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Localized and Randomized Algorithms for Electronic Structure

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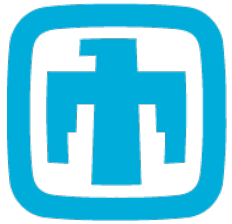

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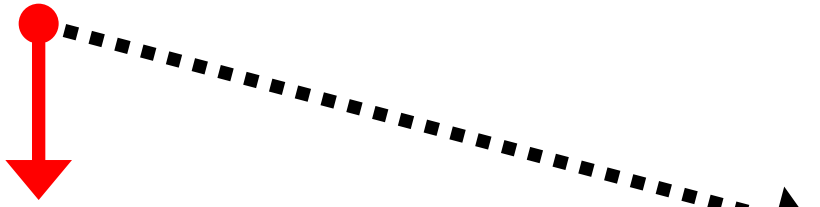
Linear-scaling electronic structure landscape

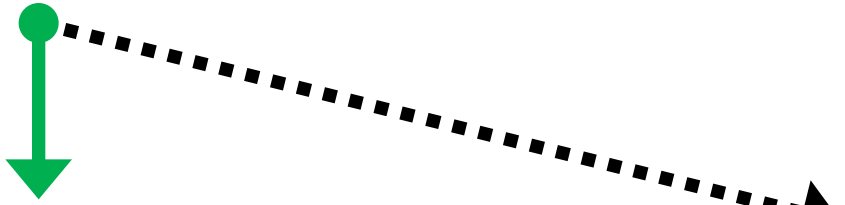


mean-field theory (physics)

[Goedecker, Rev. Mod. Phys. **71**, 1085 (1999)]

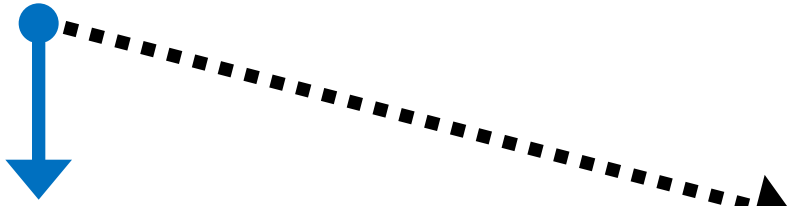
post Hartree-Fock (chemistry)

[Ayala & Scuseria, J. Chem. Phys. **110**, 3660 (1999)]



in primary basis

Wannier basis construction



Fermi-Dirac approximation ($T > 0$)

density-matrix projector ($T = 0$)

Finite-temperature methods

$$\langle X \rangle = \mathrm{tr}[X f(H)]$$

function approximation

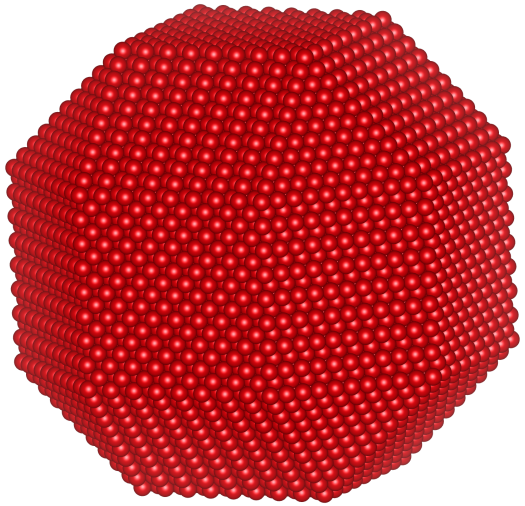
- polynomial: $f(x) \approx \sum_{i=1}^{m} c_i x^{i-1}$
- rational: $f(x) \approx \sum_{i=1}^{m} w_i (x - z_i)^{-1}$
  [Moussa, JCP **145**, 164108 (2016)]
- telescoping
- recursive

trace approximation

- exact: rational + selected inversion (PEXSI)
  [Lin *et al.*, TOMS **37**, 40 (2011)]
- localized: sparse $F \approx f(H)$
- randomized: $\mathrm{tr}[X f(H)] \approx \frac{1}{s} \sum_{i=1}^{s} y_i^\dagger X f(H) y_i$
- hybrid: $\mathrm{tr}[XF] + \frac{1}{s} \sum_{i=1}^{s} y_i^\dagger X [f(H) - F] y_i$
  or colored rotors [Wang *et al.*, **arXiv:1711.10570** (2017)]
  or embedded fragments [Arnon *et al.*, JCP **146**, 224111 (2017)]

Test problem

system:



copper clusters (unrelaxed)

@ T = 0.03 eV  ambient

⋮

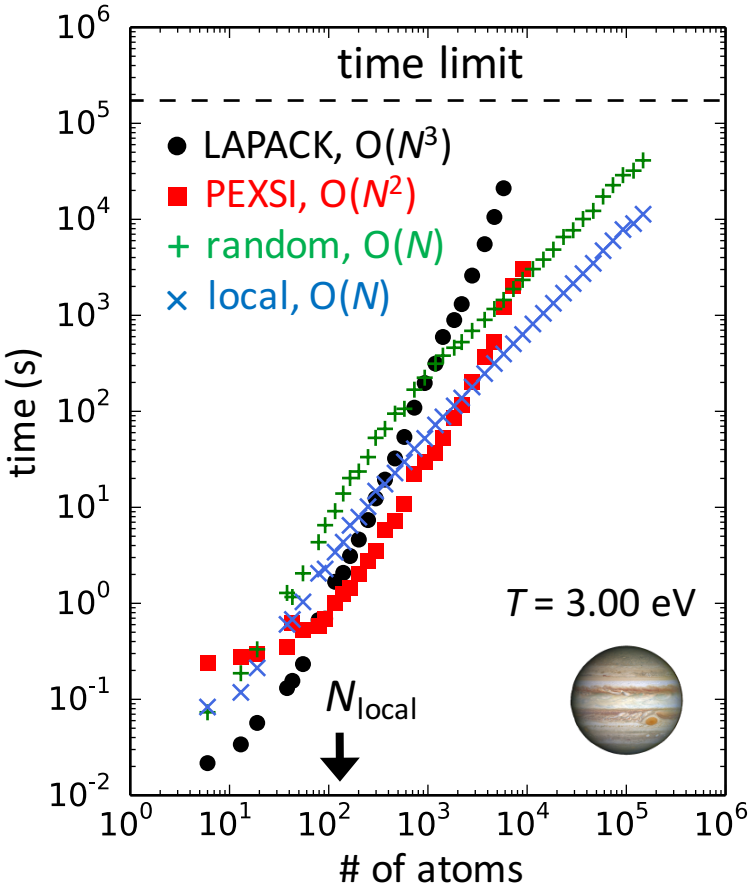
T = 3.00 eV  warm dense matter

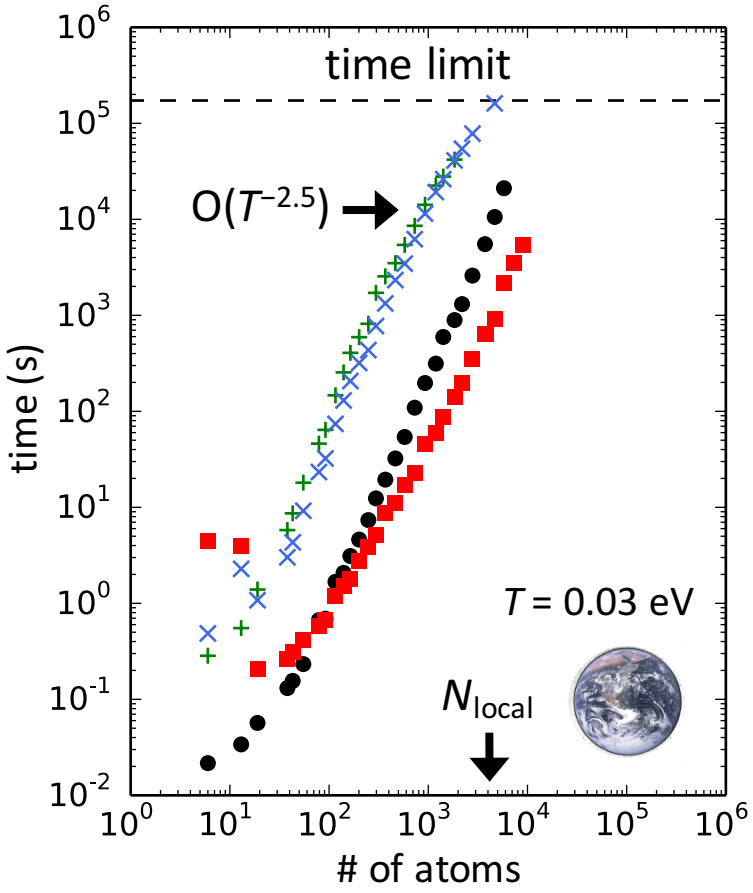
model: NRL tight binding w/ *spd* basis (9 orbitals per atom)
[Mehl & Papaconstantopoulos, PRB **54**, 4519 (1996)]

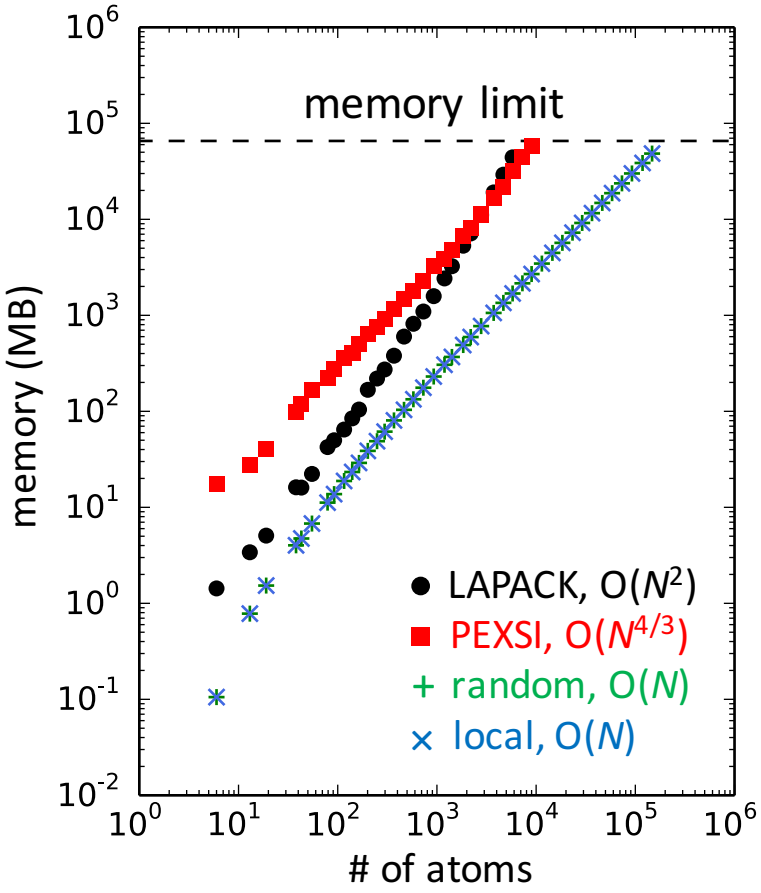
accuracy:

number	energy	force	stress
0.01 #/atom	0.01 eV/atom	0.01 eV/A	0.1 GPa

Benchmarks





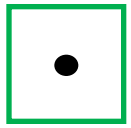


@ 1 node, 16 cores, 0.35 Tflop/s, 64 GB memory (w/ OpenMP)

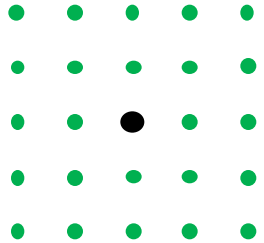
Lessons learned (little change since 1999)

- Useful preconditioner needed for rational approximation

- Hybridization failure:
  Poisson equation example



local errors

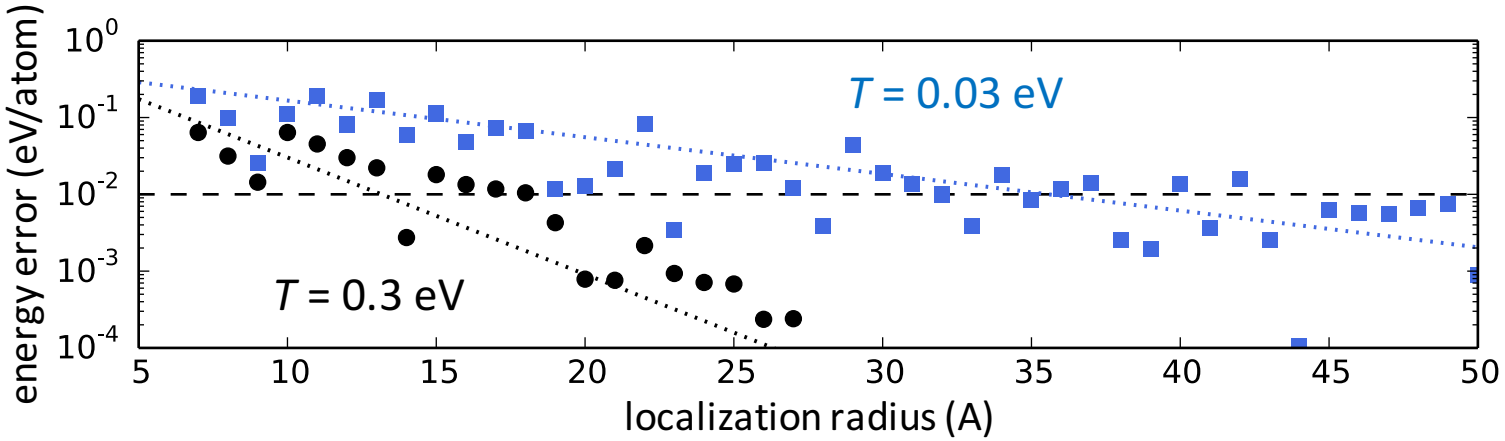


local+random errors

image charge
error analysis

- Weak localization:

$$\epsilon \propto \exp(-\sqrt{T}R_{\mathrm{loc}})$$



Localization self-energy

Truncation:

independent columns

$$\begin{bmatrix} H_{\mathrm{loc}} & \cancel{H_{\mathrm{int}}^{\dagger}} \\ \cancel{H_{\mathrm{int}}} & H_{\mathrm{env}} \end{bmatrix} \begin{bmatrix} g_{\mathrm{loc}} \\ \cancel{g_{\mathrm{env}}} \end{bmatrix} = \begin{bmatrix} e \\ 0 \end{bmatrix}$$

Embedding:

$$\begin{bmatrix} H_{\mathrm{loc}} + \Sigma_{\mathrm{loc}} & 0 \\ 0 & H_{\mathrm{env}} \end{bmatrix} \begin{bmatrix} g_{\mathrm{loc}} \\ 0 \end{bmatrix} = \begin{bmatrix} e \\ 0 \end{bmatrix}$$

Environment modeling:

$$\Sigma_{\mathrm{loc}} = -H_{\mathrm{int}}^{\dagger} H_{\mathrm{env}}^{-1} H_{\mathrm{int}}$$

Environment approximation

Localization: $$\widetilde{G} = \begin{bmatrix} \widetilde{G}_{\mathrm{loc}} & \widetilde{G}_{\mathrm{int}}^{\dagger} \\ \widetilde{G}_{\mathrm{int}} & \widetilde{G}_{\mathrm{env}} \end{bmatrix} \approx P \circ G$$

sparsity pattern

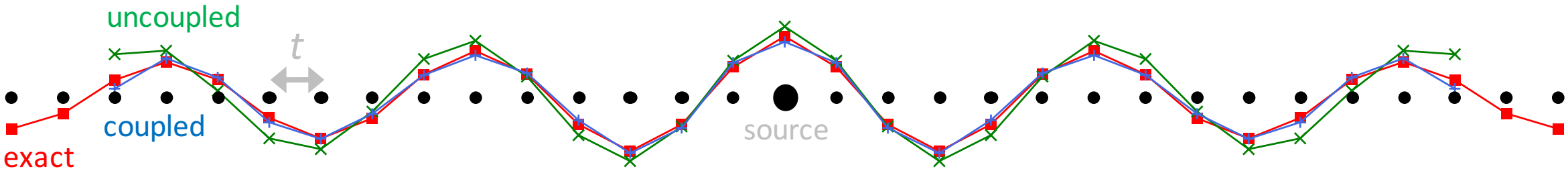
Uncoupled: $$P \circ (I - H\widetilde{G}) = 0 \rightarrow \Sigma_{\mathrm{loc}} \approx 0$$

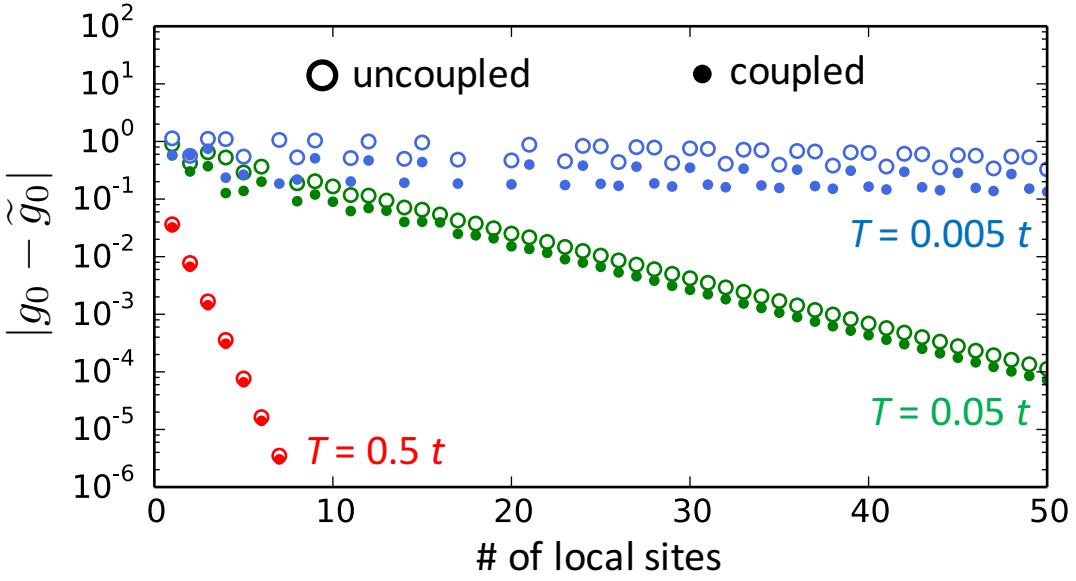
Coupled: $$P \circ (\widetilde{G} - \widetilde{G}H\widetilde{G}) = 0 \rightarrow \Sigma_{\mathrm{loc}} \approx \widetilde{G}_{\mathrm{loc}}^{-1}\widetilde{G}_{\mathrm{int}}^{\dagger}H_{\mathrm{int}}$$

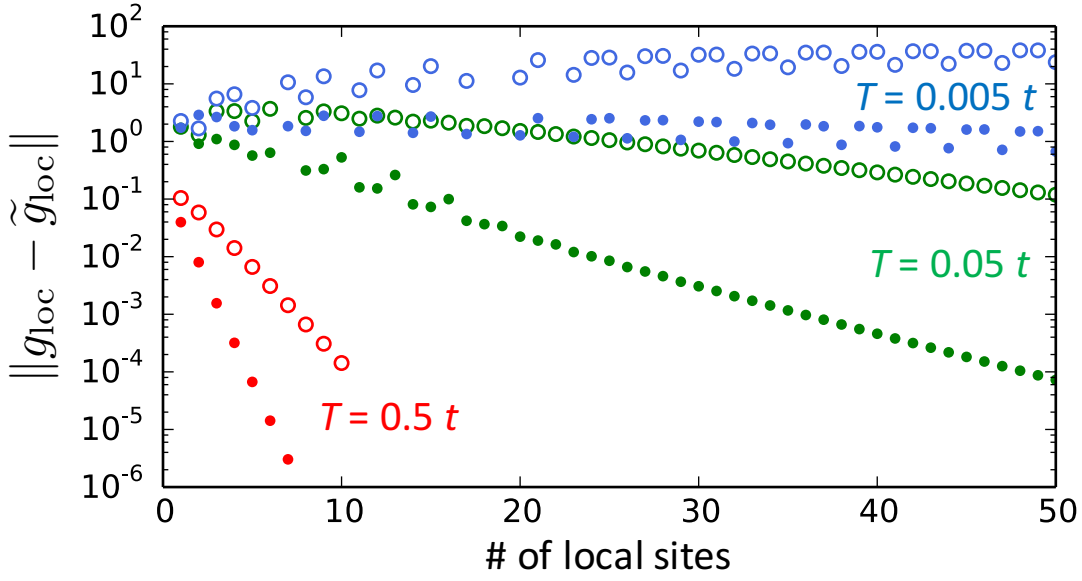
non-linear self-preconditioning

exact **Σ** for exact ***G***

1D tight-binding example







Future of linear-scaling electronic structure?

- Long overdue for warm dense matter

- Large gap in accuracy/cost curve: (semi-empirical opportunity?)

  machine-learned interatomic potential: **0.01** s/atom/time-step [Bartok *et al.*, PRL **104**, 136403 (2010)]

  large-basis localized-Wannier DFT: **60** s/atom/time-step [Osei-Kuffuor & Fattebert, PRL **112**, 046401 (2014)]

- Limited use for randomized methods:

  SCF-free Langevin dynamics?

  randomized linear algebra?

- **Software will be available w/ preprint (to benchmark new ideas)**