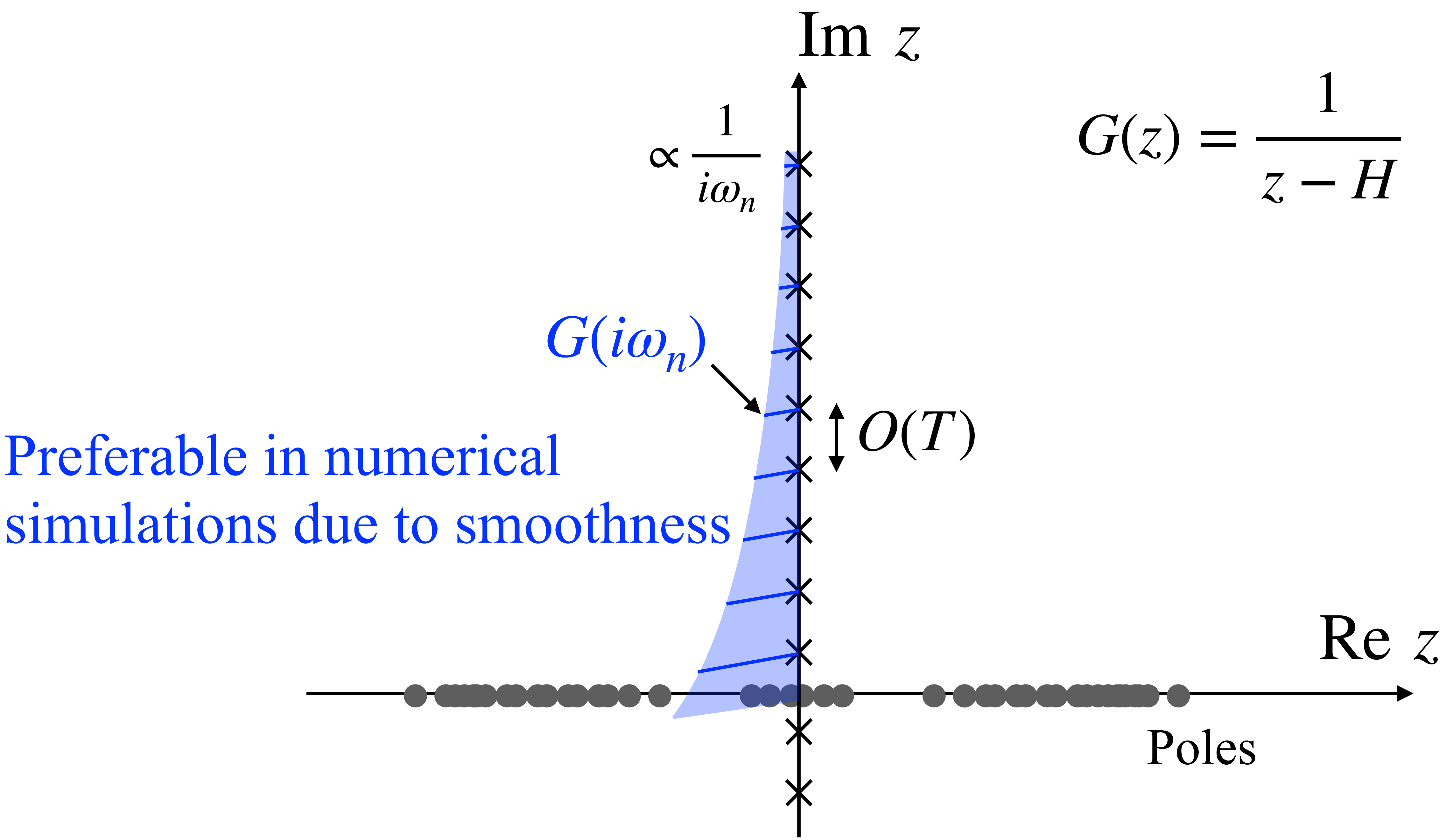
Material calculations require efficient numerical treatment of one- and two-particle response functions.

Quantum embedding for quantum chemistry

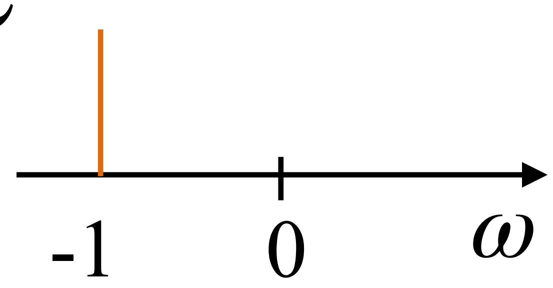


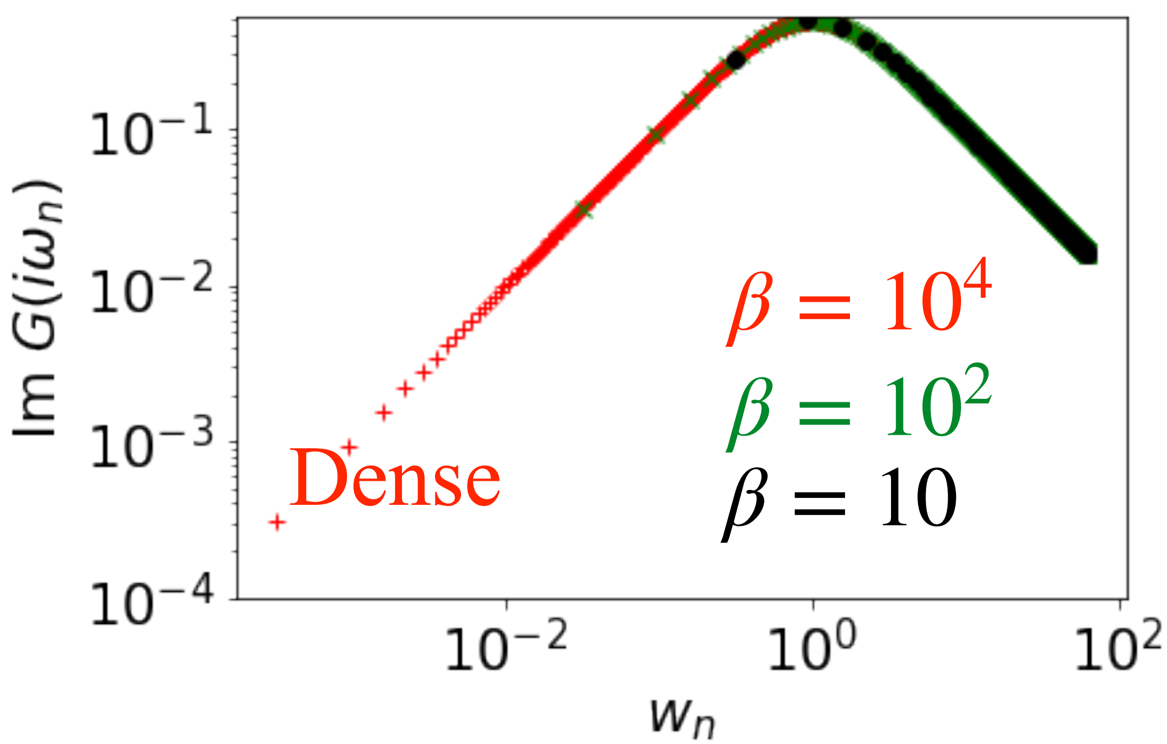
T. N. Lan, A. A. Kananenka and D. Zgid, J. Chem. Theory Comput. **12**, 4856 (2016)

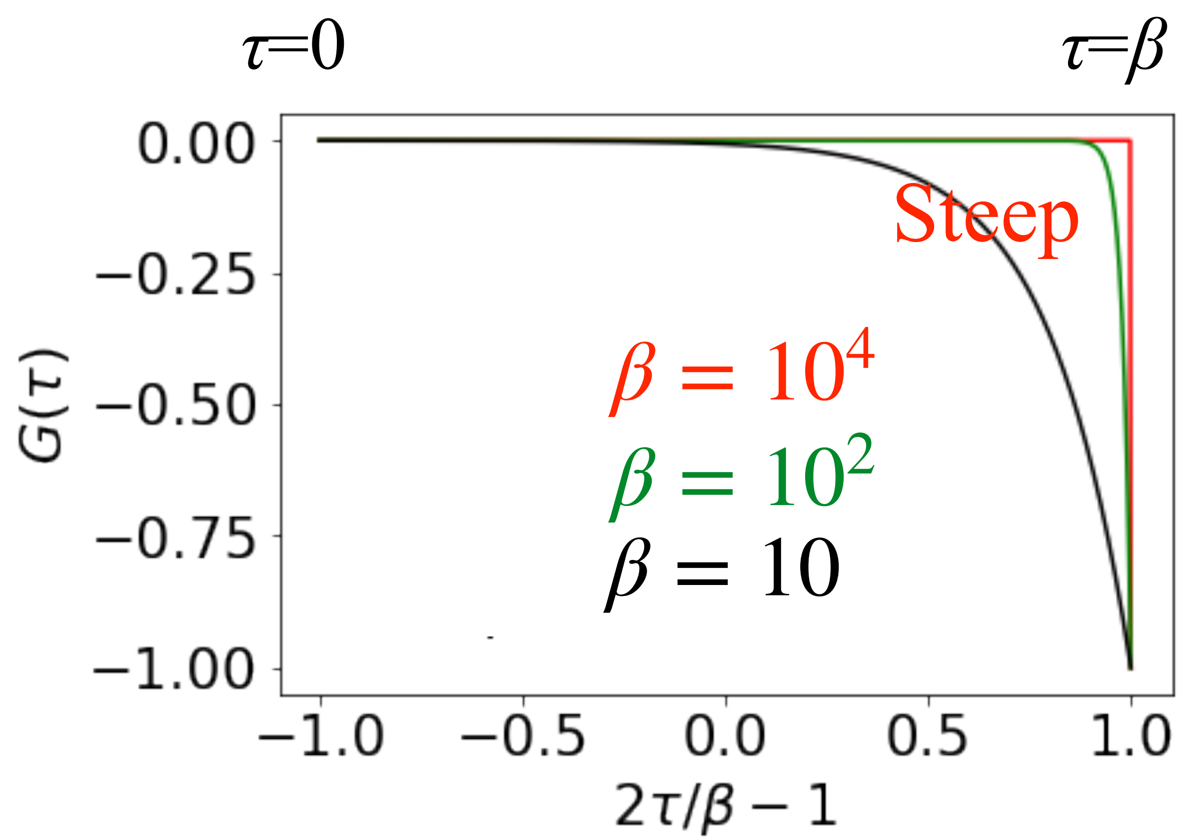
Matsubara Green's function



Example: "core" state







$$G(\tau) = T \sum_n G(i\omega_n) e^{-i\omega_n \tau}$$

Growth of data size and computational time at low T

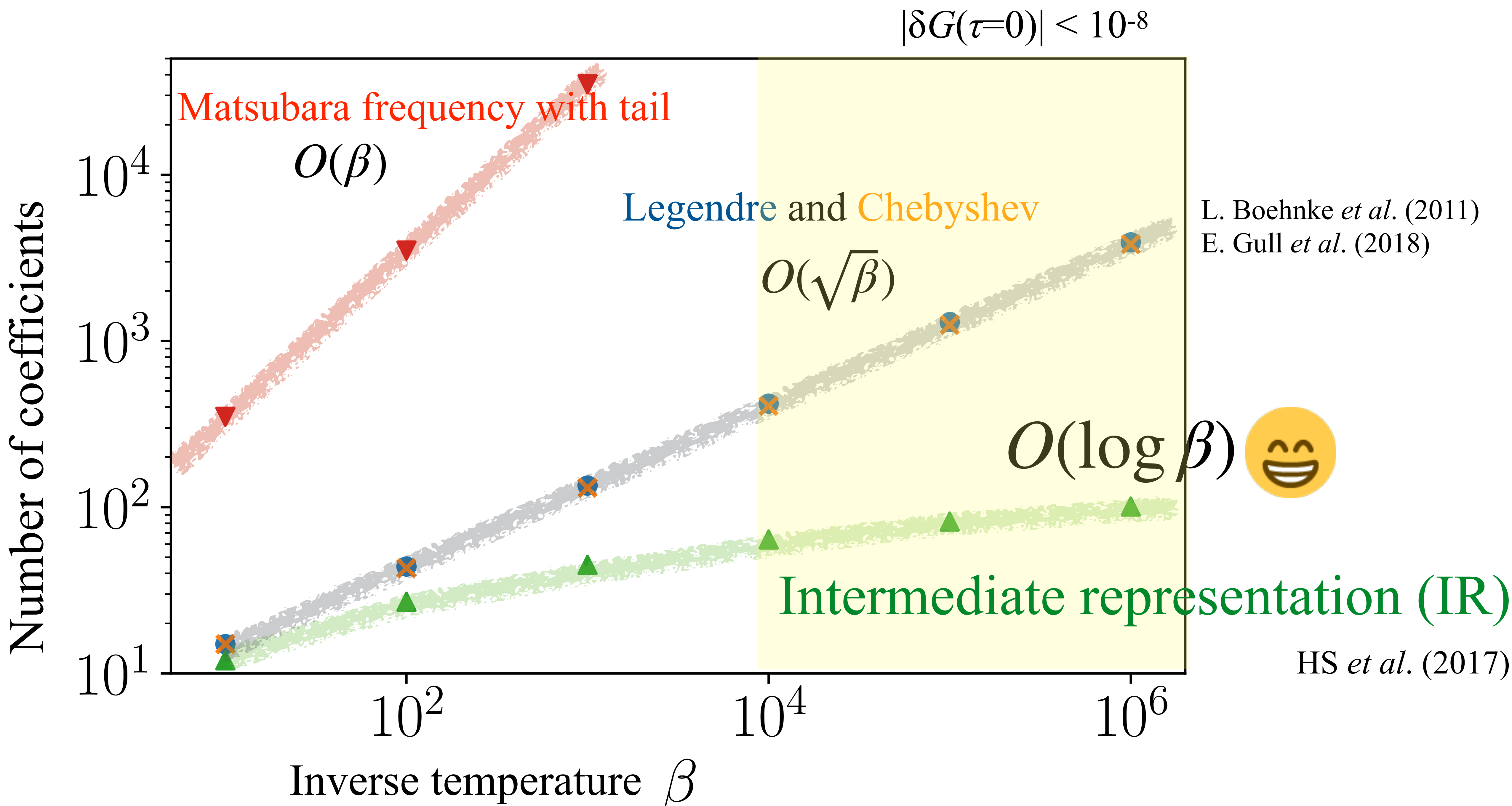
"Prescriptions"

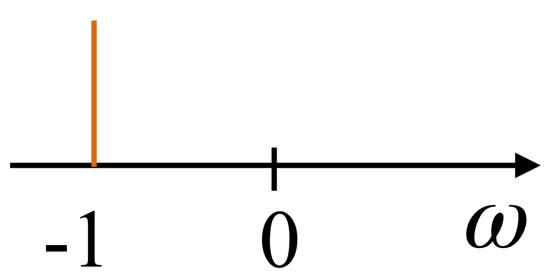
Many tuning parameters, no physical ground…

ω_n: High frequency tail expansion, cubic spline interpolation…

τ: Power mesh, polynomial expansion…

Low-T scaling of data size





$\omega_{\mathrm{band}} = 10$ eV $\simeq 10^5$ K $\rightarrow \omega_{\mathrm{band}}\beta = 10^4$

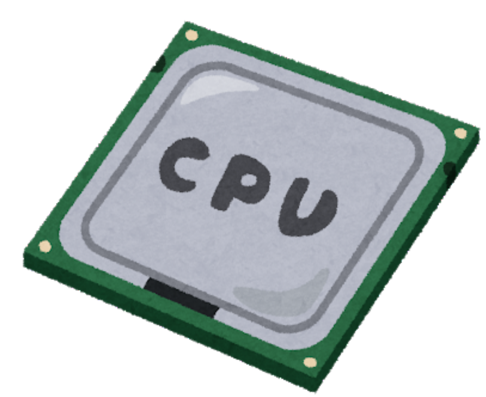
$T = 10$ K

$\hbar = 1$

Motivation

- Storage
  "IR" basis based on physical ground

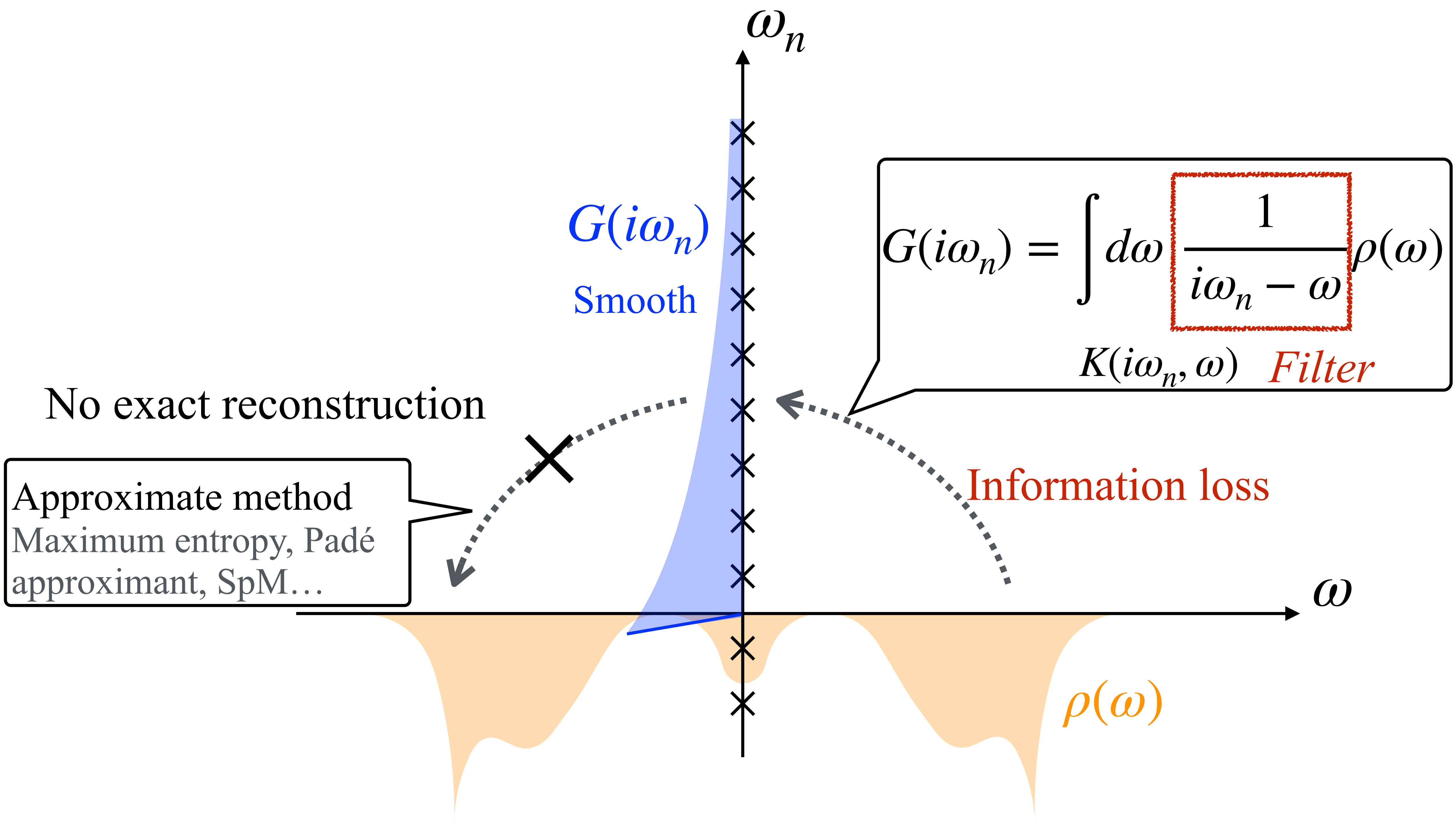
- Computation
  Sparse sampling approach to solving diagrammatic equations (*GW*, Dyson, RPA, FLEX, …)



Outline

- Single-particle Green's function
  - IR basis and sparse sampling
  - Applications to *ab initio* calculations
- Extension to two-particle Green's function

Matsubara Green's function and spectral function



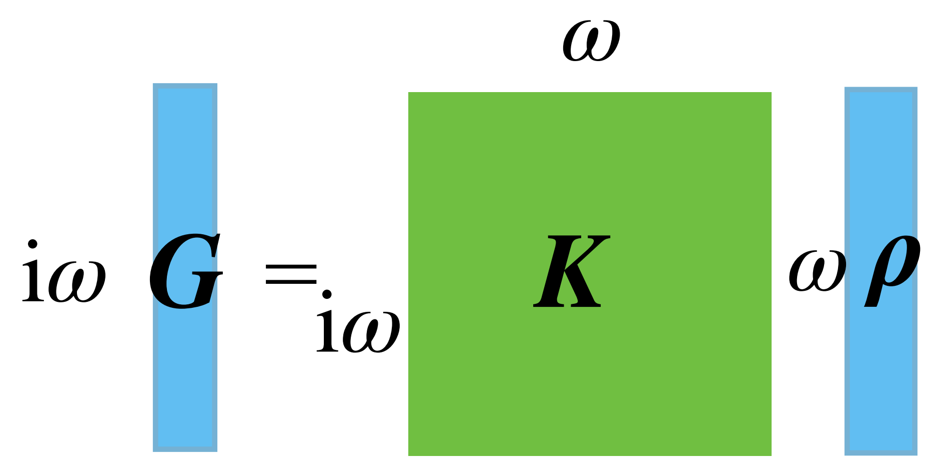
The kernel knows typical structures appearing in $G(i\omega_n)$.

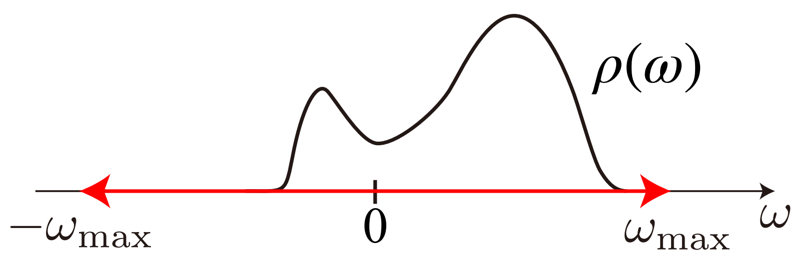
Intermediate representation (IR)

HS, J. Otsuki, M. Ohzeki, K. Yoshimi, PRB **96**, 035147 (2017)
J. Otsuki, M. Ohzeki, HS, K. Yoshimi, PRE **95**, 061302(R) (2017)

$$\hat{G}(\mathrm{i}\omega) = \int_{-\omega_{\max}}^{\omega_{\max}} \mathrm{d}\omega' \, \underbrace{\frac{\omega'^{\delta_{\alpha,\mathrm{B}}}}{\mathrm{i}\omega - \omega'}}_{K^{\alpha}(\mathrm{i}\omega,\omega')} \rho(\omega')$$

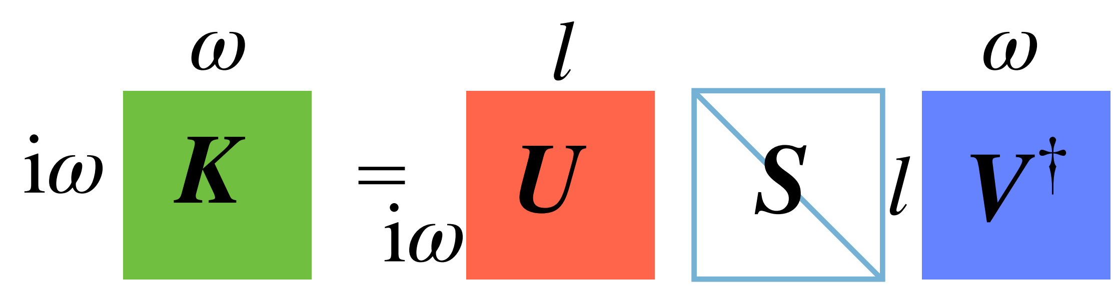
α = F (fermion), B (boson)





Singular value expansion

$$K^{\alpha}(\mathrm{i}\omega, \omega) = \sum_{l=0}^{\infty} S_l^{\alpha} U_l^{\alpha}(\mathrm{i}\omega) V_l^{\alpha}(\omega)$$

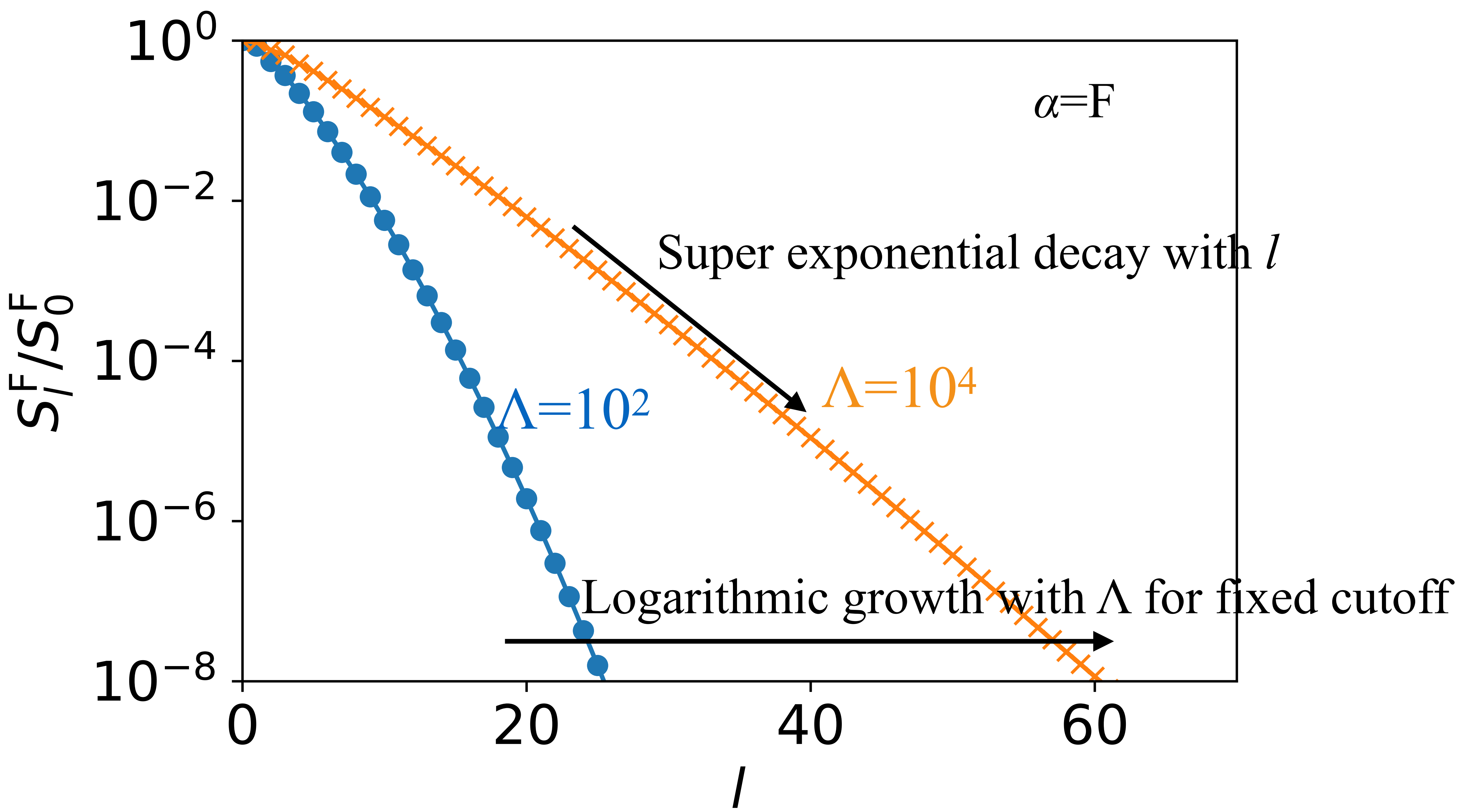


IR basis functions

- Orthonormal basis sets
- Parameterized by $\Lambda \equiv \beta\omega_{\max}$

Singular values

$\Lambda \equiv \beta\omega_{\max}$



Compact representation of Green's function

$$G(\mathrm{i}\omega) = \int_{-\omega_{\max}}^{\omega_{\max}} d\omega \; K^\alpha(\mathrm{i}\omega, \omega)\rho(\omega) \iff G_l = -S_l^\alpha \rho_l$$

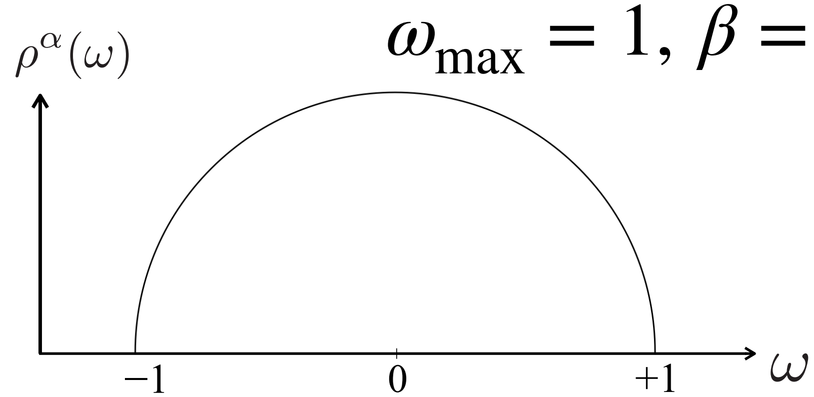
$$G_l \equiv \sum_{\mathrm{i}\omega} U_l^\alpha(\mathrm{i}\omega)^* G(\mathrm{i}\omega)$$

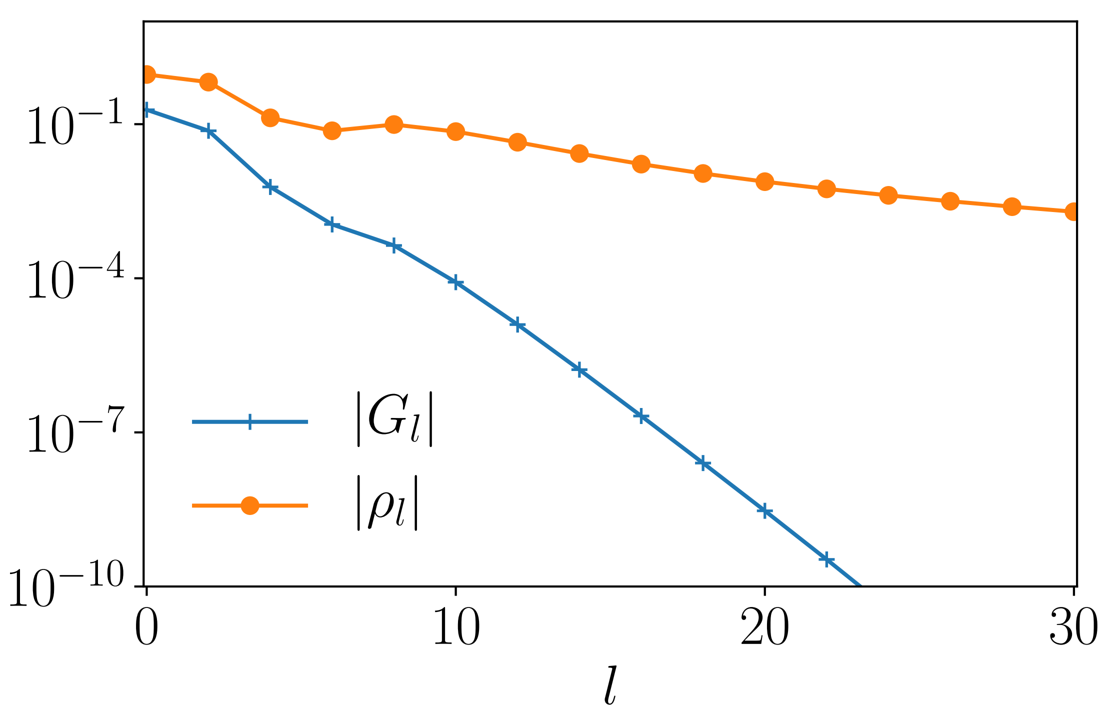
$$\rho_l \equiv \int_{-\omega_{\max}}^{\omega_{\max}} d\omega \rho(\omega) V_l^\alpha(\omega)$$

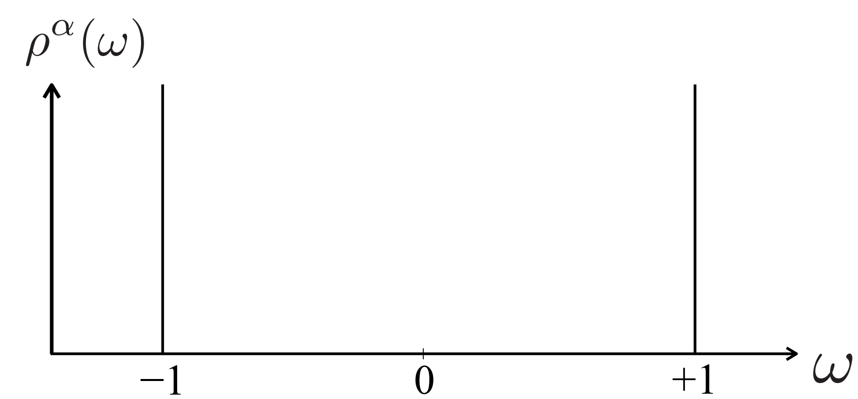
$O(1)$

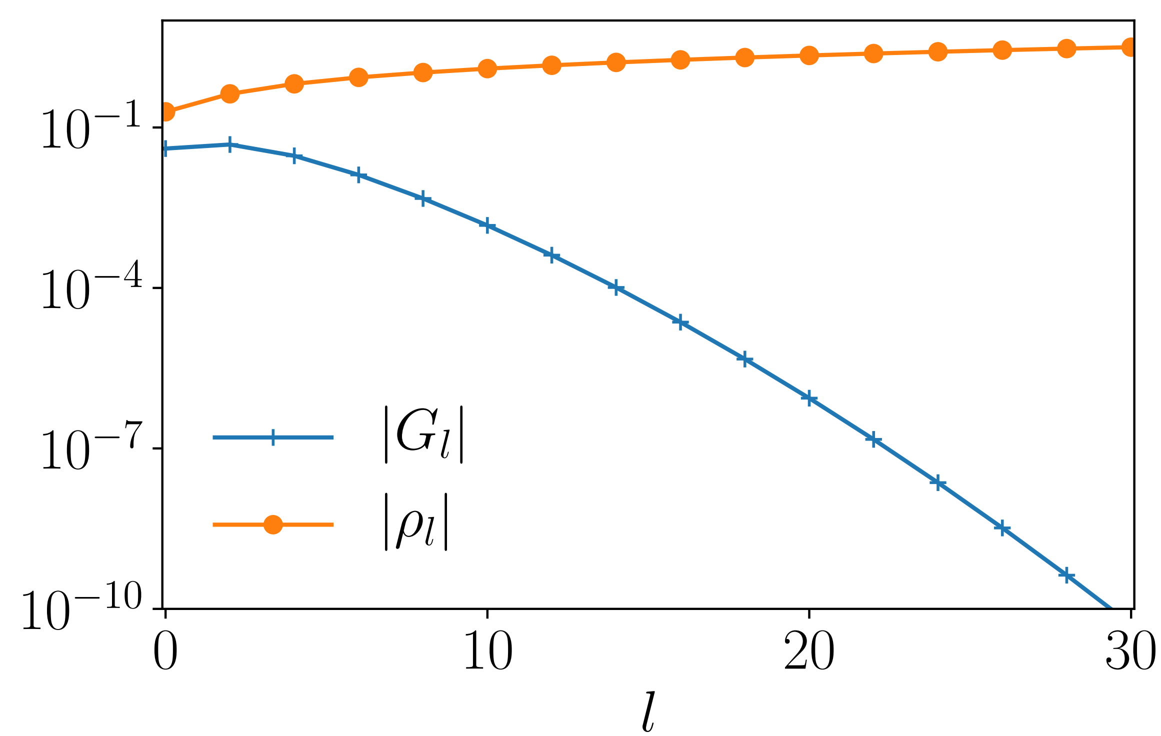
G_l decay as fast as S_l!

$\omega_{\max} = 1,\ \beta = 100$ (fermion)







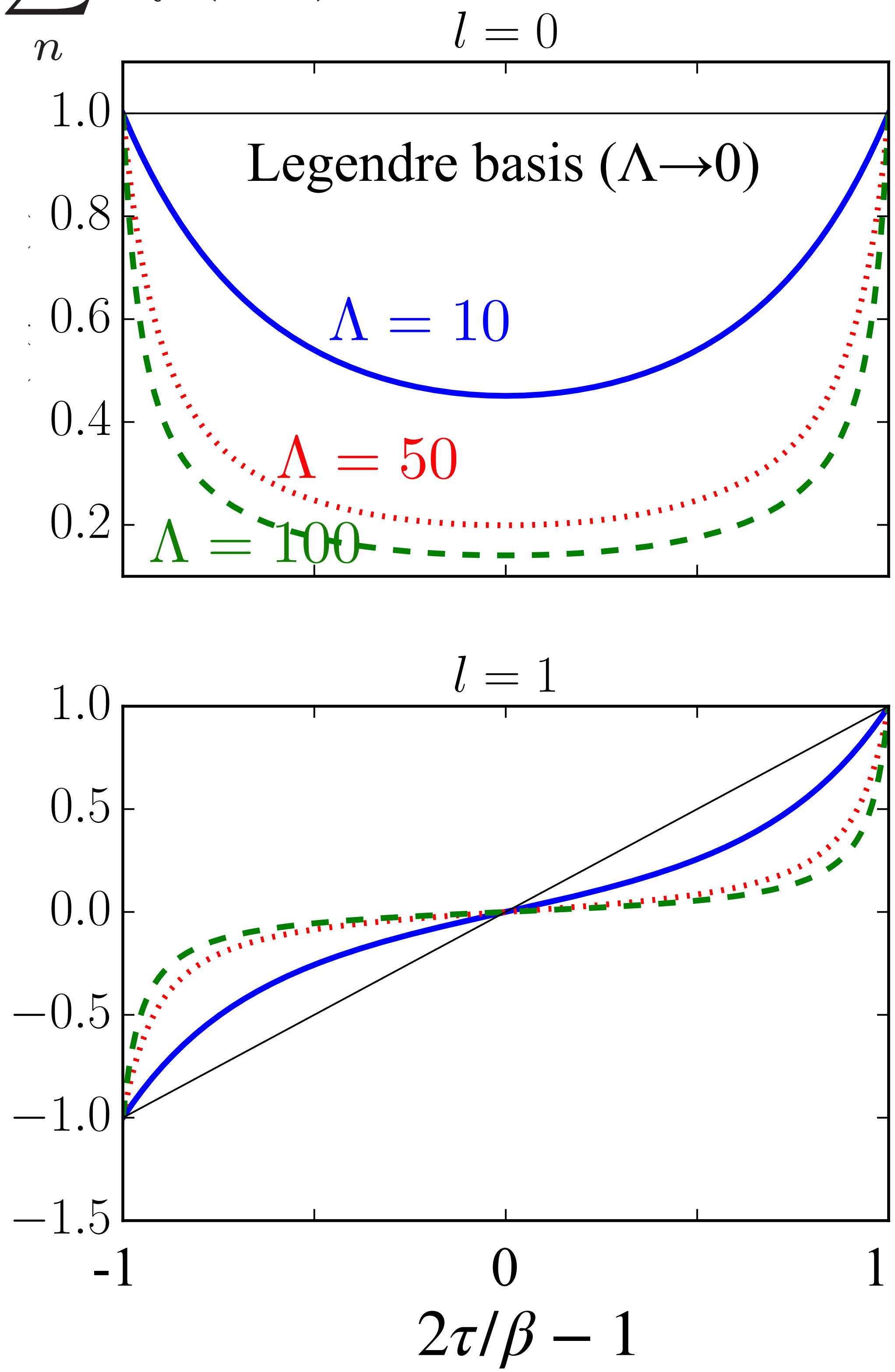


IR basis functions

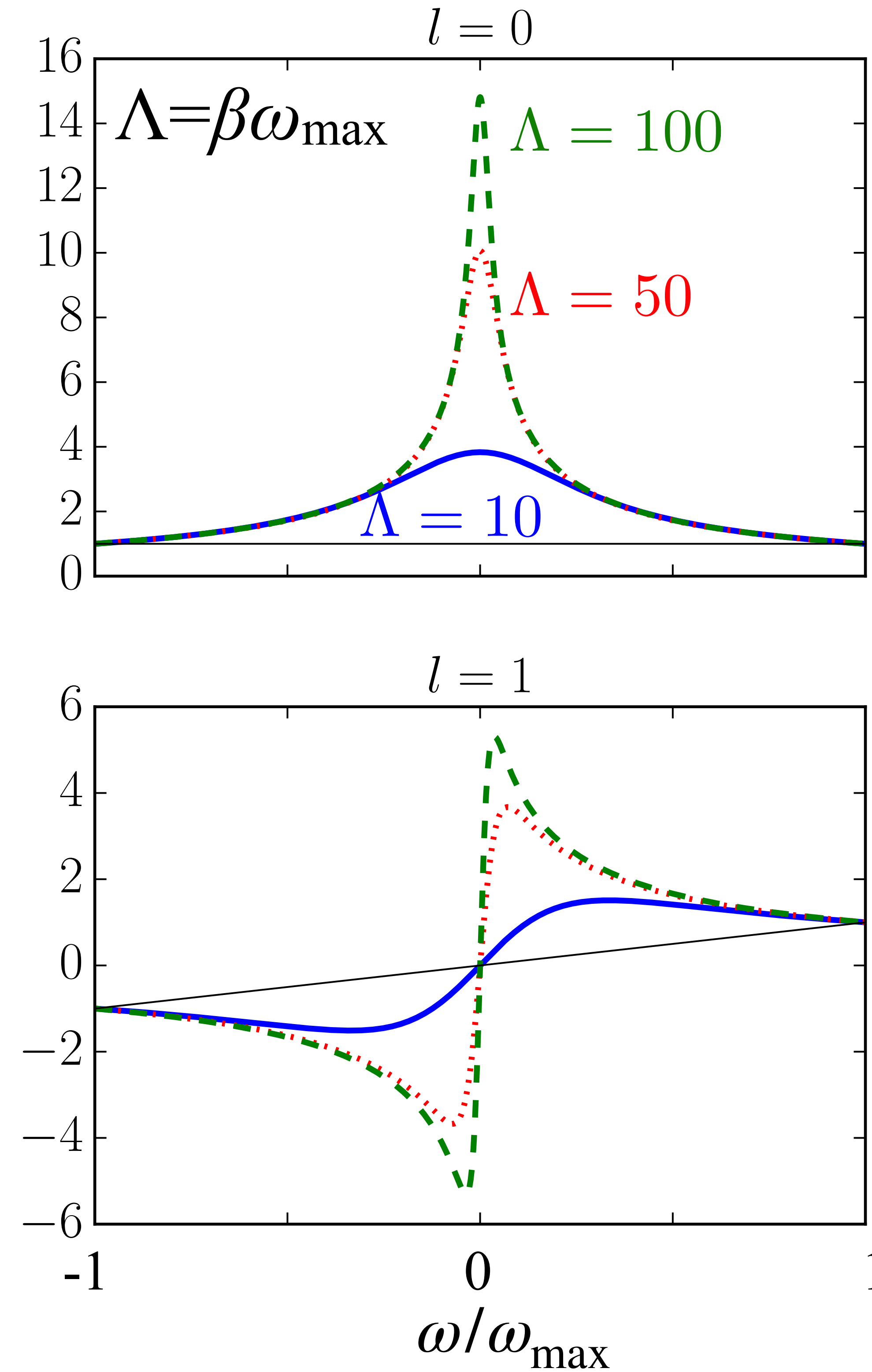
$\Lambda \equiv \beta\omega_{\max}$

$$U_l^{\mathrm{F}}(\tau) \equiv \sum_n U_l^{\mathrm{F}}(\mathrm{i}\omega_n)e^{-\mathrm{i}\omega_n\tau}$$

$$V_l^{\mathrm{F}}(\omega)$$



Fine resolution at $\tau = 0, \beta$



Fine resolution at low ω

Problems to be addressed in Part I

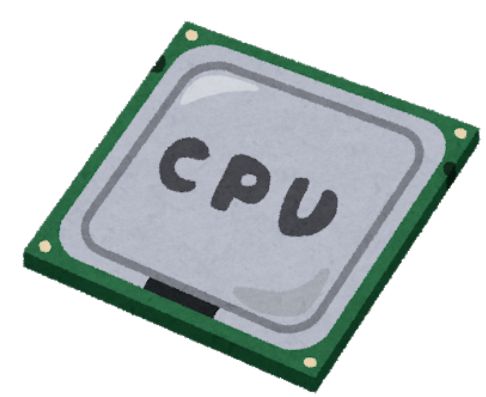
- Storage

  "IR" basis based on physical ground

$$O(\log \beta)$$

- Computation

  Sparse sampling approach to
  diagrammatic equations (*GW*, Dyson, RPA, FLEX, DMFT…)



How to solve diagrammatic equation efficiently?

Dyson equation $$G(i\omega_n) = \frac{1}{i\omega_n - H - \Sigma(i\omega_n)}$$

- Solve linear equation for $G(\mathrm{i}\omega)$

$$A(\mathrm{i}\omega)G(\mathrm{i}\omega_n) = 1$$ diagonal in $\mathrm{i}\omega$

$$A_{\mathrm{i}\omega} \equiv \mathrm{i}\omega_n - H - \Sigma(\mathrm{i}\omega_n)$$

Computational complexity: $O(\beta)$

- Solve linear equation for G_l

$$\sum_{l'=0}^{N} \underline{A_{ll'}} G_l = 1_l$$

$N \times N$ dense matrix ($N \propto \log \beta$)

Computational complexity: $O(\log^3 \beta)$

Asymptotically faster but not super efficient in practical calculation

Trick: sparse sampling

$$\underline{G_l} = \sum_{\mathrm{i}\omega=-\mathrm{i}\infty}^{\mathrm{i}\infty} U_l^{\alpha}(\mathrm{i}\omega)^{*} \underline{G(\mathrm{i}\omega)}$$

What we want to compute eventually

Do we really need to know $G(\mathrm{i}\omega)$ on all frequencies?

No. We do not have to compute this equation because frequency dependence of $G(\mathrm{i}\omega)$ has only N degrees of freedom:

$$G(\mathrm{i}\omega) = \sum_{l=0}^{N} G_l U_l^{\alpha}(\mathrm{i}\omega) + \epsilon(N)\,.$$

$\epsilon(N)$: exponentially small error

Thus, we can determine G_l from $G(\mathrm{i}\omega_n)$ only on appropriately selected *N sampling frequencies*.

Similar idea of sampling frequencies: T. Ozaki, PRB **75**, 035123 (2007), M. Kaltak and G. Kresse, PRB **101**, 205145 (2020) and so on.

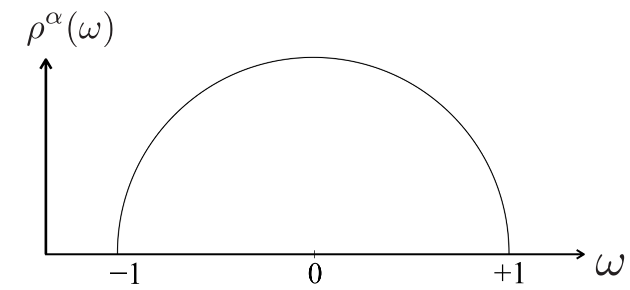
Numerical demonstration

J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)

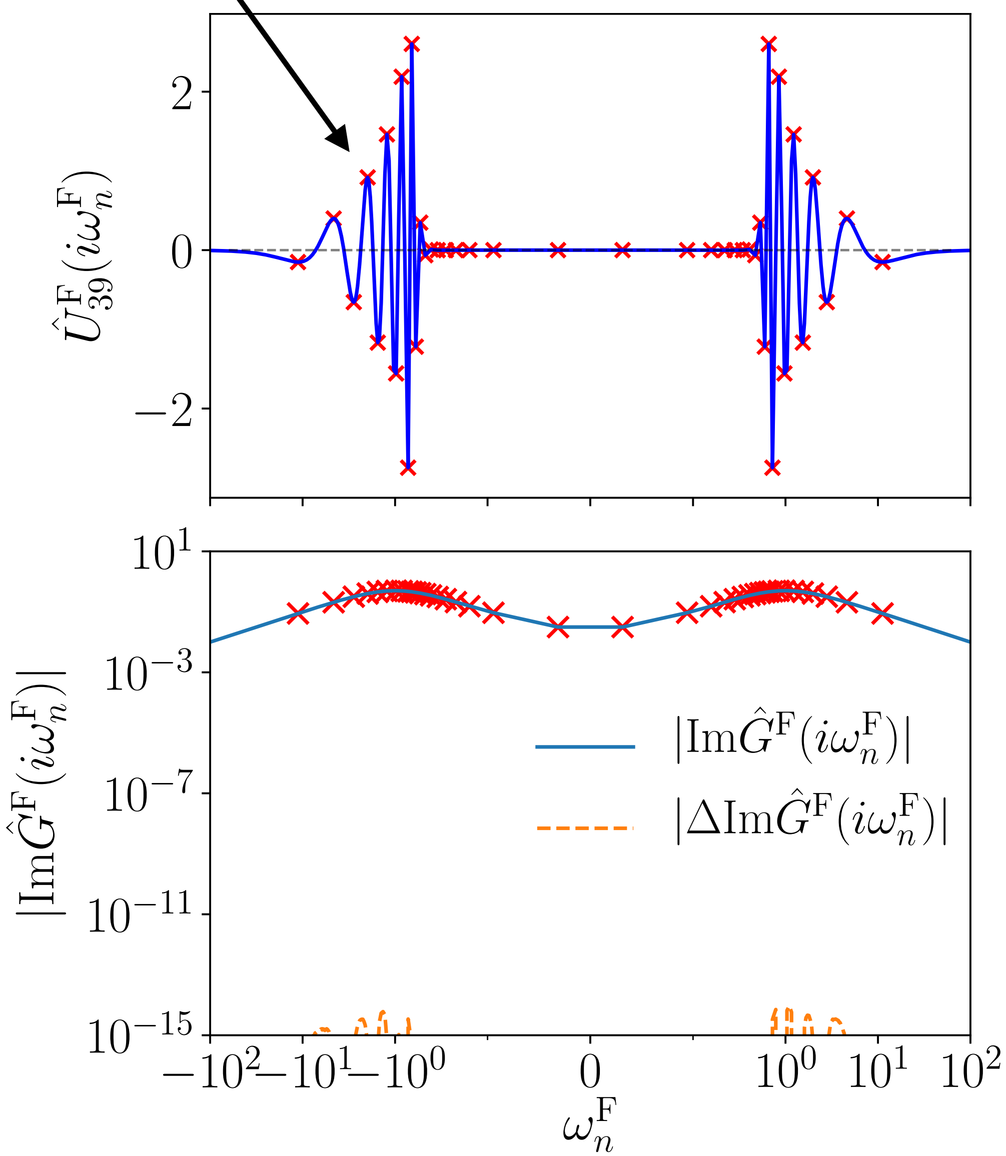
We choose sampling points near extrema of the highest basis function.

$\{\mathrm{i}\bar{\omega}_1^{\mathrm{F}}, \cdots, \mathrm{i}\bar{\omega}_N^{\mathrm{F}}\}$

β=100, $\omega_{\max}$=1
Cutoff of $S_l / S_0 > 10^{-15}$
N=40



$\{G(\mathrm{i}\bar{\omega}_i^{\mathrm{F}})\} \rightarrow \{G_l\} \rightarrow G(\mathrm{i}\omega)$



$$G_l = \underset{G_l}{\mathrm{argmin}} \sum_{\mathrm{i}\omega \in \mathcal{W}^{\alpha}} \left| \hat{G}(\mathrm{i}\omega) - \sum_{l=0}^{L-1} \hat{U}_l^{\alpha}(\mathrm{i}\omega) G_l \right|^2$$

$$= \left(\hat{\mathbf{F}}_{\alpha}^{+} \hat{\boldsymbol{g}} \right)_l \qquad (\hat{\mathbf{F}}_{\alpha})_{kl} = \hat{U}_l^{\alpha}(\mathrm{i}\omega_k^{\alpha})$$

Numerical demonstration

J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)

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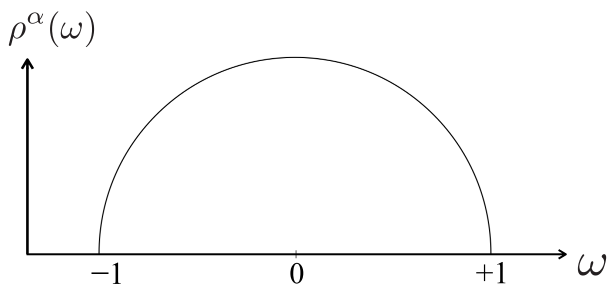
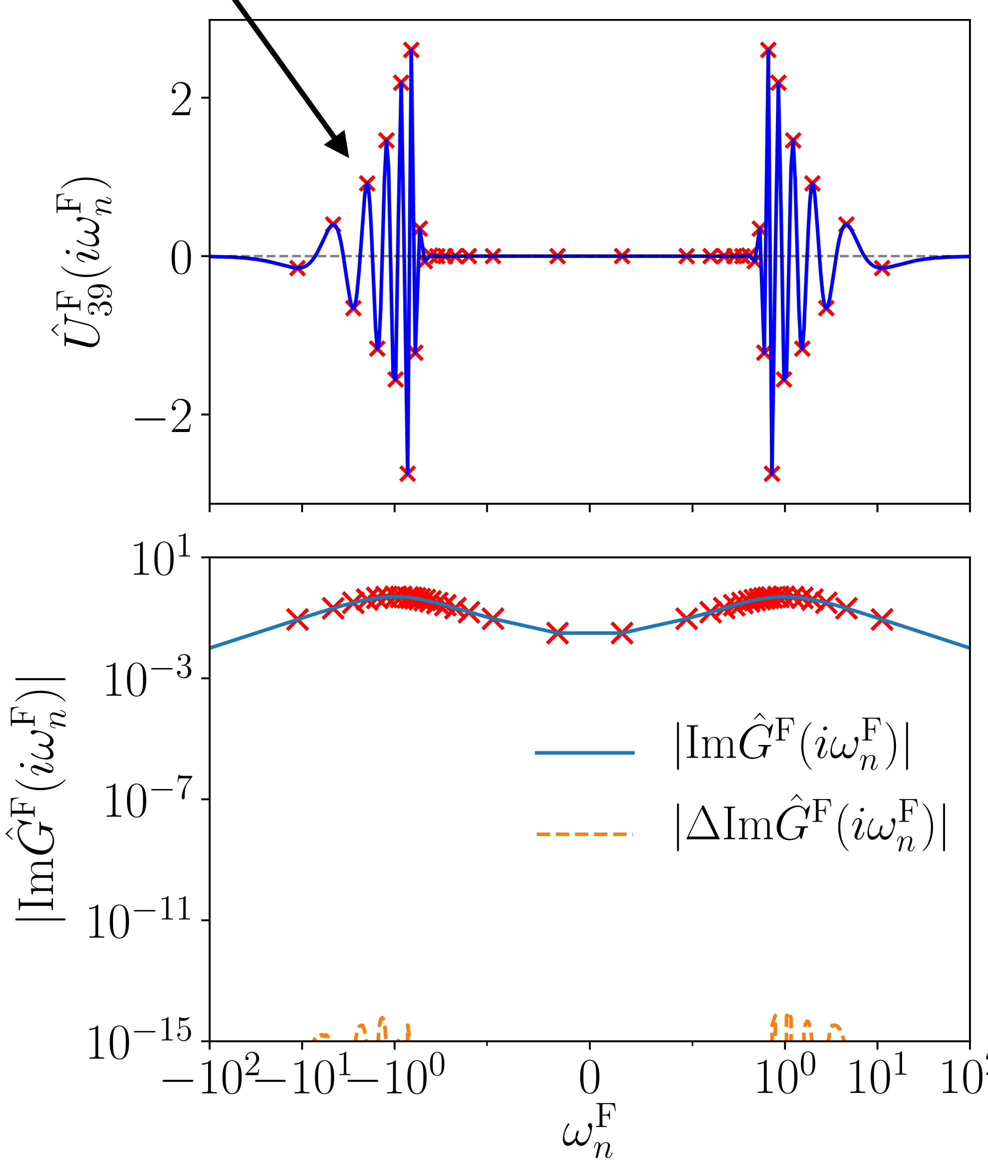


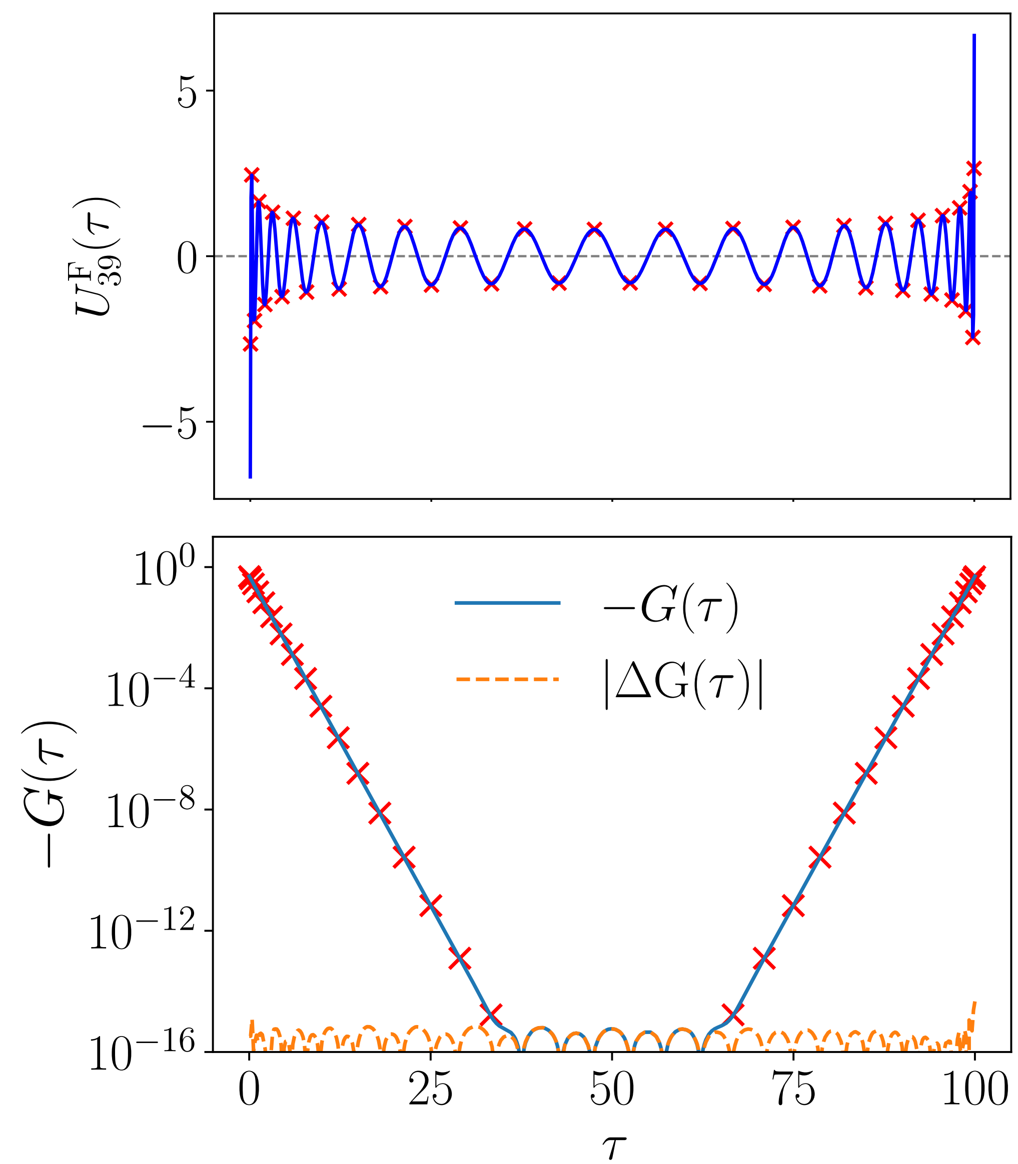
We choose sampling points near extrema of the highest basis function.

$\{\mathrm{i}\bar{\omega}_1^{\mathrm{F}}, \cdots, \mathrm{i}\bar{\omega}_N^{\mathrm{F}}\}$

$\{G(\mathrm{i}\bar{\omega}_i^{\mathrm{F}})\} \rightarrow \{G_l\} \rightarrow G(\mathrm{i}\omega)$



$\{G(\bar{\tau}_i)\} \rightarrow \{G_l\} \rightarrow G(\tau)$



How to solve Dyson equation

J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)

1. Solve Dyson equation on sampling frequencies

$$G(\mathrm{i}\bar{\omega}_k^{\mathrm{F}}) = \frac{1}{\mathrm{i}\bar{\omega}_k^{\mathrm{F}} - H - \Sigma(\mathrm{i}\bar{\omega}_k^{\mathrm{F}})} \qquad k = 1, \cdots, N$$

Computational complexity: $O(N_{\mathrm{orb}}^3 \log \beta)$

2. Compute G_l from sampled values

$$\{G(\mathrm{i}\bar{\omega}_k^{\mathrm{F}})\} \rightarrow \{G_l\}$$

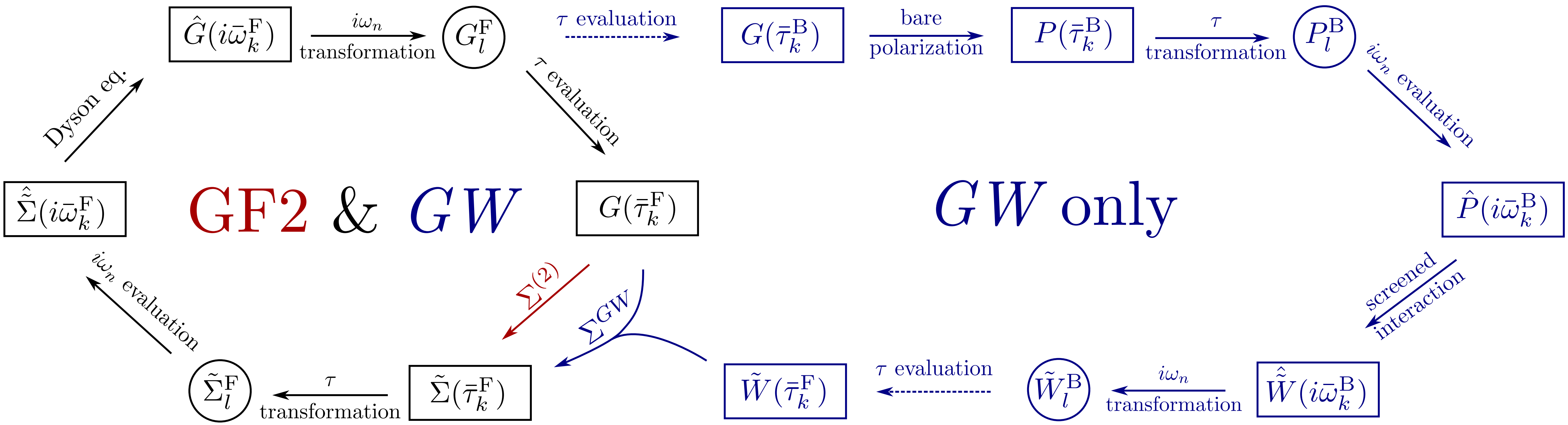
Fitting by applying pseudo inverse matrix

Computational complexity: $O(N_{\mathrm{orb}}^2 \log^2 \beta)$

3. Evaluate $G(\mathrm{i}\omega_n)$/ $G(\tau)$ from G_l

Self-consistent GF2 & *GW* calculations

J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)



$$\Sigma_{ij}^{(2)}(\tau) = -G_{kl}(\tau)G_{qm}(\tau)G_{np}(-\tau)V_{ikpq}(2V_{ljmn} - V_{mjln})$$

$$\Sigma^{GW}(\tau) = -G(\tau)W(\tau)$$

The data always stay compact! (N<200)

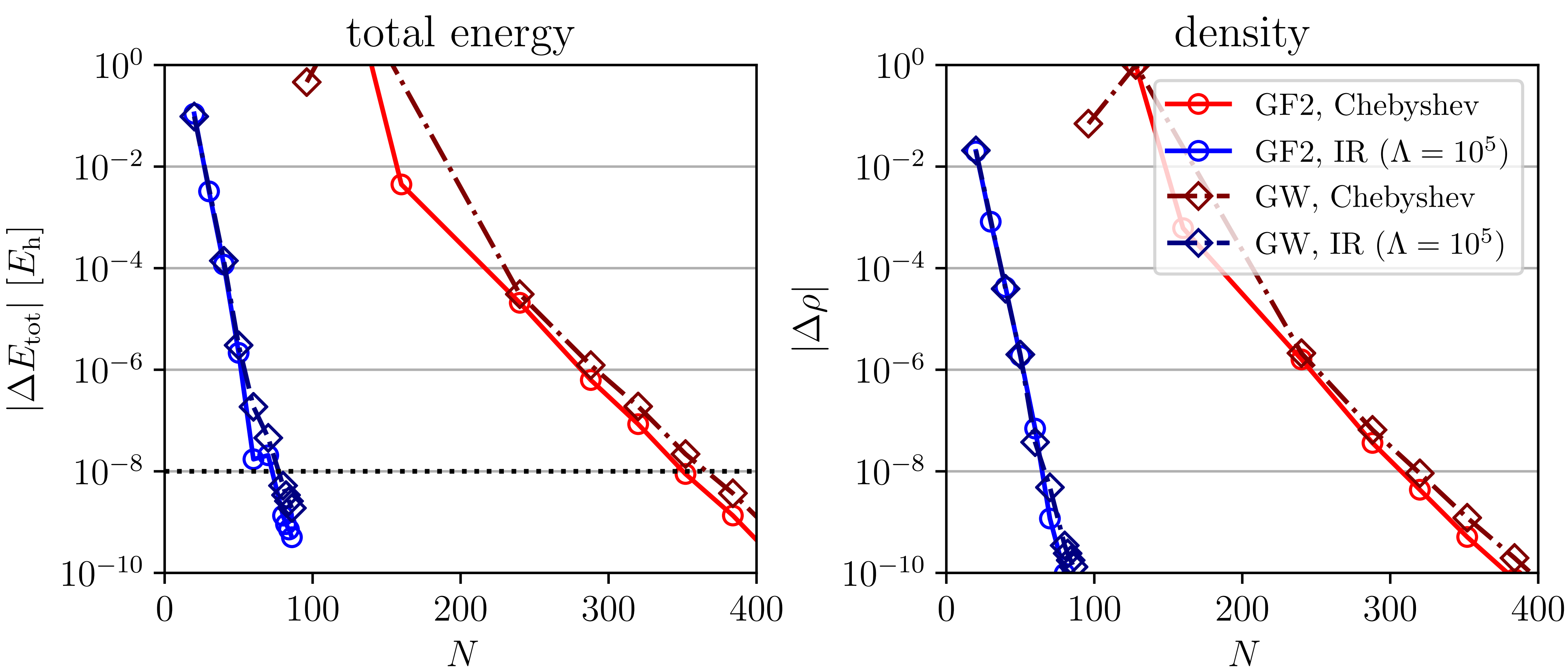
Example: Molecules

J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)

10 hydrogen atoms placed on a straight line with equal spacing a_0

cf. M. Motta *et al.*, PRX **7**, 031059 (2017)

$\beta = 1000\ E_\mathrm{h}^{-1}$ ($T \simeq 315.8$ K)



julia + python

Reference data: $N \to +\infty$

Example: noble gas atoms with deep core states

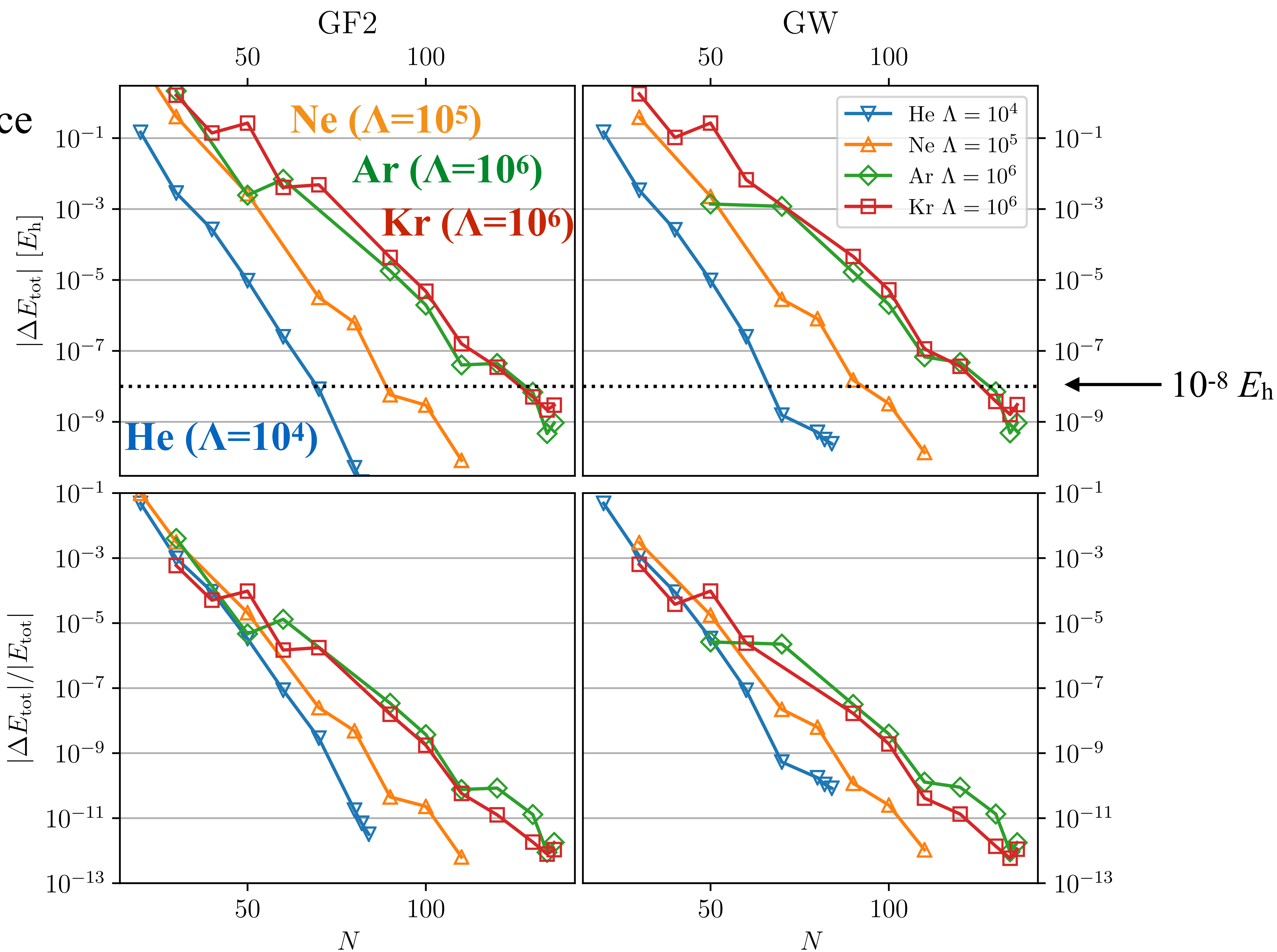
J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)

Chebyshev is no more doable.

$\beta = 1000\ E_\mathrm{h}^{-1}$ ($T \simeq 315.8$ K)

Energy difference

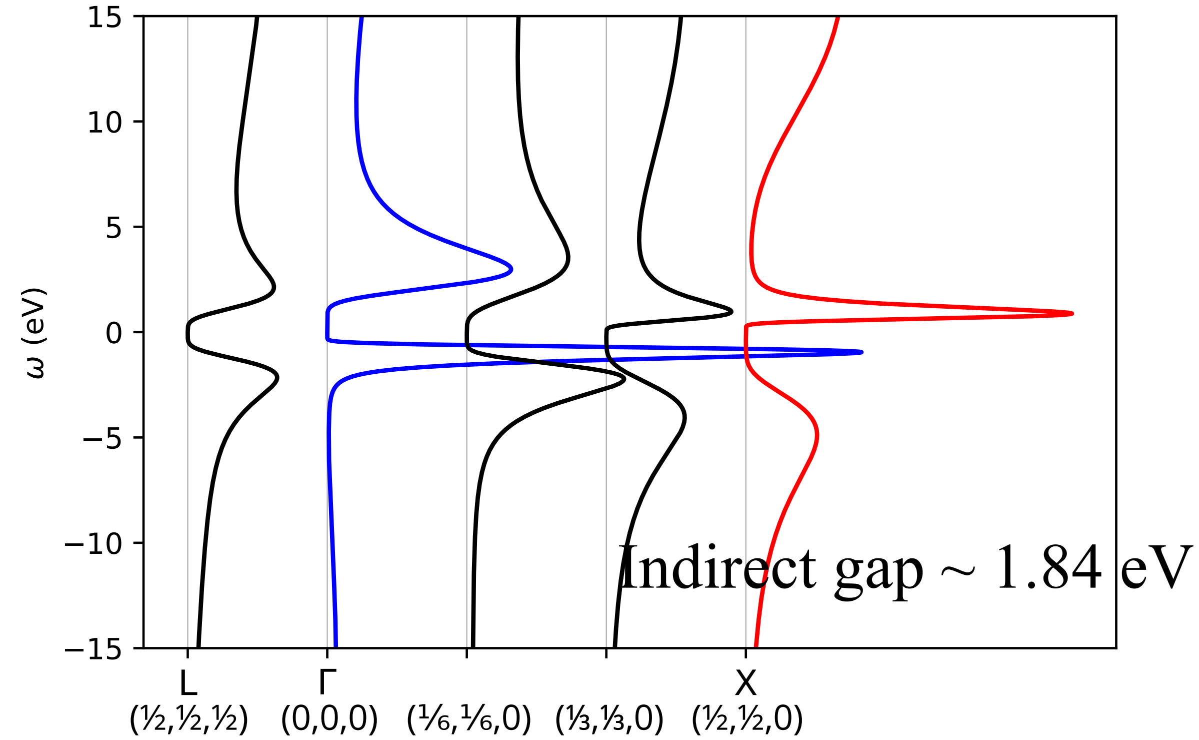
Relative energy difference



Example: *GW* of silicon crystal

J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, <u>HS</u>, PRB **101**, 035144 (2020)

Chebyshev		IR $\Lambda = 10^4$		IR $\Lambda = 10^5$	
N	E_{tot} $[E_{\mathrm{h}}]$	N	E_{tot} $[E_{\mathrm{h}}]$	N	E_{tot} $[E_{\mathrm{h}}]$
100	-8.0270874	80	-7.8804300	110	-7.8804300
150	-7.8861508	82	-7.8804300	112	-7.8804300
200	-7.8806642	84	-7.8804300		
250	-7.8804379	86	-7.8804300		
300	-7.8804302				
350	-7.8804298				



$4 \times 4 \times 4$ mesh, $\beta = 1000\ E_{\mathrm{h}}^{-1}$ ($T \simeq 315.8$ K)

GTH-DZVP basis of Gaussian orbitals (13 orbitals per Si atom)

Example: Migdal-Eliashberg theory

T. Wang, T. Nomoto, Y. Nomura, HS, J. Otsuki, T. Koretsune, and R. Arita, PRB **102**, 134503 (2020)

Numerically demanding to estimate T_c of $O(10)$ K considering the retardation effect from first principles

Linearized gap equation

$$\tilde{\lambda}\Delta_m(\boldsymbol{k}, i\omega_n) = -\frac{T}{N_{\boldsymbol{k}}}\sum_{m'}\sum_{\boldsymbol{k}', i\omega_{n'}} \mathcal{K}_{mm'}(\boldsymbol{k}-\boldsymbol{k}', i\omega_n - i\omega_{n'}) \times |G_{m'}(\boldsymbol{k}', i\omega_{n'})|^2 \Delta_{m'}(\boldsymbol{k}', i\omega_{n'})$$

Convolution can be performed efficiently using a sparse grid in τ.

- Pairing interaction kernel $\mathcal{K}_{mm'} = \mathcal{K}^{\mathrm{el-ph}}_{mm'} + \mathcal{K}^{\mathrm{C}}_{mm'}$

- Renormalized electron Green's function

$$G_m(\boldsymbol{k}, i\omega_n) = \frac{1}{i\omega_n - \varepsilon_{m\boldsymbol{k}} - \Sigma_m(\boldsymbol{k}, i\omega_n)}$$

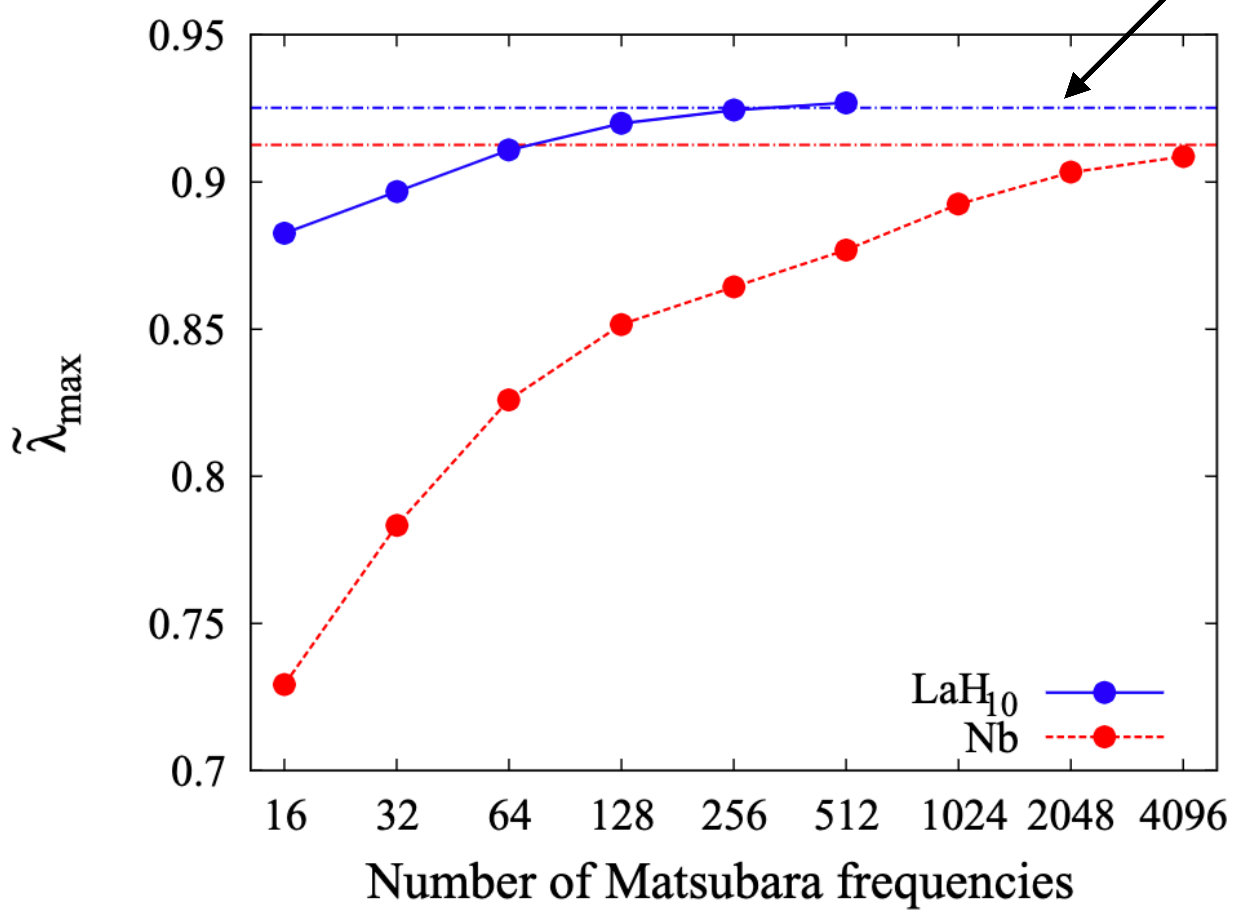
$$\Sigma_m(\boldsymbol{k}, i\omega_n) = -\frac{T}{N_{\boldsymbol{k}}}\sum_{m'}\sum_{\boldsymbol{k}', i\omega_{n'}} \mathcal{K}^{\mathrm{el-ph}}_{mm'}(\boldsymbol{k}-\boldsymbol{k}', i\omega_n - i\omega_{n'}) G_{m'}(\boldsymbol{k}', i\omega_{n'}).$$

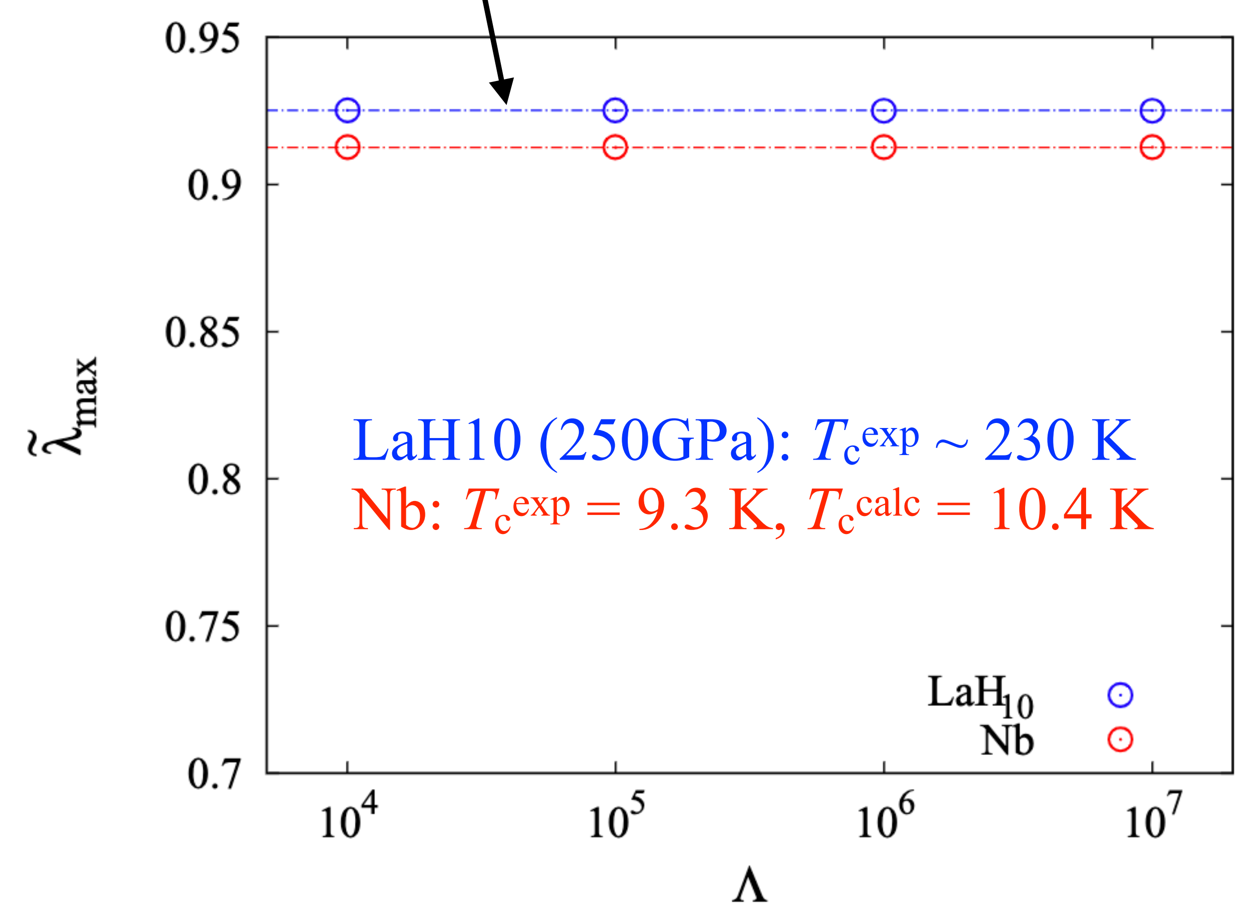
Example: Migdal-Eliashberg equation

T. Wang, T. Nomoto, Y. Nomura, HS, J. Otsuki, T. Koretsune, and R. Arita, PRB **102**, 134503 (2020)

Just above T_c ($\tilde{\lambda}_\mathrm{max} = 1$)

Results obtained by using IR + sparse sampling





Converged results even for $T_\mathrm{c} = O(10)$ K!

- Memory consumption 1/30
- Computational time 1/20

$\Lambda = 10^5$ v.s. $N_\omega = 4096$

Open source software: irbasis

https://github.com/SpM-lab/irbasis

N. Chikano, K. Yoshimi, J. Otsuki, H. Shinaoka (2018) + M. Wallerberger (2019)

- Python and C++
- Step-by-step tutorial

python™

Integral form

$$S_l^\alpha U_l^\alpha(\tau) = \int_{-\omega_{\max}}^{\omega_{\max}} d\omega K^\alpha(\tau,\omega) V_l^\alpha(\omega)$$

$$\Big\Downarrow \; x \equiv 2\tau/\beta - 1,\ y \equiv \omega/\omega_{\max}$$

$$\underline{s_l^\alpha u_l^\alpha(x)} = \int_{-1}^{1} dy k^\alpha(x,y) \underline{v_l^\alpha(y)} \qquad x, y \in [-1,1]$$

Unknown analytic solution

Precomputed in multiprecision arithmetic for Λ=10,10²,…, 10⁷

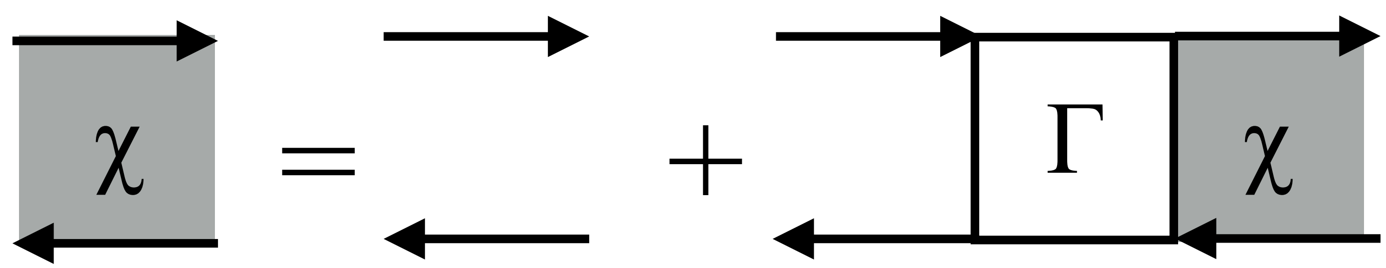
N. Chikano, J. Otsuki, H. Shinaoka, PRB **98**, 035104 (2018)

Outline

- Single-particle Green's function
  - IR basis and sparse sampling
  - Applications to *ab initio* calculations
- Extension to two-particle Green's function

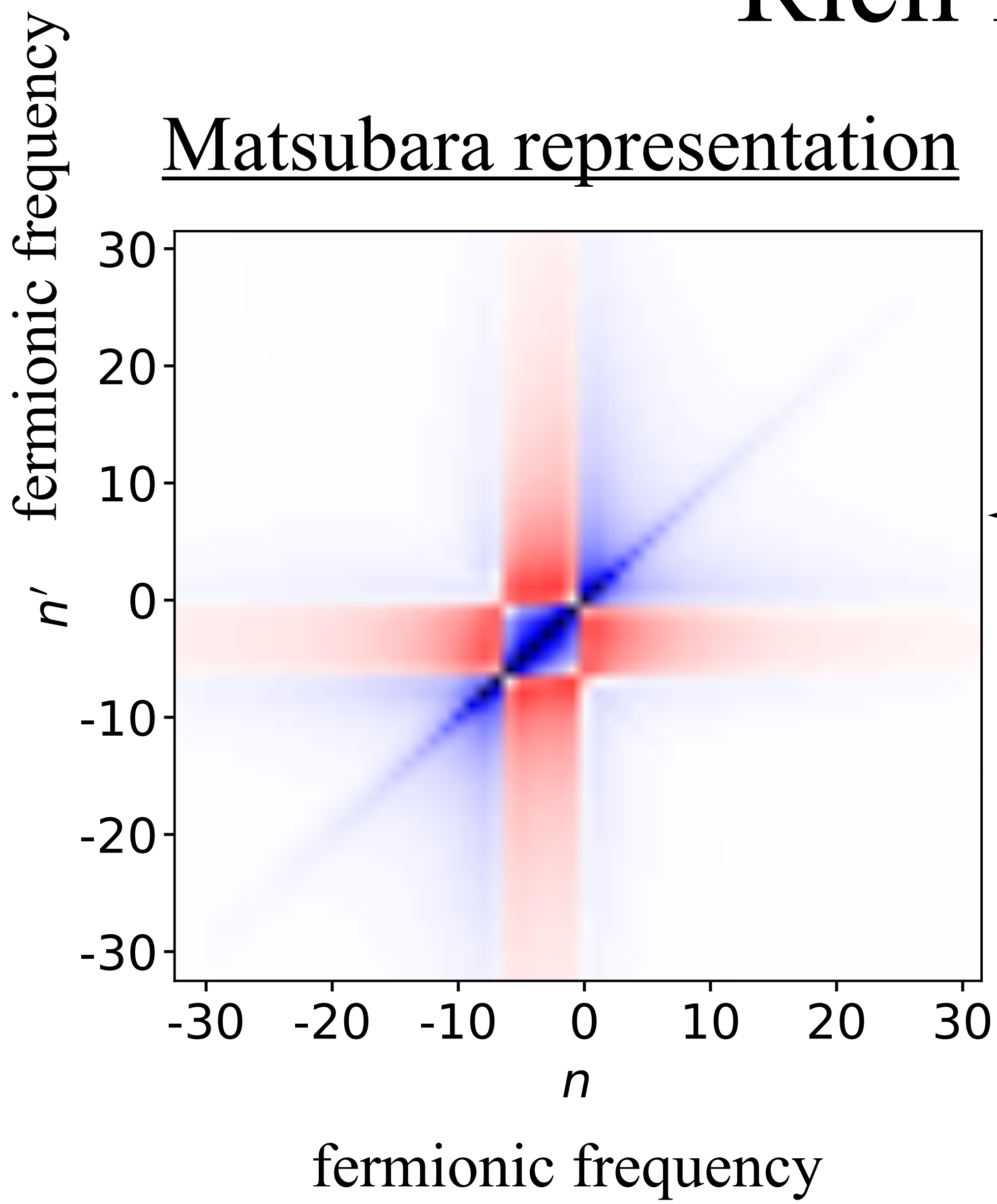
Why two-particle objects?

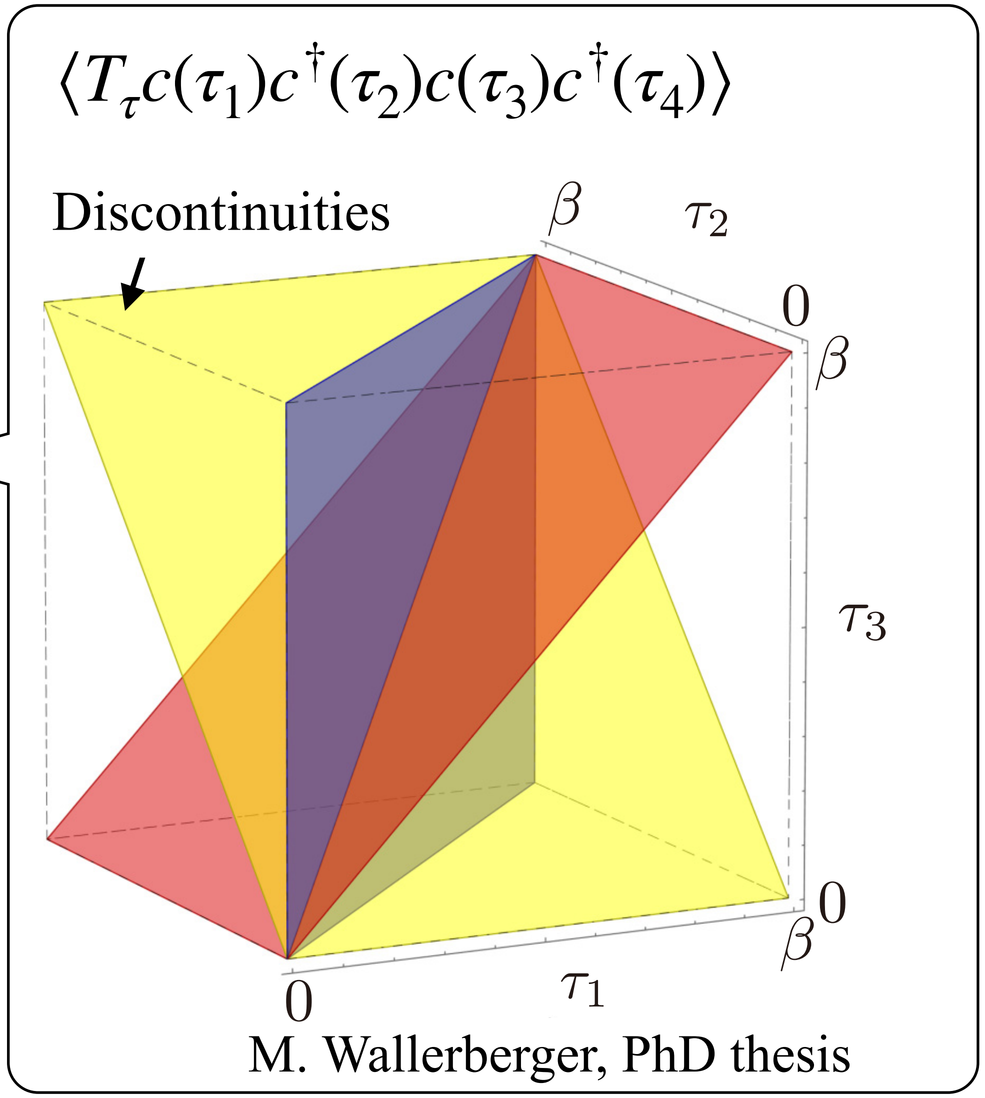
- Vertex corrections
- My motivation: Dynamical mean-field theory (DMFT)
  - Dynamic susceptibility (inelastic scattering experiments)
  - Non-local correlations beyond DMFT

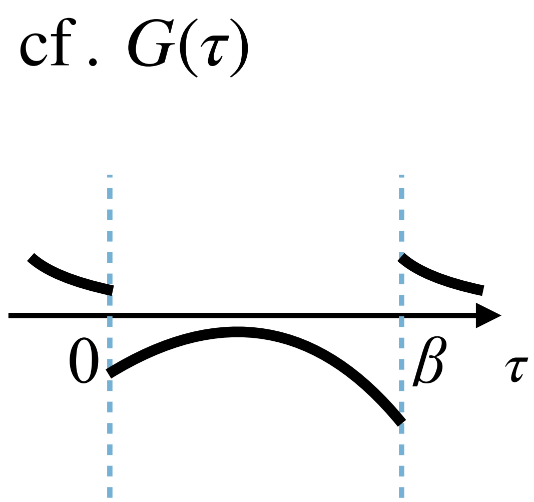


Bethe-Salpeter equation

Rich frequency structure







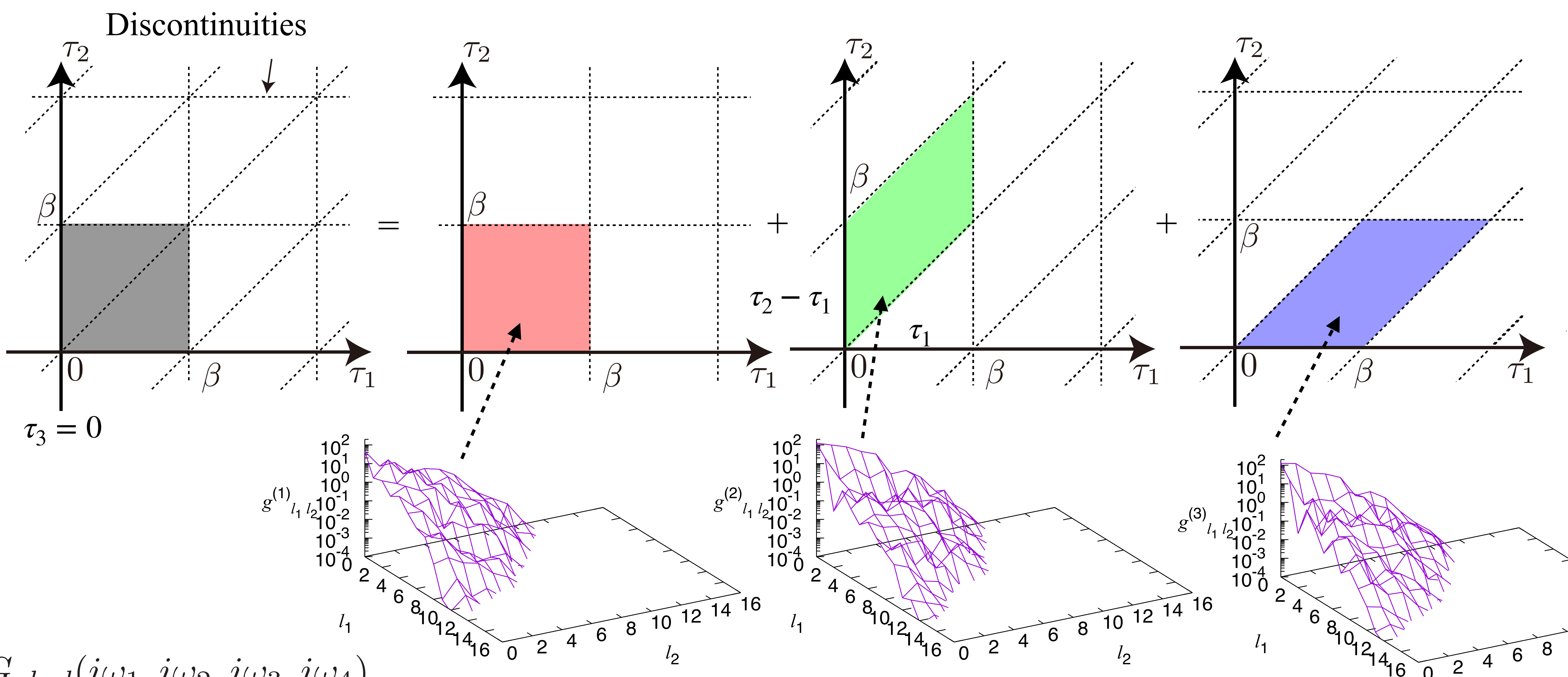
- Large size: $O(\beta^3)$
- More indices for spin, orbital, wave vector…

NEW

- IR approach frequency dependence
- Sparse sampling and tensor network representation

IR approach

HS, J. Otsuki, M. Ohzeki, K. Yoshimi, K. Haule, M. Wallerberger, E. Gull, PRB **97**, 205111 (2018)



$$G_{abcd}(i\omega_1, i\omega_2, i\omega_3, i\omega_4)$$

$$= \delta_{\sum_i \omega_i, 0} \sum_{l_1, l_2, l_3 = 0}^{\infty} \Big\{ G^{(1)}_{abcd; l_1 l_2 l_3} U^{\mathrm{F}}_{l_1}(i\omega_1) U^{\mathrm{F}}_{l_2}(i\omega_2) U^{\mathrm{F}}_{l_3}(i\omega_3) + \cdots +$$

$$G^{(16)}_{abcd; l_1 l_2 l_3} U^{\mathrm{F}}_{l_1}(i\omega_3) U^{\overline{\mathrm{B}}}_{l_2}(i\omega_3 + i\omega_2) U^{\mathrm{F}}_{l_3}(-i\omega_4) \Big\}$$

$$= \delta_{\sum_i \omega_i, 0} \sum_{r=1}^{16} \sum_{l_1 l_2 l_3} G^{(r)}_{abcd; l_1 l_2 l_3} U^{\alpha}_{l_1}(i\omega) U^{\alpha'}_{l_2}(i\omega') U^{\alpha''}_{l_3}(i\omega'')$$

- Exponential decay
- Small size: $O((\log \beta)^3)$

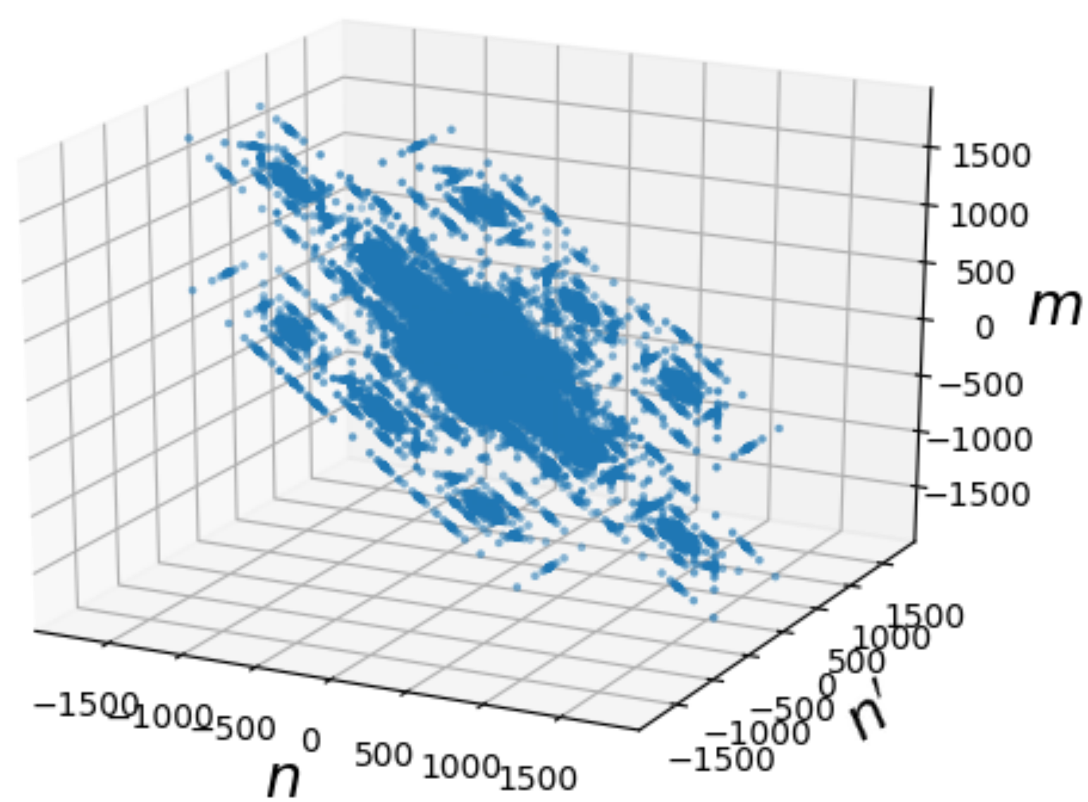
Spectrum representation of two-particle quantities

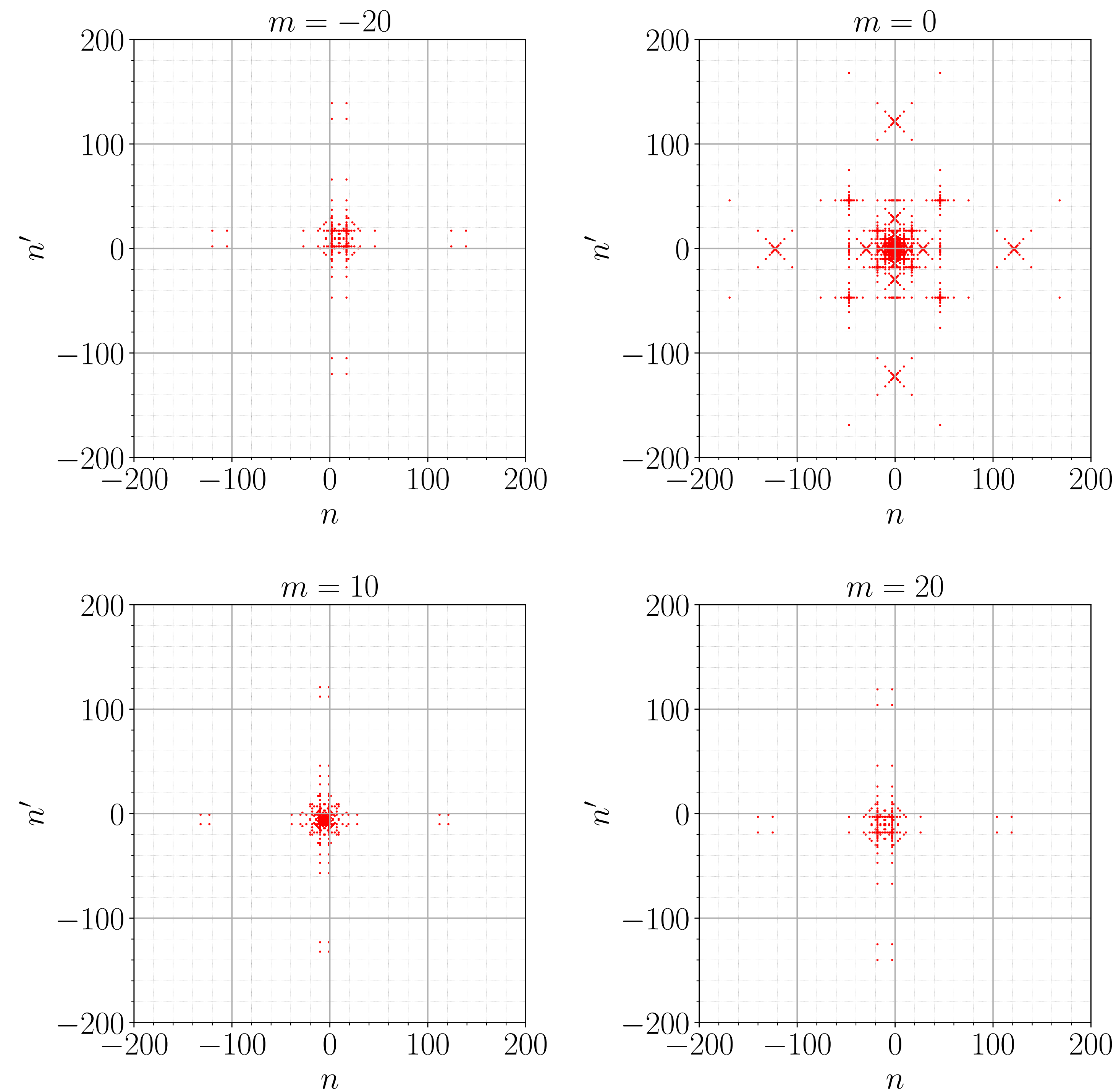
A. Toschi *et al.*, PRB **75**, 045118 (2007)
H. Hafermann *et al.*, EPL **85**, 27007 (2009)
A. Shvaika, CMP **19**, 33004 (2016)

1. Sparse sampling

HS, D. Geffroy, M. Wallerberger, J. Otsuki, K. Yoshimi, E. Gull, J. Kuneš, SciPost Phys. **8**, 012 (2020)

$\Lambda=10^3$ $\quad X^{\mathrm{loc}}_{ijkl}(i\omega_n, i\omega_{n'}; i\nu_m)$





1.5×10^5 sampling points $\ll$ 3×10^9 points in box

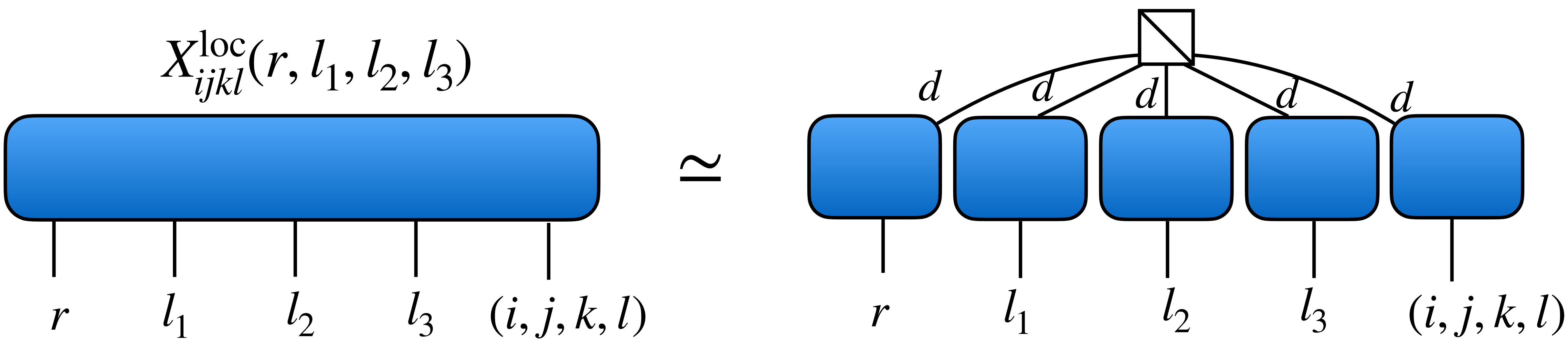
2. Dimensionality reduction by tensor network

<u>HS</u>, D. Geffroy, M. Wallerberger, J. Otsuki, K. Yoshimi, E. Gull, J. Kuneš, SciPost Phys. **8**, 012 (2020)

$$X^{\mathrm{loc}}_{ijkl}(r, l_1, l_2, l_3) \simeq \sum_{d=1}^{D} x^{(1)}_{dr} x^{(2)}_{dl_1} x^{(3)}_{dl_2} x^{(4)}_{dl_3} x^{(5)}_{dijkl}$$

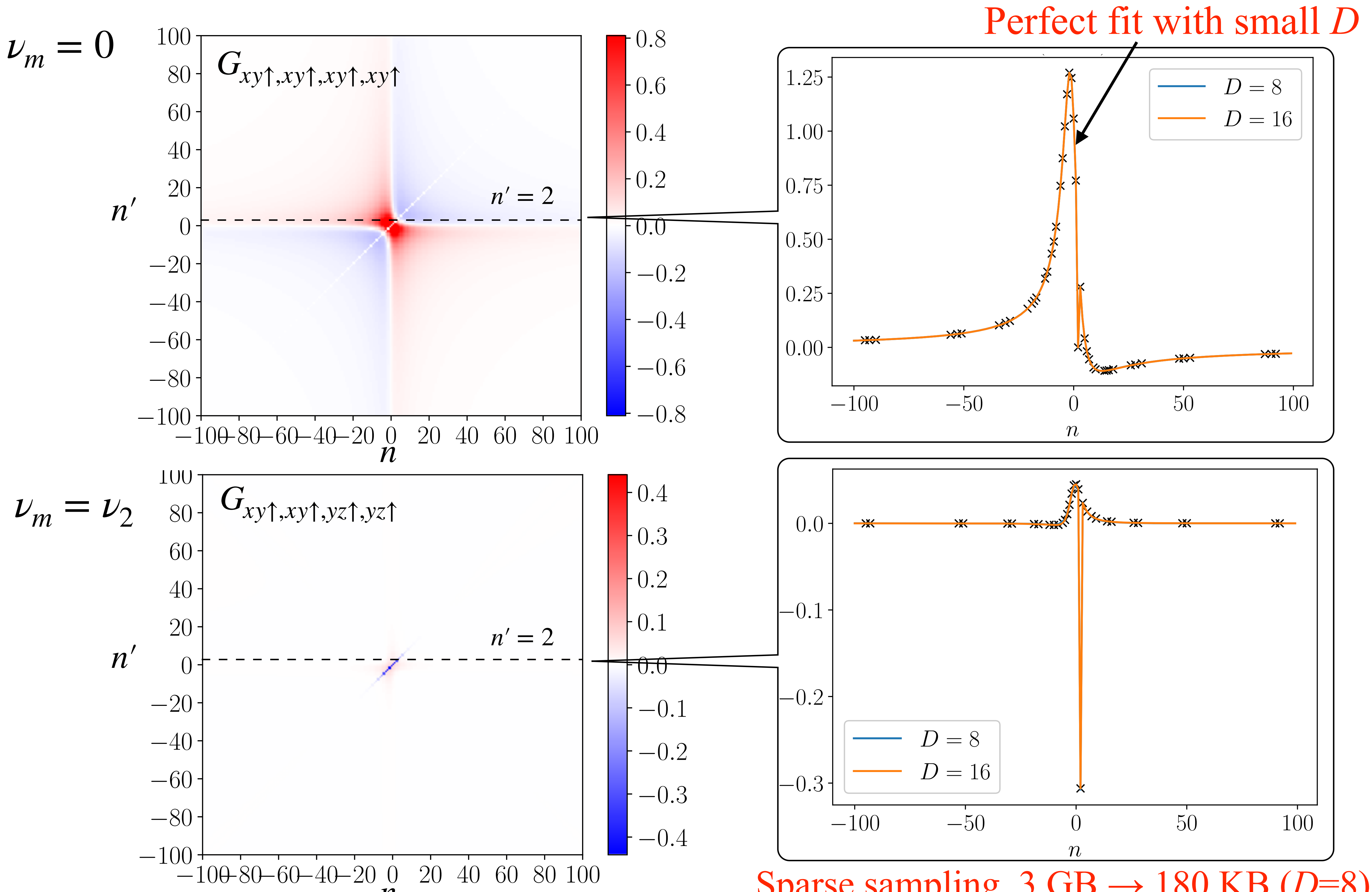
16   < 30 Assumption: D is small.

- Further compactification of IR tensor



Three-orbital t_{2g} model (atom) $U = 5$ eV, $J = 0.1$ eV, $\beta = 10$

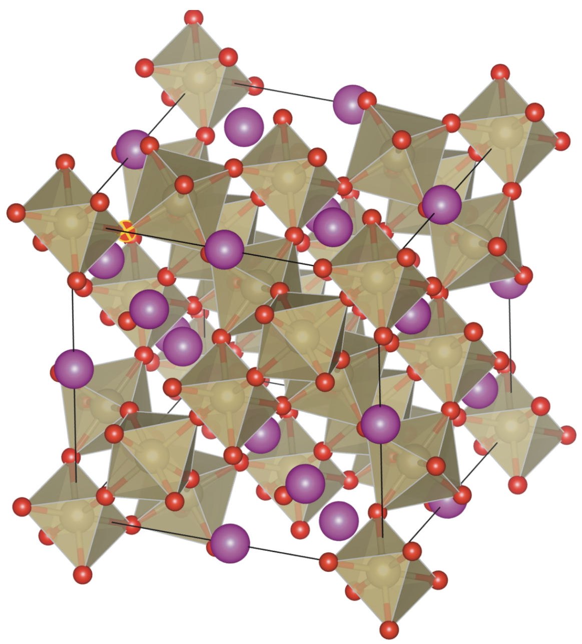
Exact diagonalization $G_{a\sigma_1,b\sigma_2,c\sigma_3,d\sigma_4}(i\omega_n, i\omega_{n'}, i\nu_m)$ $a,b,c,d = d_{xy}, d_{yz}, d_{zx}$ $\Lambda = 1000$

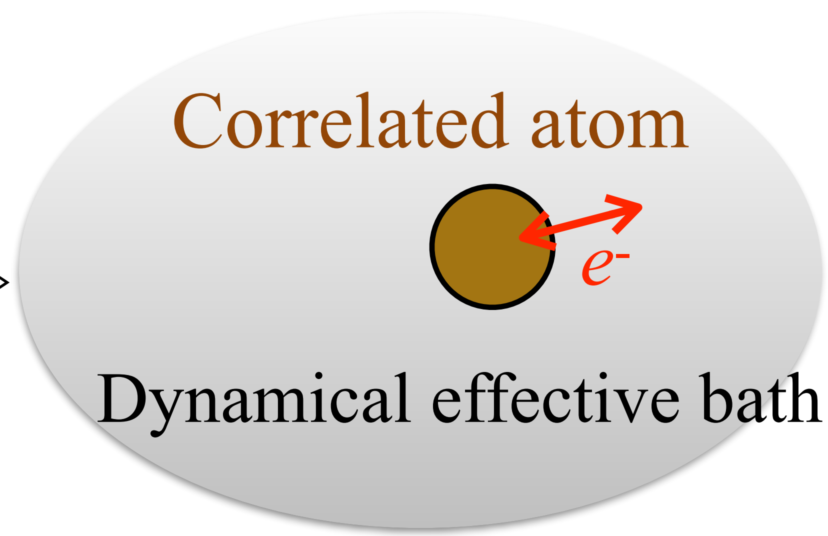


Sparse sampling 3 GB → 180 KB (D=8)

Computing dynamic susceptibility in DMFT

Review: G. Kotliar *et al*., Rev. Mod. Phys. **78**, 865 (2006)





QMC

Local two-particle Green's functions

$$X^{\mathrm{loc}}_{abcd}(i\omega_n, i\omega_{n'}; i\nu_m)$$

spin/orbital fermion boson

Huge data

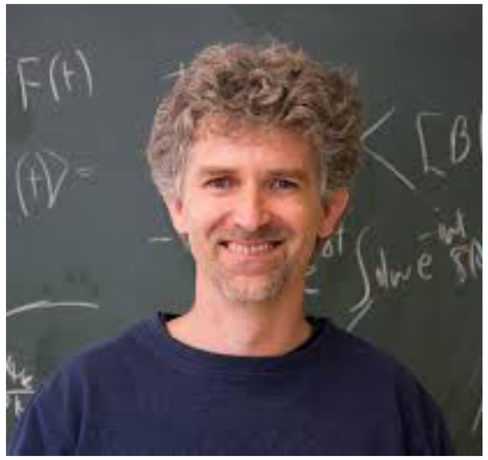
Bethe-Salpeter equation

$$\chi_{abcd}(q, i\nu_m)$$

Comparison with neutron scattering *etc*.

More realistic systems

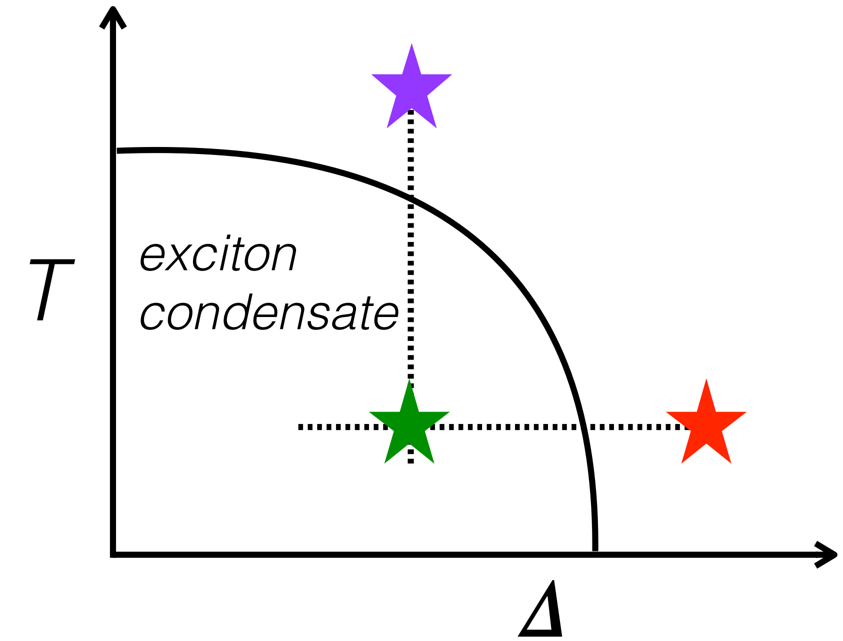
D. Geffroy@Brno

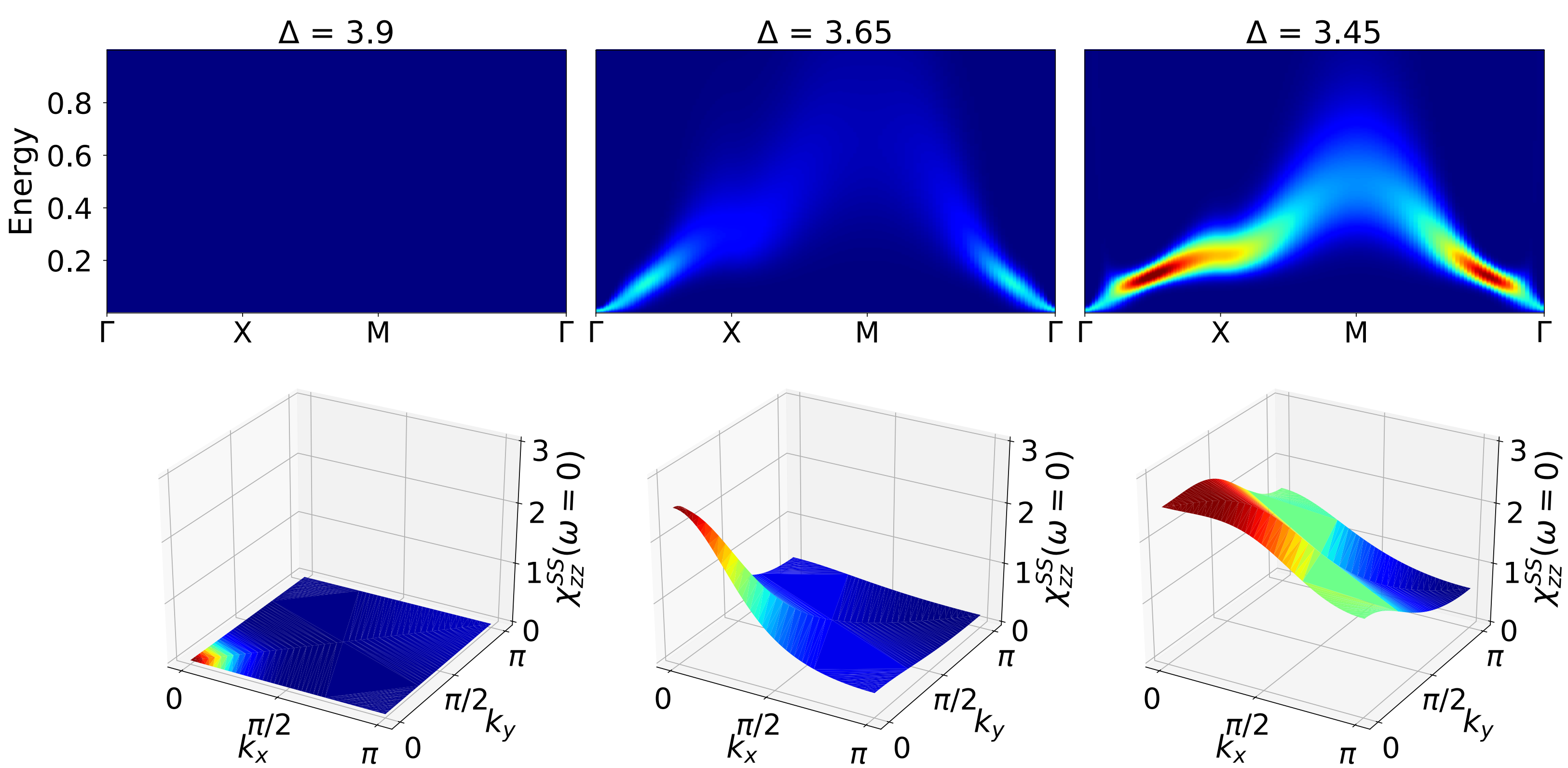


J. Kuneš@Wien

Two-band model with crystal-field splitting

D. Geffroy, J. Kaufmann, A. Hariki, P. Gunacker, A. Hausoel and J. Kuneš, PRL **122**, 127601 (2019)

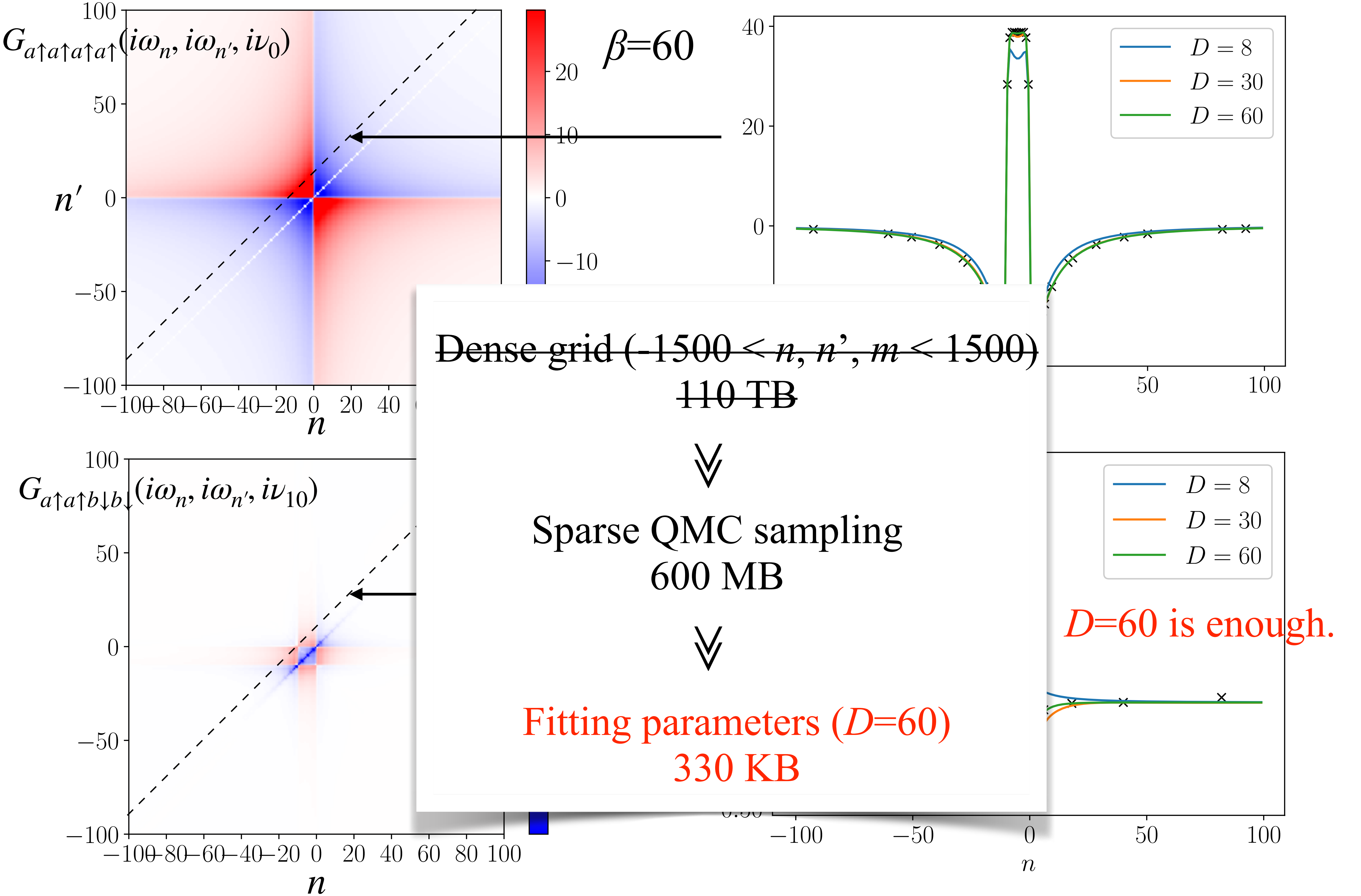




- Worm sampling in Legendre basis by ALPS/CT-HYB

  40 hours with 840 processes

Sparse QMC sampling



~~Dense grid (-1500 < n, n', m < 1500)~~
~~110 TB~~

⩔

Sparse QMC sampling
600 MB

⩔

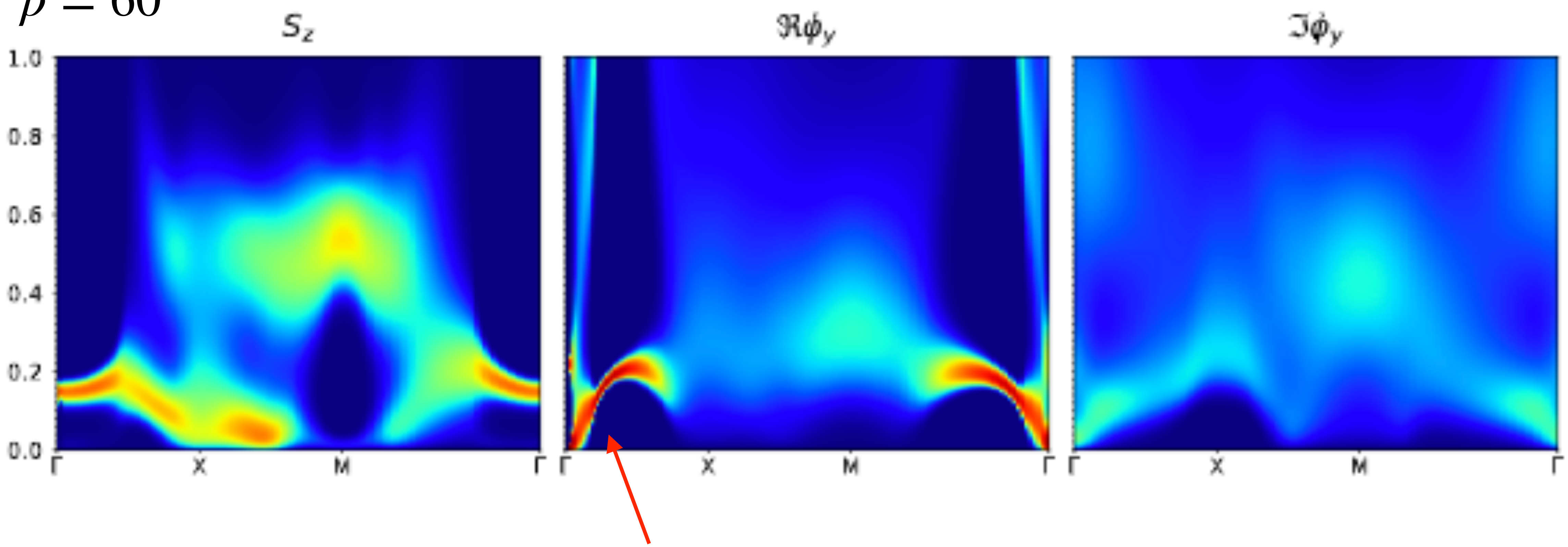
Fitting parameters (D=60)
330 KB

Solving Bethe-Salpeter equation

HS, D. Geffroy, M. Wallerberger, J. Otsuki, K. Yoshimi, E. Gull, J. Kuneš, SciPost Phys. **8**, 012 (2020)

Interpolating the local vertex, one can solve Bethe-Salpeter equation using a dense large Matsubara mesh.

$\beta = 60$



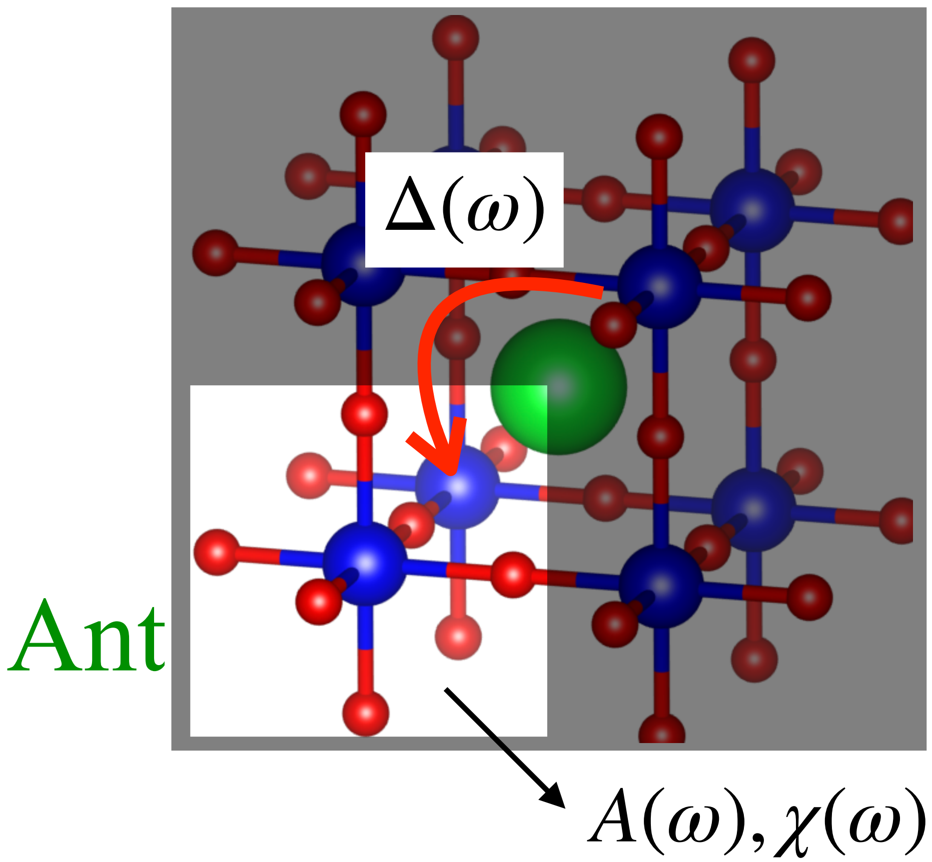
Goldstone mode in the condensed state

Remaining issue: How to solve diagrammatic equations at the two-particle level in IR?

M. Wallerberger*, HS*, A. Kauch, arXiv:2012.05557

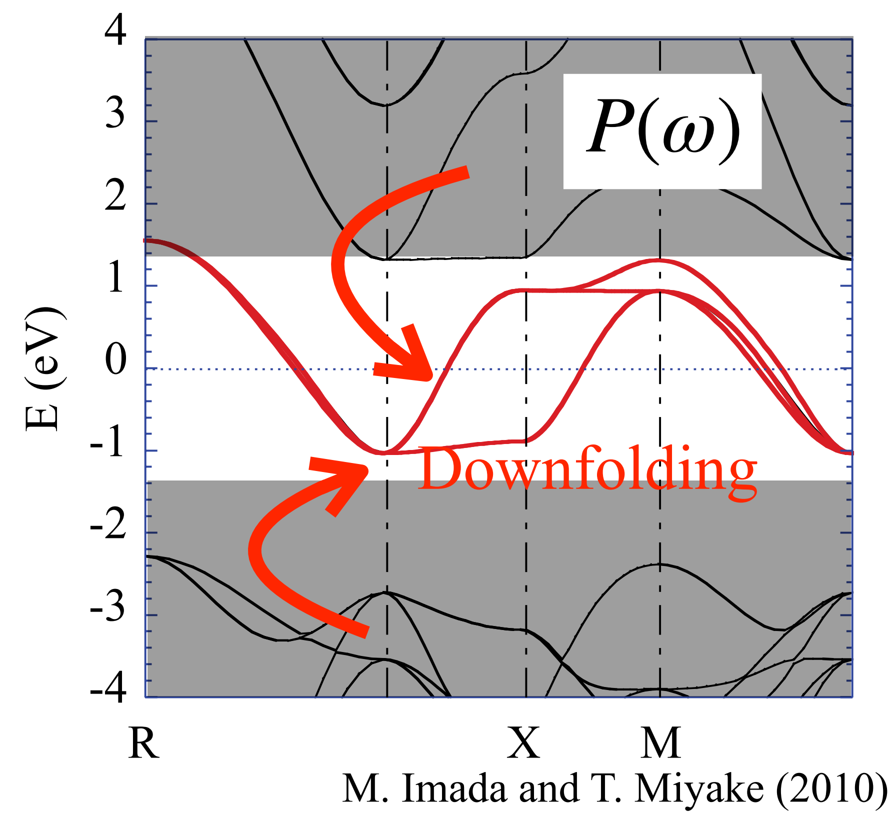
Future perspectives: Quantum embedding theories

Embedding in space



Dynamical mean-field theory, dynamical vertex approximation *etc*.

Embedding in energy



Constrained RPA *etc*.

Dynamical susceptibility calculations in DMFT, non-local diagrammatic calculations of quantum critical and unconventional superconductivity…

Summary

Intermediate representation + Sparse sampling + Tensor networks

- ☑ Compact representation of Green's functions
- ☑ Diagrammatic equations at single-particle level
- ☑ Sparse QMC measurement
- ☐ Computation at two-particle level (ongoing)

⇨ Unconventional superconductivity *etc.*

「固体物理」に解説記事を執筆中！

One-particle theory

- HS, J. Otsuki, M. Ohzeki, K. Yoshimi, PRB **96**, 035147 (2017)
- J. Otsuki, M. Ohzeki, HS, K. Yoshimi, PRE **95**, 061302(R) (2017)
- N. Chikano, J. Otsuki, HS, PRB **98**, 035104 (2018)
- N. Chikano, K. Yoshimi, J. Otsuki, HS, Compt. Phys. Commun. **240**, 181 (2019)
- J. Li, M. Wallerberger, C.-N. Yeh, N. Chikano, E. Gull, HS, PRB **101**, 035144 (2020)
- T. Wang, T. Nomoto, Y. Nomura, HS, J. Otsuki, T. Koretsune, and R. Arita, PRB **102**, 134503 (2020)

Two-particle theory

- HS, J. Otsuki, M. Ohzeki, K. Yoshimi, K. Haule, M. Wallerberger, E. Gull, PRB **97**, 205111 (2018)
- HS, D. Geffroy, M. Wallerberger, J. Otsuki, K. Yoshimi, E. Gull, J. Kuneš, SciPost Phys. **8**, 012 (2020)
- M. Wallerberger*, HS*, A. Kauch, arXiv:2012.05557