A22.6

APS MARCH MEETING

MARCH 04, 2019

The University of Texas at Austin

RECENT ADVANCES IN PARSEC FOR PERFORMING LARGE-SCALE ELECTRONIC STRUCTURE CALCULATIONS IN REAL SPACE

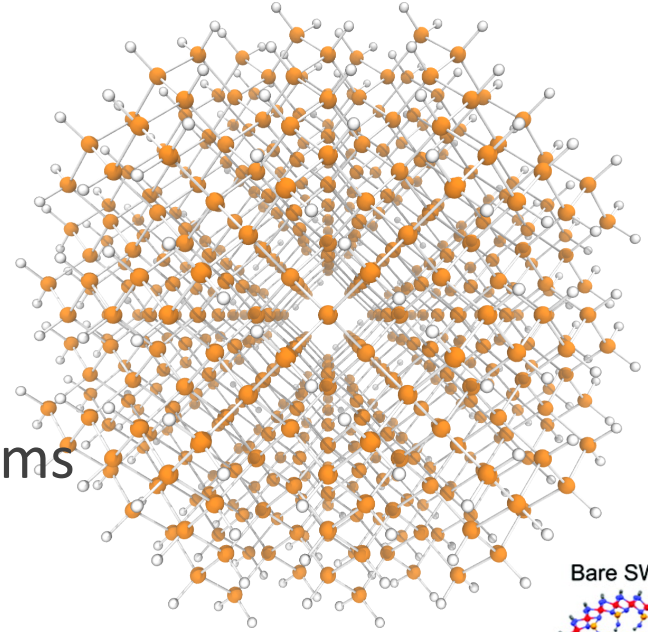
Kai-Hsin Liou[1*], James Chelikowsky[1], and Chao Yang[2]

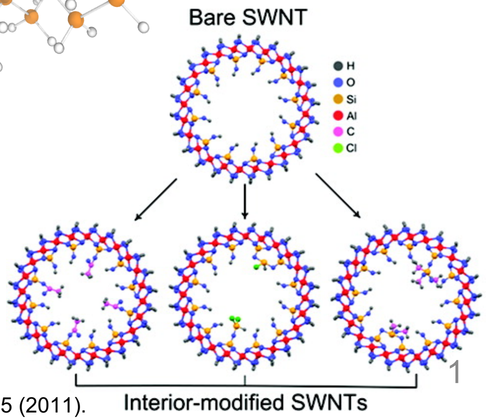
[1] The University of Texas at Austin

[2] Lawrence Berkeley National Laboratory

Motivation

- Silicon nanocrystals
  - Optoelectronics
  - 10 nm in diameter ~ 30,000 atoms
- Inorganic nanotubes
  - Catalysis
  - Water desalination
  - 20 nm in length ~ 5,000 atoms

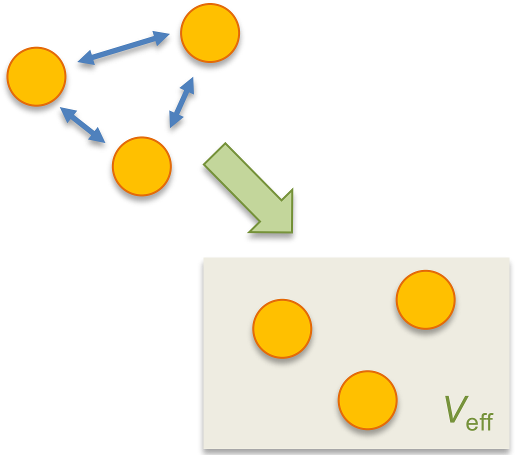


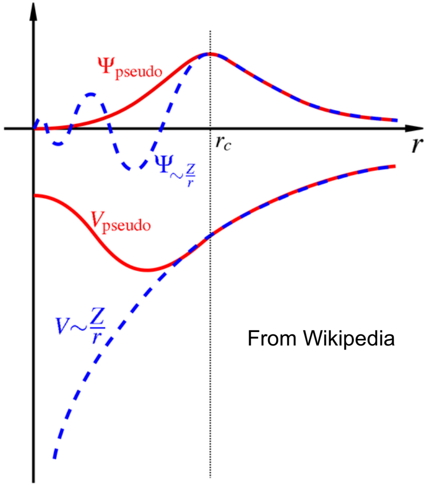


[1] D.-Y. Kang, J. Zang, C. Jones, S. Nair, Single-Walled Aluminosilicate Nanotubes with Organic-Modified Interiors, *J Phys Chem C* **115**, 7676–7685 (2011).

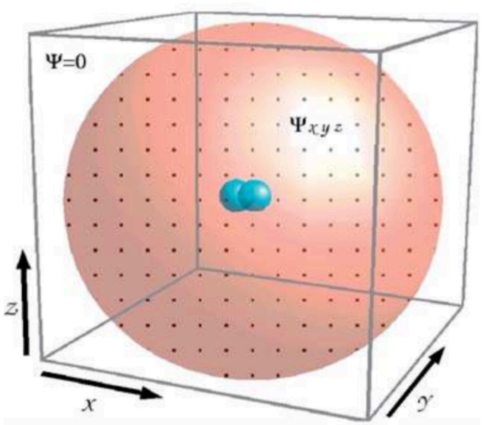
PARSEC

Pseudopotential **A**lgorithm for **R**eal-**S**pace **E**lectronic Structure **C**alculation
https://real-space.org/





From Wikipedia



Kohn–Sham DFT

N interacting electrons →

N independent e's

Pseudopotential

All electrons →

Only valence electrons

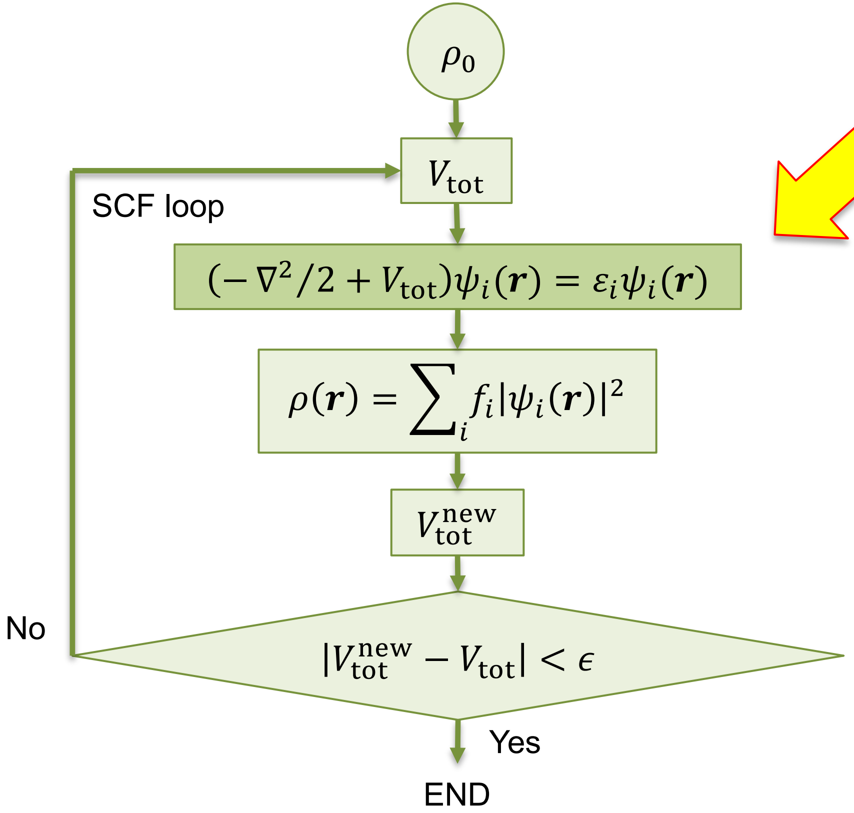
Real-Space Grids

Finite difference method

Ease of implementation

Suitable for confined systems

Solving the Kohn–Sham Equations



Key to large systems:

An efficient eigensolver

Observation: intermediate SCF iterations require only converged charge density, not individual wfns

Chebyshev-filtered Subspace Iteration

1. Filtering
2. Orthonormalization
3. Rayleigh–Ritz method

Filtering

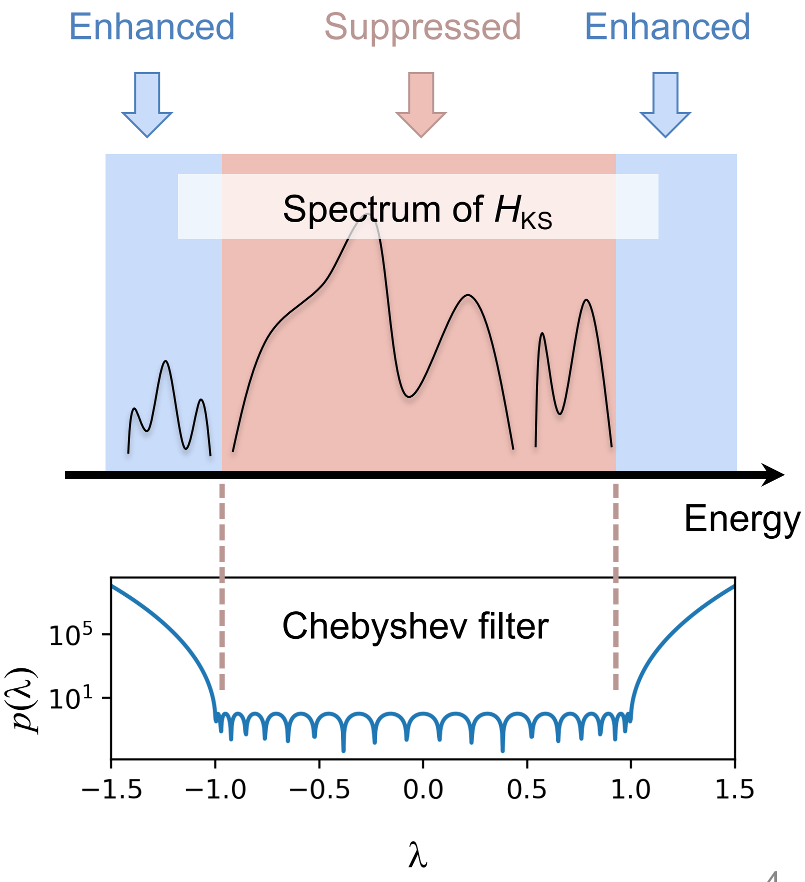
Enhance the subspace constituting the occupied states

$\tilde{p}(H_{\mathrm{KS}})$: Chebyshev polynomial of H_{KS} u_i: eigenstates
v: a state λ_i: eigenvalues

$$\tilde{p}(H_{\mathrm{KS}})v = \tilde{p}(H_{\mathrm{KS}})\sum_{i=1}^{n} c_i u_i = \sum_{i=1}^{n} c_i \tilde{p}(H_{KS}) u_i = \sum_{i=1}^{n} p(\lambda_i) c_i u_i$$

Expand v in terms of eigenstates

Operate the polynomial of H_{KS} to each eigenstate



Orthonormal. & Rayleigh–Ritz method

Generate approximate eigenpairs from a subspace

Algorithm 2 Cholesky QR

1: **procedure** $V = \text{ORTH}(W)$
2: $\quad A = W^T W$ $\qquad \triangleright$ A is positive definite
3: $\quad$ Find an upper triangular R such that $A = R^T R$ $\qquad \triangleright$ Cholesky factorization
4: $\quad V = WR^{-1}$

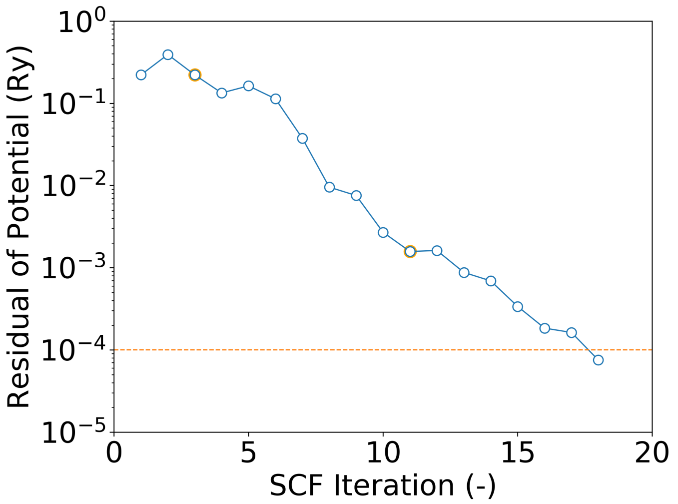
Algorithm 3 Rayleigh–Ritz refinement

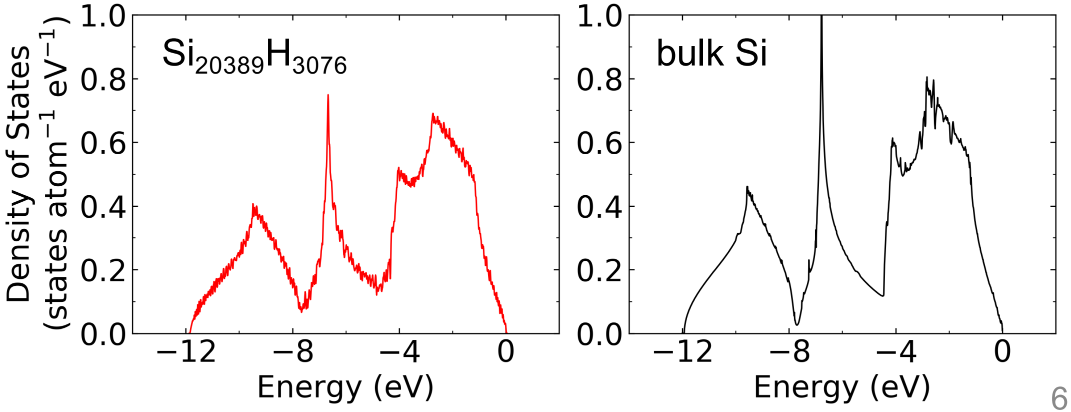
1: **procedure** $(V, D) = \text{RAYLEIGHRITZ}(V)$
2: $\quad A = V^T H V$
3: $\quad$ Compute Q, D such that $AQ = QD$ and $Q^T Q = I$. $\qquad \triangleright$ D has the Ritz values
4: $\quad V \leftarrow VQ$

Still need to diagonalize
a smaller dense matrix

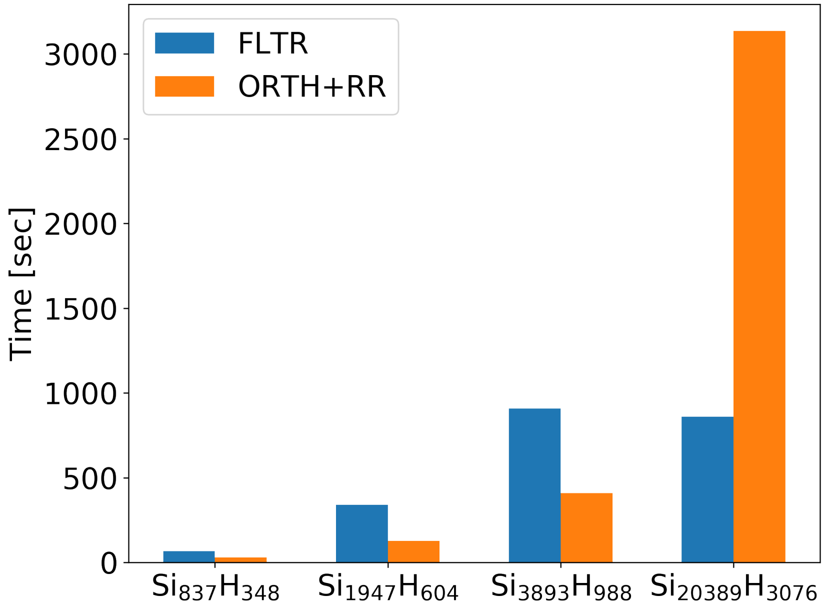
Results

- $Si_{20389}H_{3076}$
- # of occ. states = 42,316
- Cori Haswell
  - 4,096 cores
  - 19 hours





O(N^3) Ops Start to Dominate

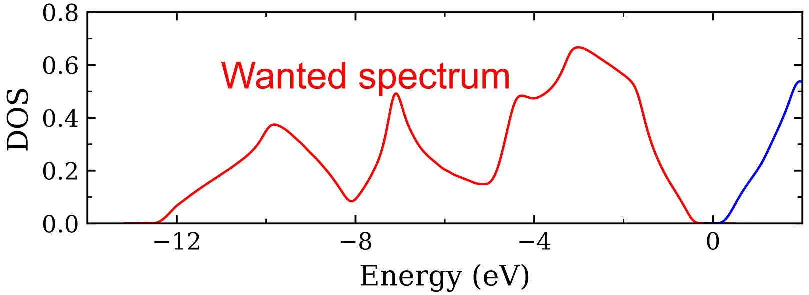


- Filtering O(Nsk)
- Orthonormal. O(Ns^2)
- Rayleigh–Ritz O($Ns^2 + s^3$)

N: # of grid points
s: # of states
k: degrees of filters

Goal: To reduce *s*

Idea of Spectrum Slicing



Chebyshev–Jackson filter

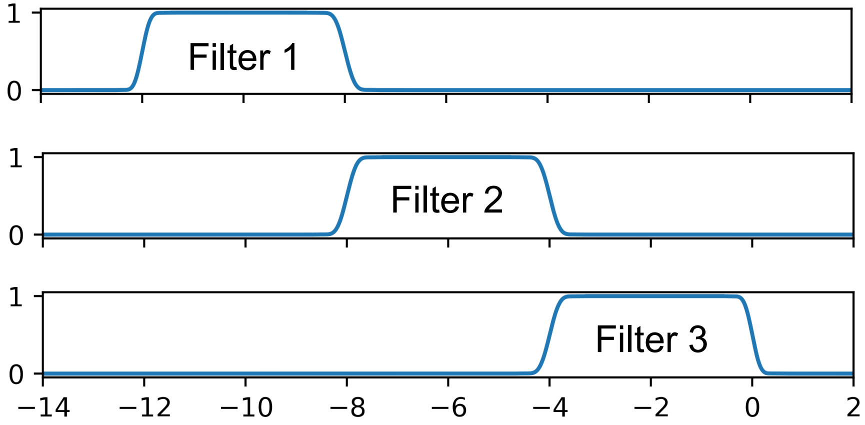
$$\tilde{p}(H_{\mathrm{KS}}) = \sum_{i=0}^{k} \gamma_i(a,b) g_i(k)\, T_i(H_{\mathrm{KS}})$$

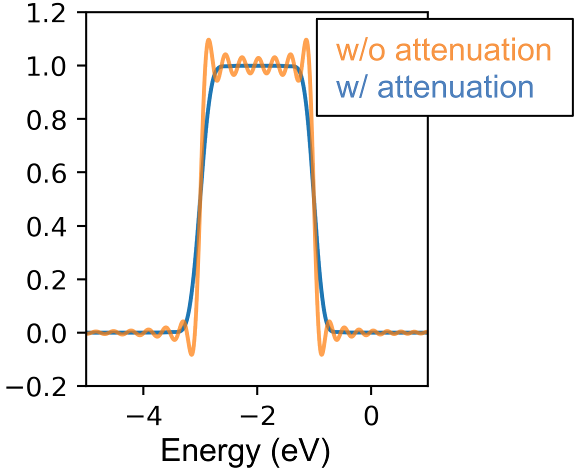
$g_i(k)$: attenuation coefficient

Slice 1

Slice 2

Slice 3





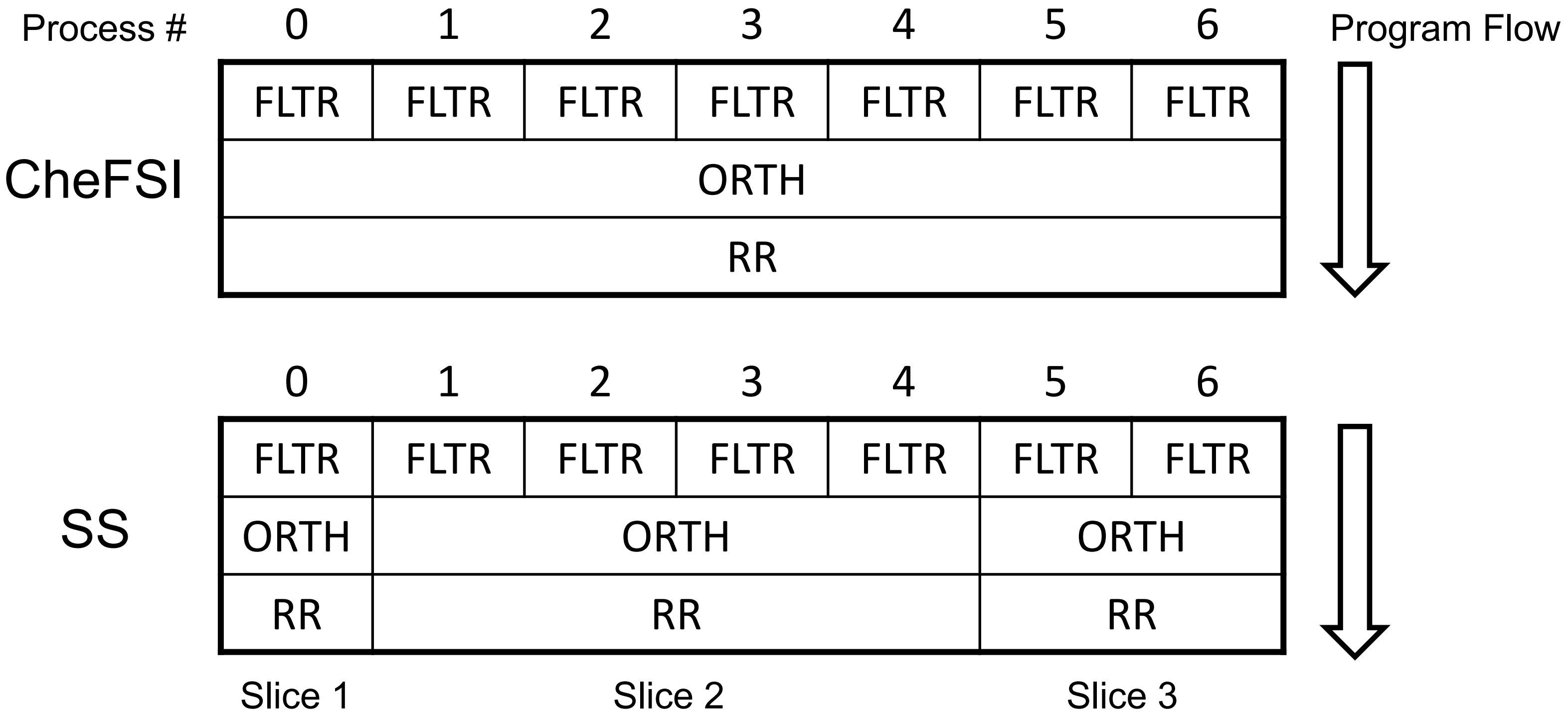
Spectrum Slicing Method

1. Dividing the wanted spectrum into slices
2. Performing polynomial-filtered subspace iteration in parallel
3. Combine spectra and calculate new charge density

- Shifting the burden of diagonalization to cheaper filtering
  - Diagonalization in Rayleigh–Ritz is $O(s^3) \sim O(N^3)$
  - Filtering is $O(Nsk) \sim O(N^2)$

N: # of grid points
s: # of states
k: degrees of filters

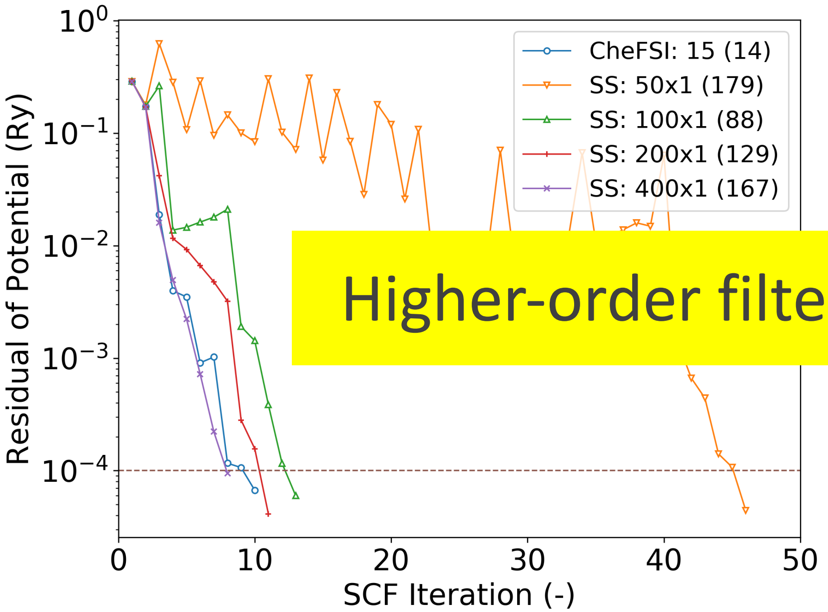
CheFSI vs. Spectrum Slicing

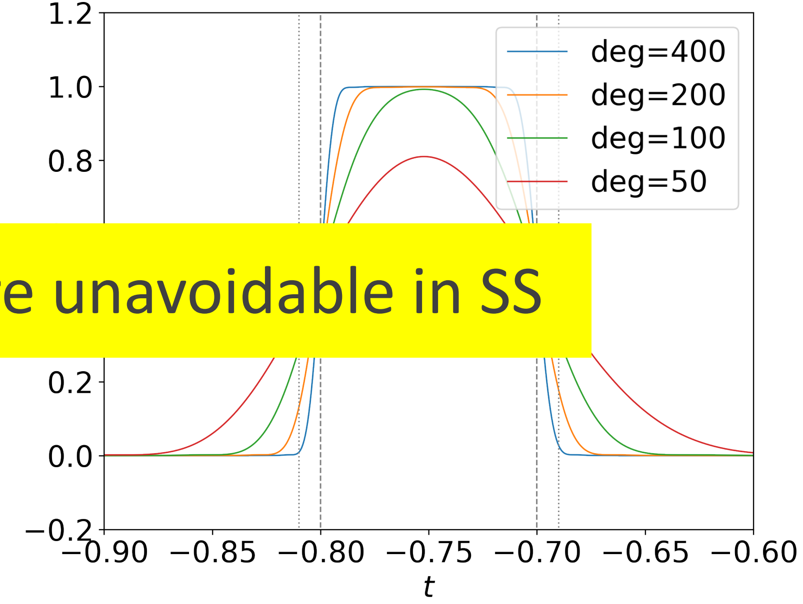


Influence of Degree of Filters

[DOF]x[SI cycles] (Time to solution)

$$\tilde{p}(H_{\mathrm{KS}}) = \sum_{i=0}^{k} \gamma_i(a,b) g_i(k)\, T_i(H_{\mathrm{KS}})$$

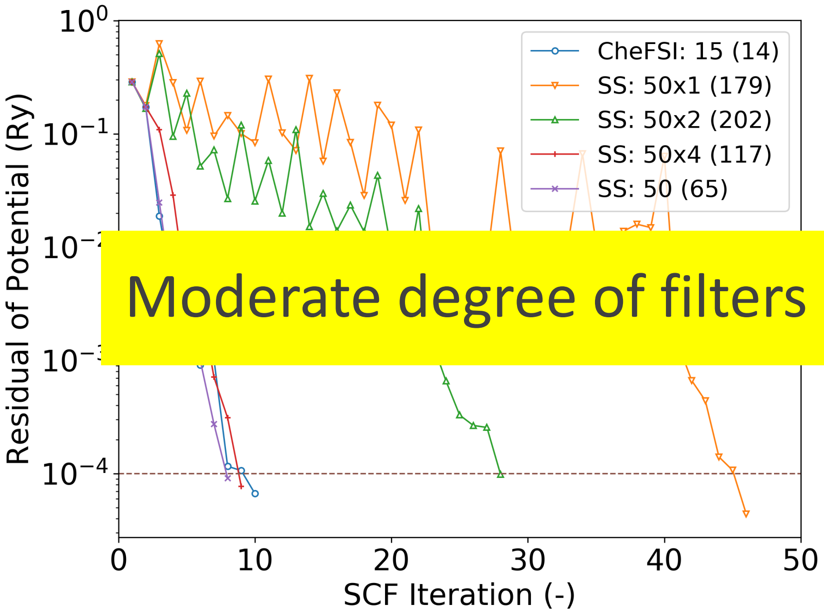




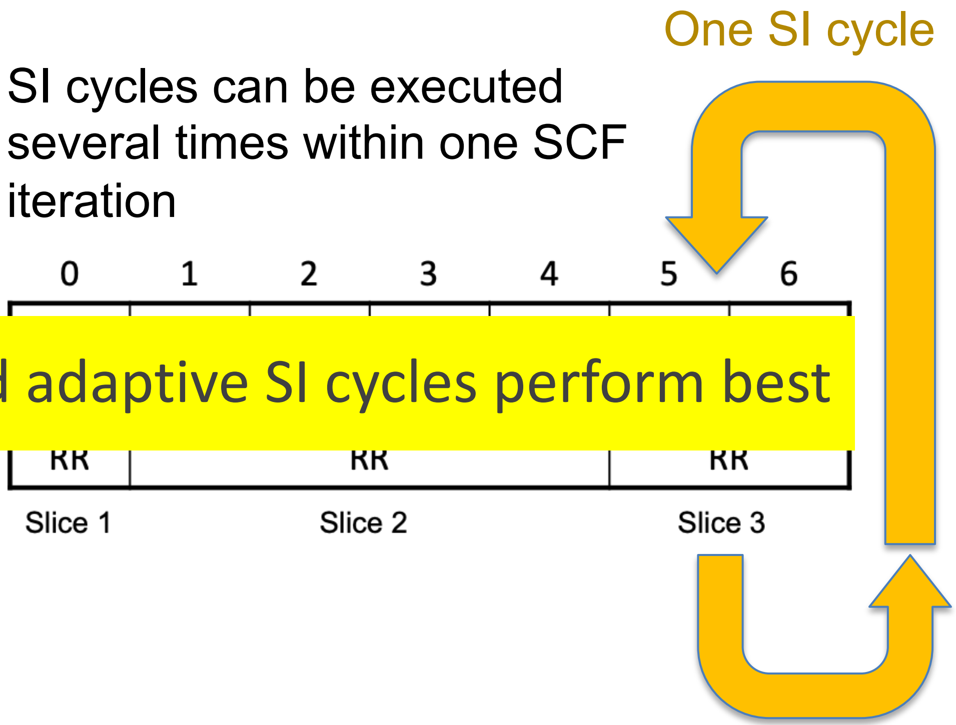
Higher-order filters are unavoidable in SS

Influence of SI cycles

[DOF]x[SI cycles] (Time to solution)



SI cycles can be executed several times within one SCF iteration



Moderate degree of filters and adaptive SI cycles perform best

Summary

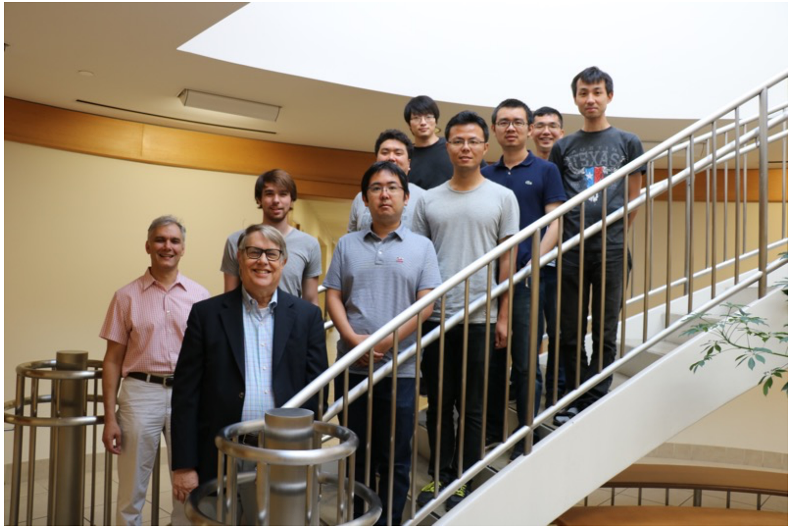
- Electronic structure of a confined system with over 20,000 atoms was solved by Chebyshev-filtered subspace iteration (filtering + Rayleigh–Ritz method)
- A subspace iteration spectrum slicing method is proposed for large electronic structure calculation
- There are more! →

Session H36: Real-Space Methods for Large Scale Electronic Structure Problems
H36.00001
James Chelikowsky
2:30 PM–3:06 PM, Tuesday, March 5, 2019
BCEC Room: **205C**

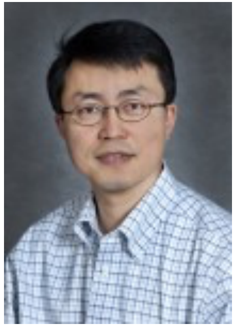
Acknowledgements

U.S. DEPARTMENT OF ENERGY

Office of Science



Center for Computational Materials



Chao Yang

TACC

NERSC