Numerical Methods for DFT and Beyond

Ville Havu

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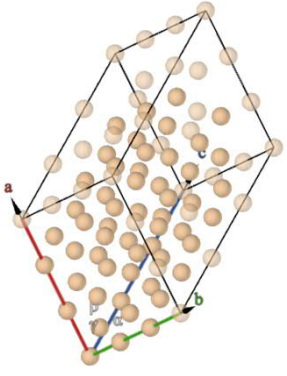
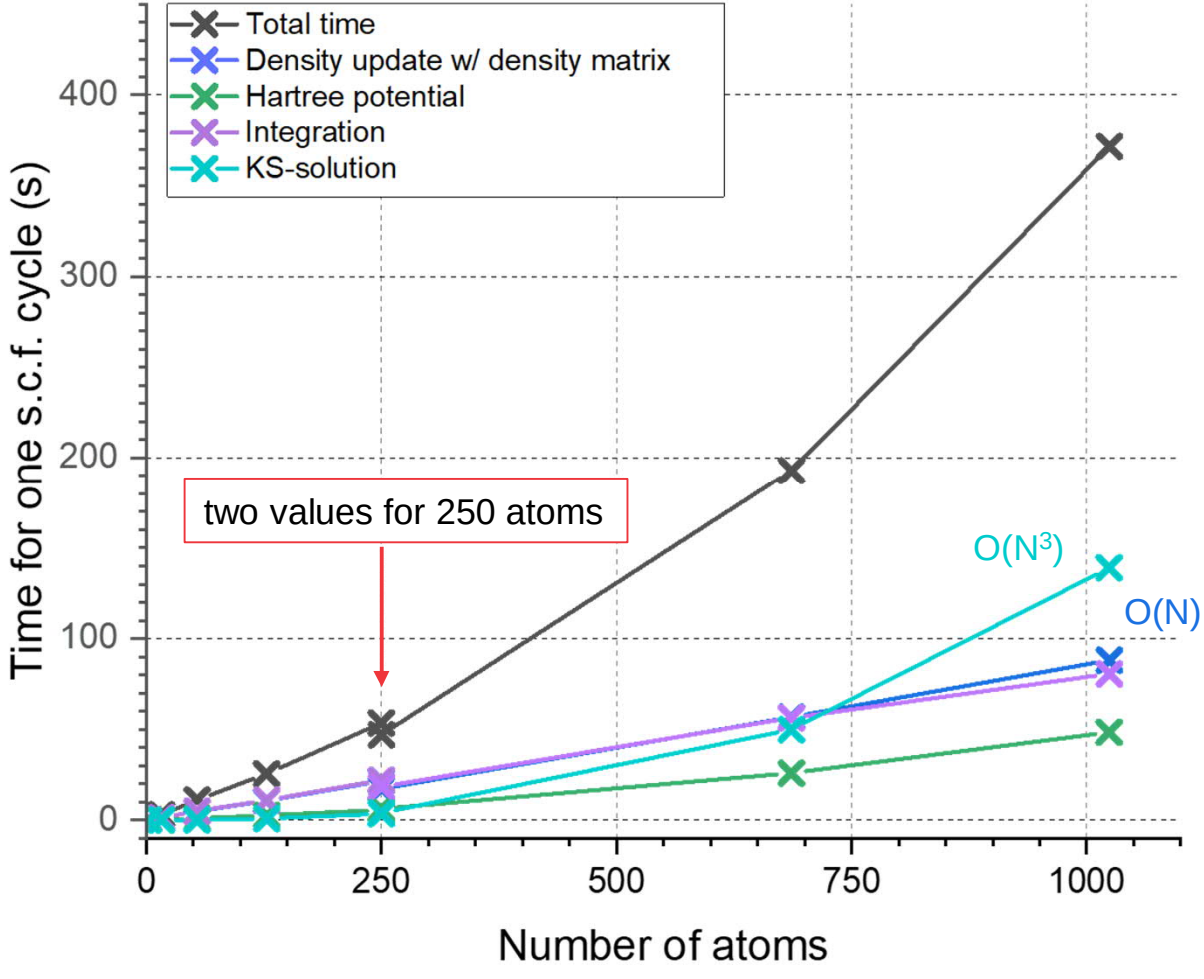
First look

Bulk Si, tight settings, 2 – 1024 atoms in a unit cell, 1 k-point, 128 MPI-tasks (2 x 64 AMD Rome), 256 GB memory

gcc, OpenMPI, OpenBLAS, ScaLAPACK





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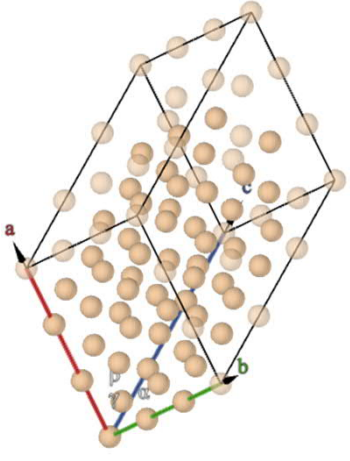
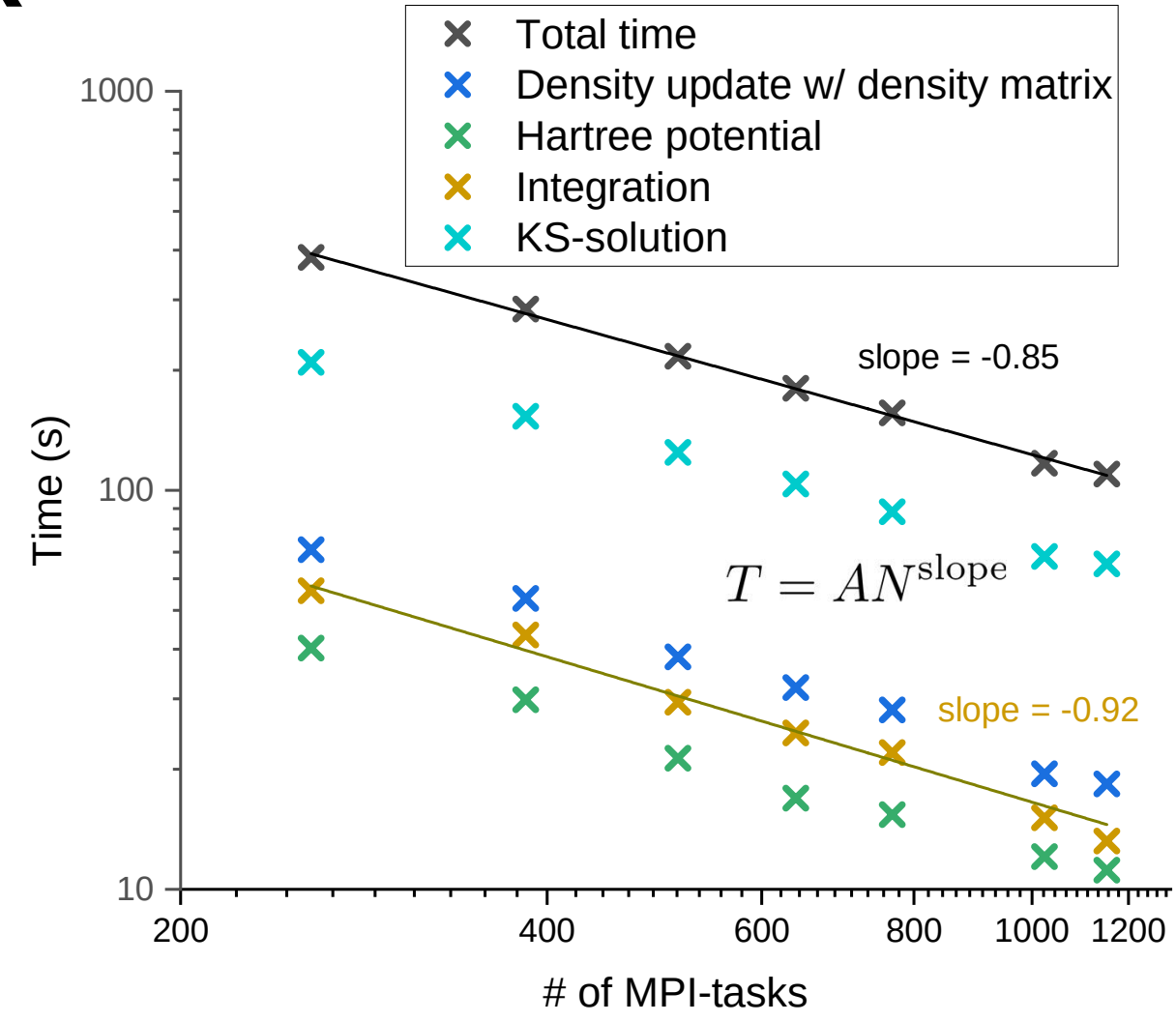
Second look

Bulk Si, tight settings, 1458 atoms
in a unit cell
256 - 1152 MPI-tasks





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Integration in DFT

Integration – calculation of matrix elements

$$h_{ij} = \int d\mathbf{r}\, \varphi_i(\mathbf{r}) h_{KS}(\mathbf{r}) \varphi_j(\mathbf{r}) \qquad h_{KS}(\mathbf{r}) = -\frac{1}{2}\Delta + V_{\text{eff}}(\mathbf{r})$$

$$S_{ij} = \int d\mathbf{r}\, \varphi_i(\mathbf{r}) \varphi_j(\mathbf{r})$$

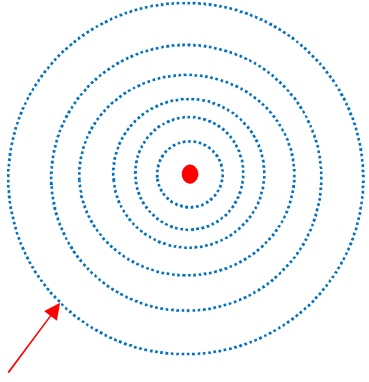
Gaussian basis functions → many analytic formulae available

Plane wave basis functions → fast Fourier transform does the trick

Numerical atom centered basis functions → need atom centered grids

$$h_{ij} = \sum_k \varphi_i(\mathbf{r}_k) h_{KS}(\mathbf{r}_k) \varphi_j(\mathbf{r}_k) w_k$$

$$s_{ij} = \sum_k \varphi_i(\mathbf{r}_k) \varphi_j(\mathbf{r}_k) w_k$$

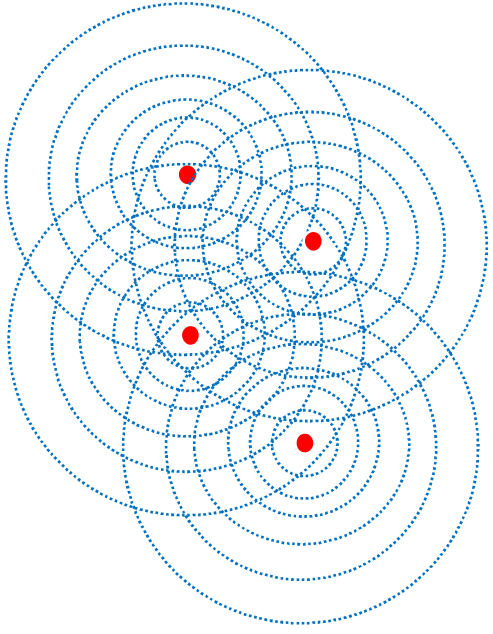


Grid points around the atom, location $\mathbf{r}_k$, weight w_k

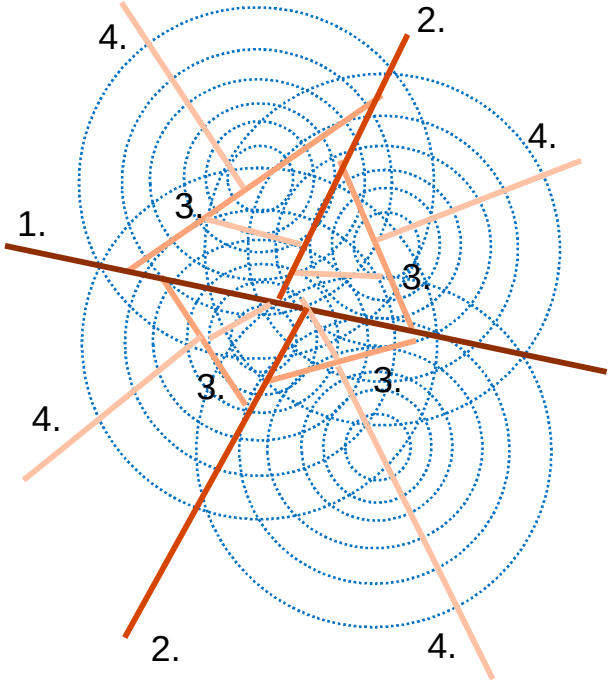
Grid-based integration

Many atoms, large overlapping grids, need to consider integration points in batches

Overlapping grid



Split recursively into batches of equal size



In each batch:

1. Compute the matrices

$$M_{jk} = \varphi_j(\mathbf{r}_k)\sqrt{w_k}$$

$$N_{ik} = h_{KS}(\mathbf{r}_k)\varphi_i(\mathbf{r}_k)\sqrt{w_k}$$

2. Sum up the local contribution

$$h_{ij,loc} = \sum_k M_{jk}N^*_{ik}$$

When all batches done sum up:

$$h_{ij} = \sum h_{ij,loc}$$

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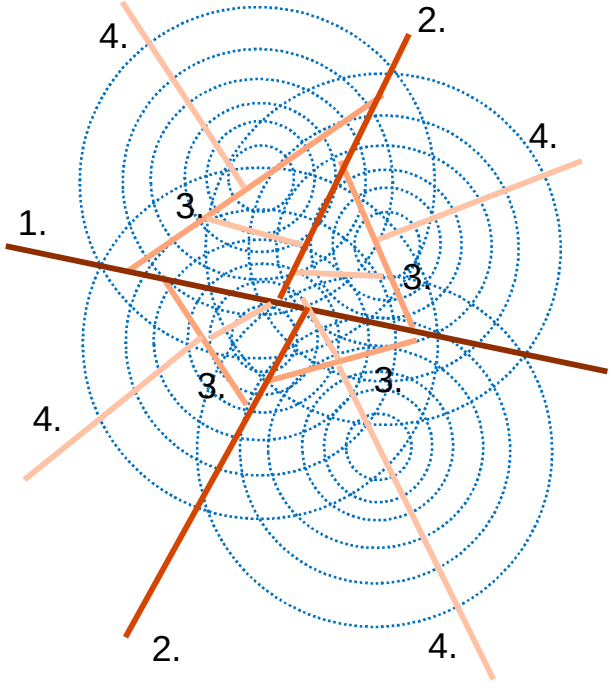
Havu et. at. *Journal of Computational Physics* **228**, 8367-8379

Grid-based integration

Many atoms, large overlapping grids, need to consider integration points in batches



<u>In each batch:</u>

1. Compute the matrices

$$M_{jk} = \varphi_j(\mathbf{r}_k)\sqrt{w_k}$$

$$N_{ik} = h_{KS}(\mathbf{r}_k)\varphi_i(\mathbf{r}_k)\sqrt{w_k}$$

Need to evaluate only non-zero basis functions in each batch

2. Sum up the local contribution

$$h_{ij,loc} = \sum_k M_{jk} N^*_{ik}$$

Matrix-matrix product (BLAS3 operation)

<u>When all batches done</u> sum up:

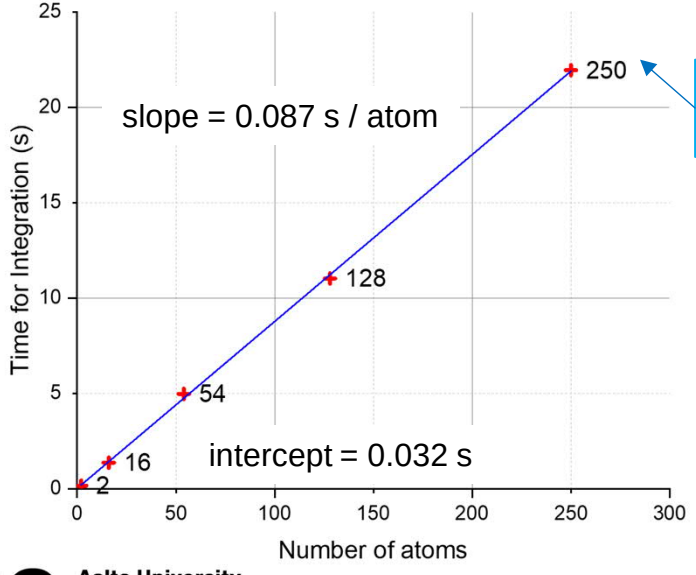
$$h_{ij} = \sum h_{ij,loc}$$

MPI_Allreduce makes operation parallel

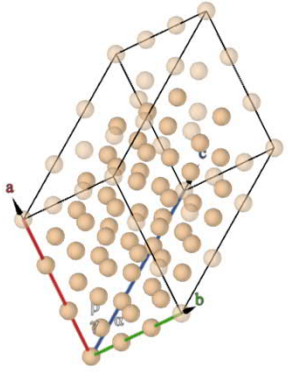
Complexity: O(*N*) points, O(1) functions / batch → O(*N*) in total

Performance and scalability: basic algorithm

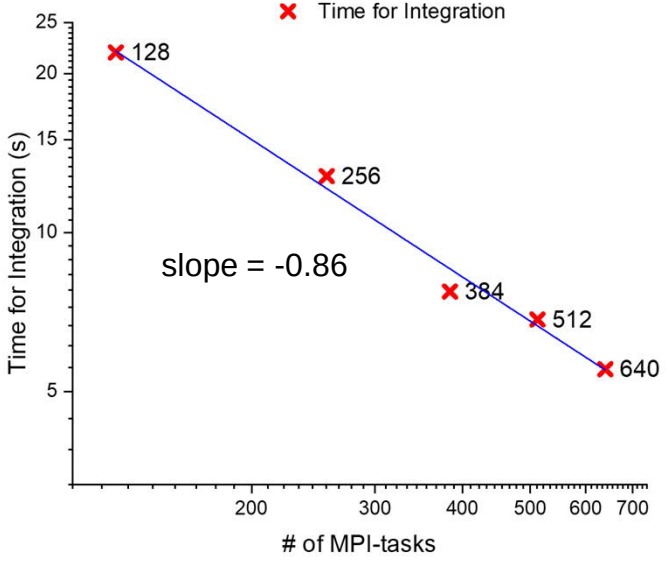
Bulk Si, tight settings, 2 – 250 atoms in a unit cell
128 MPI-tasks (2 x 64 AMD Rome), 256 GB memory, single s.c.f. iteration



out of memory for the next unit cell



Bulk Si, tight settings, 250 atoms in a unit cell
128 – 640 MPI-tasks, single s.c.f. iteration



Improving integration with local basis storage and load balancing

The basic algorithm works well in many cases but can be improved

Per batch for each MPI-task:

1. Compute the matrices

$$M_{jk} = \varphi_j(\mathbf{r}_k)\sqrt{w_k}$$
$$N_{ik} = h_{KS}(\mathbf{r}_k)\varphi_i(\mathbf{r}_k)\sqrt{w_k}$$

2. Sum up the local contribution

$$\tilde{h}_{ij,loc} = \sum_k M_{jk}N^*_{ik}$$

When all batches done sum up:

$$h_{ij} = \sum \tilde{h}_{ij,loc}$$

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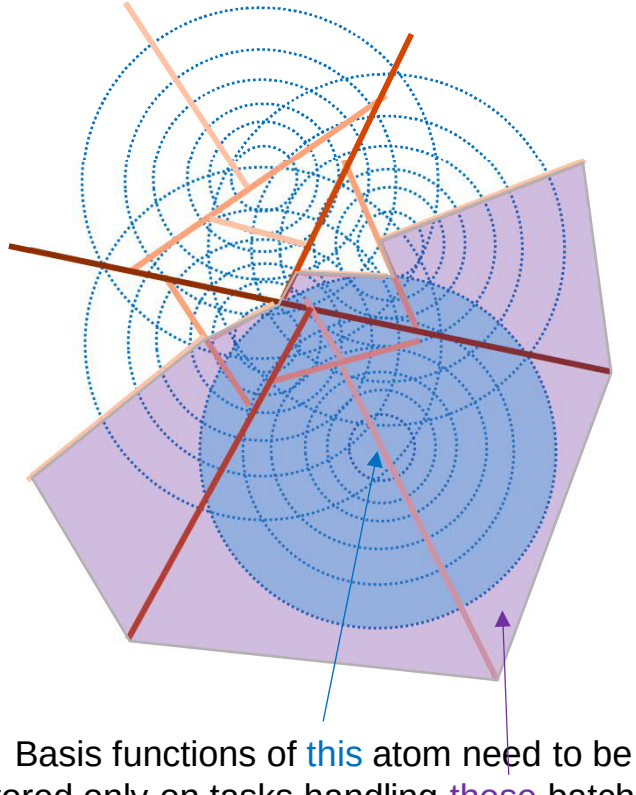
Store only the elements relevant to the MPI-task doing the integration in $h_{ij,loc}$ → no global matrix storage. It follows:

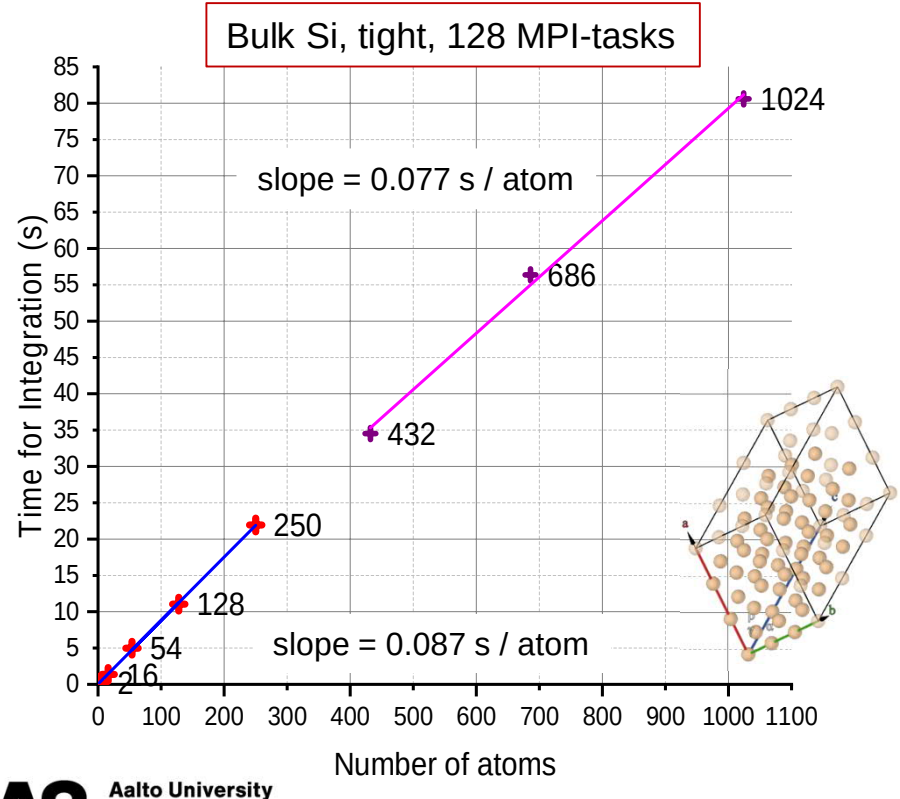
1. Batches need to be distributed to MPI-tasks **by location**, not by load
2. Load imbalance can follow to be compensated by **load balancing**
3. Drawback: some more exotic functionality may lack support

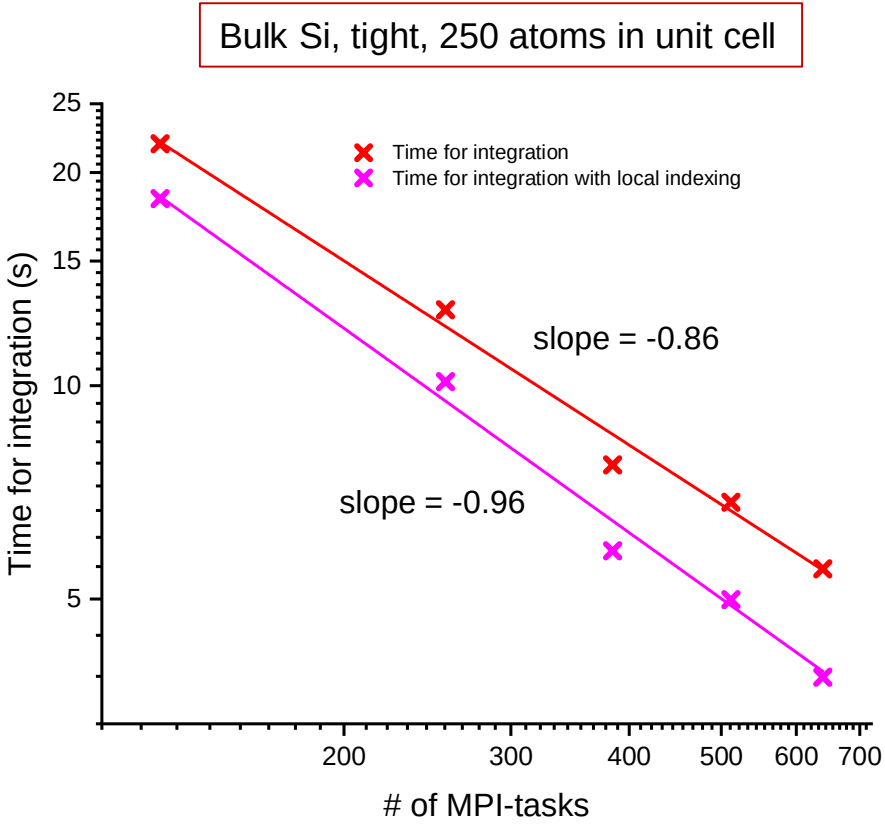
```
use_local_index
load_balancing
```



Basis functions of this atom need to be stored only on tasks handling these batches

Improving integration with local basis storage and load balancing





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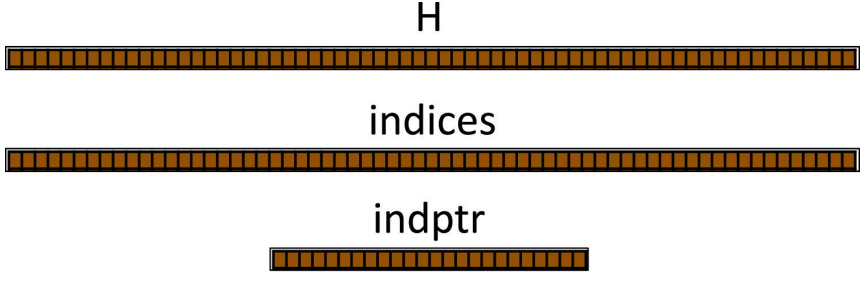
Local indexing storage schemes

Global sparse matrix storage

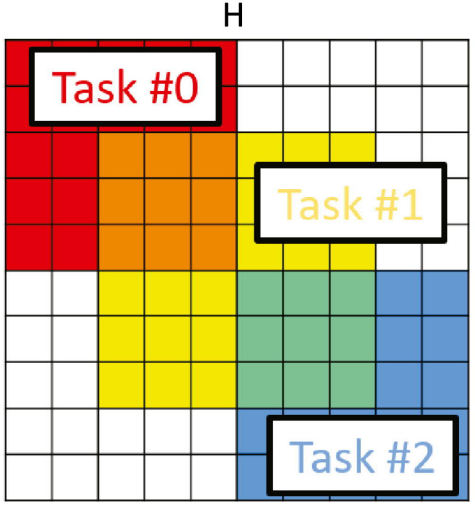
- All MPI-tasks store the same matrix
- Sparse storage format
  - Entries
  - Row indices of entries
  - Column pointers to row indices
- Storage cost O(*N*)
- Simply synchronization with MPI_AllReduce

Local dense storage

- MPI-tasks store only elements relevant to its batches
- Dense storage format
  - Larger system – storage cost up
  - More MPI-tasks – storage cost down
- Complex synchronization with point-to-point communication



(a) Globally-Indexed Sparse



(b) Locally-Indexed Dense

Huhn et al. *Comp. Phys. Comm.*, **254**, 107314

Density update

The density at each grid point follows from the single-particle orbitals: $n(\mathbf{r}_k) = \sum_n f_n |\psi_n(\mathbf{r}_k)|^2$

O(*N*) orbitals x O(*N*) grid points = O(N^2) if calculated directly

But $\psi_n(\mathbf{r}) = \sum_i c_n^i \varphi_i(\mathbf{r})$ and it follows

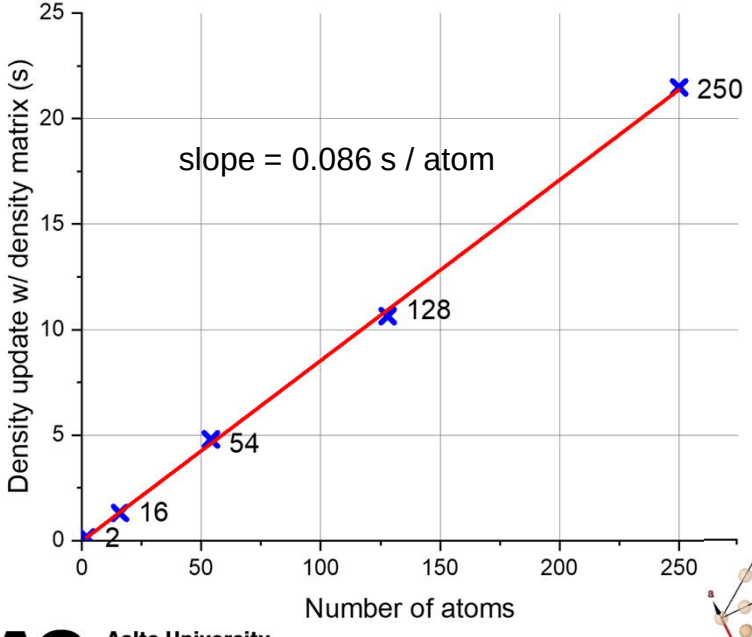
$$n(\mathbf{r}_k) = \sum_n f_n \sum_i c_n^{i*} \varphi_i(\mathbf{r}_k) \sum_j c_n^j \varphi_j(\mathbf{r}_k) = \sum_{ij} \varphi_i(\mathbf{r}_k)\varphi_j(\mathbf{r}_k) \sum_n f_n c_n^{i*} c_n^j$$

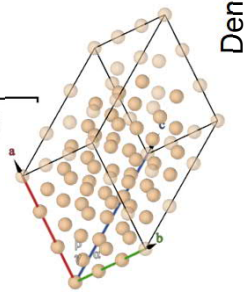
O(1) for localized basis $= D_{ij}$

Formally O(N^3) but
- a very small prefactor
- D_{ij} is sparse

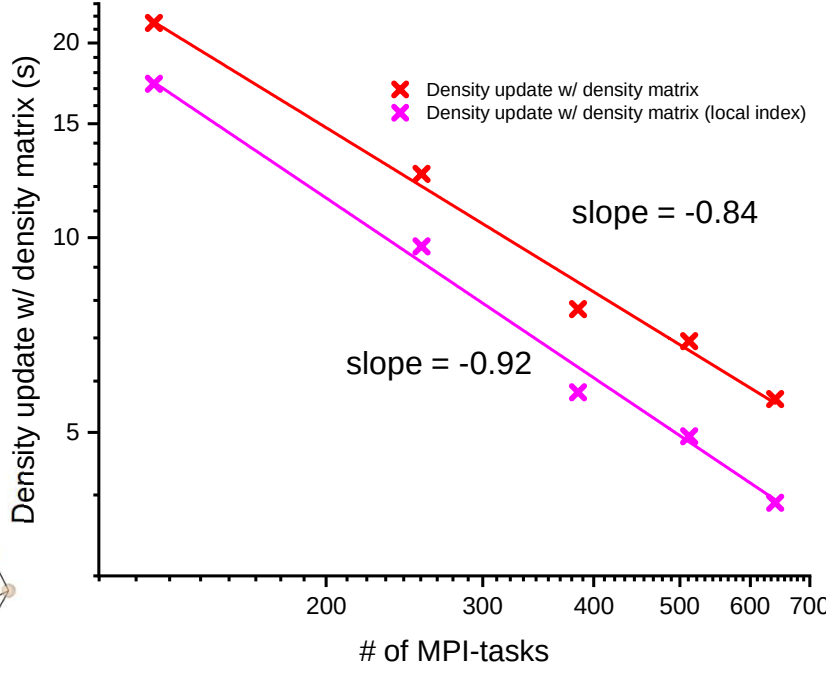
Performance and scalability

Bulk Si, tight settings, 2 – 250 atoms in a unit cell
128 MPI-tasks, single s.c.f. iteration





Bulk Si, tight, 250 atoms in unit cell, single s.c.f. iteration



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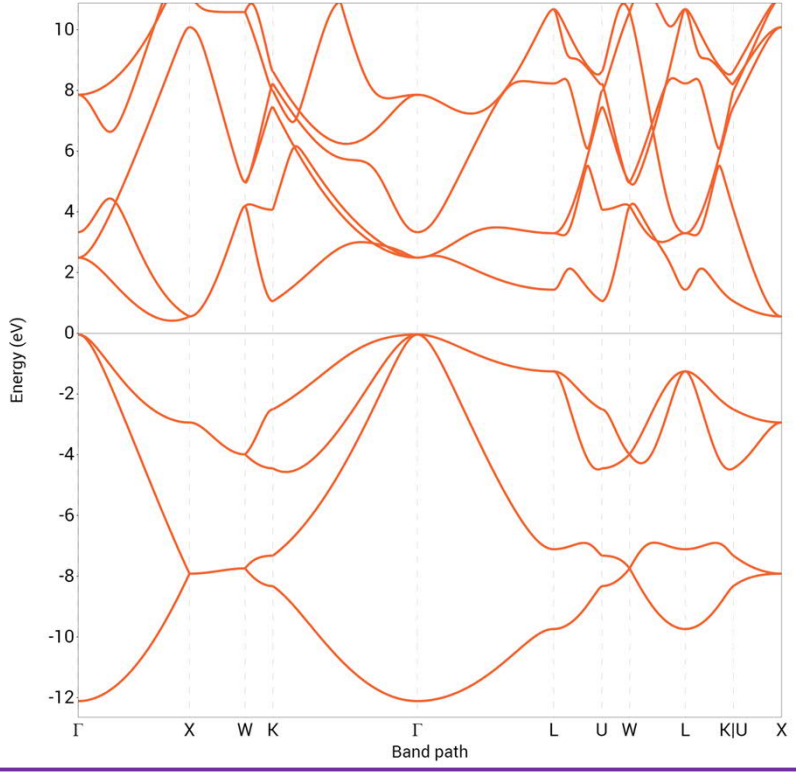
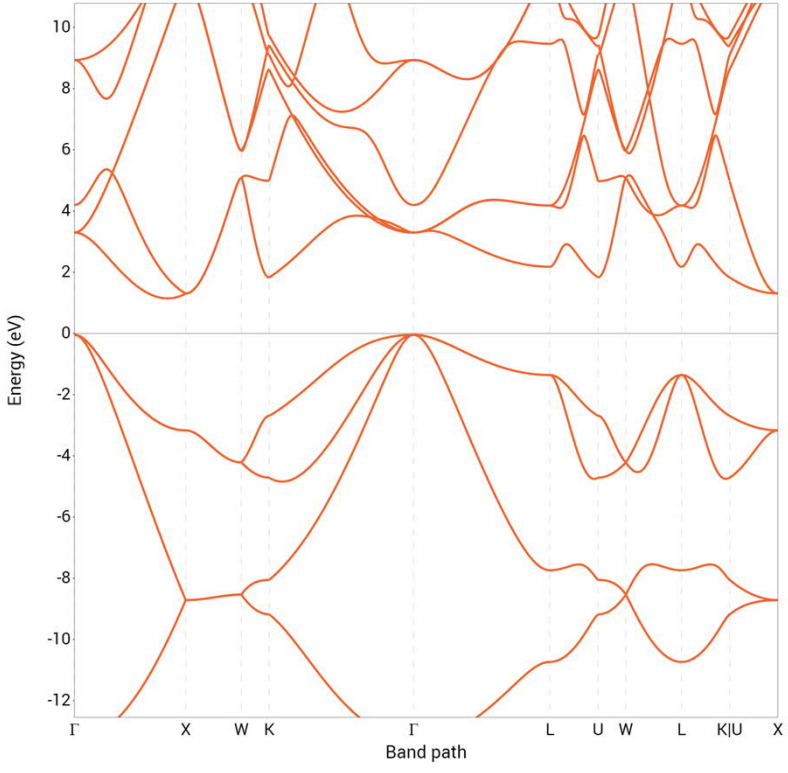
Beyond DFT – local is not enough

Si-bulk, LDA, tight, 15x15x15 k-grid: E_g = 0.466 eV



Si-bulk, HSE06, tight, 12x12x12 k-grid: E_g = 1.188 eV



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Input files and output file analysis made with GIMS: https://gims.ms1p.org/

Beyond DFT – the exchange operator

To go beyond local and semilocal approximations one very often needs the exchange operator:

$$\hat{K}\psi_n(\mathbf{r}) = \sum_m f_m \int \frac{\psi_m^*(\mathbf{r}')\psi_n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\, d\mathbf{r}'\psi_m(\mathbf{r})$$

In basis representation, that is:

$$K_{ij} = \sum_{kl}(ik|lj)D_{kl} \qquad (ij|kl) = \iint \frac{\varphi_i^*(\mathbf{r})\varphi_j(\mathbf{r})\varphi_k^*(\mathbf{r}')\varphi_l(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\, d\mathbf{r}d\mathbf{r}'$$

Resolution of identity

Four center integrals (ij | kl) are very expensive. Need to reduce costs. $$(ij|kl) = \iint \frac{\varphi_i^*(\mathbf{r})\varphi_j(\mathbf{r})\varphi_k^*(\mathbf{r}')\varphi_l(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\, d\mathbf{r}d\mathbf{r}'$$

Pair products are too many, expand in auxiliary functions (resolution of identity (RI), variational density fitting):

$$\varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) = \sum_\mu C_{ij}^\mu P_\mu(\mathbf{r})$$

→

$$\begin{cases} (ij|kl) = \sum_{\mu\nu} C_{ij}^\mu V_{\mu\nu} C_{kl}^\nu \\ V_{\mu\nu} = \iint \frac{P_\mu(\mathbf{r})P_\nu(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} \end{cases}$$

Four centers becomes two indices. Need to:

1. Select the auxiliary basis functions
2. Define, how to compute the coefficients C_{ij}

Resolution of identity

$$\varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) = \sum_\mu C_{ij}^\mu P_\mu(\mathbf{r})$$

Four centers becomes two indices. Need to:

1. Select the auxiliary basis functions
2. Define, how to compute the coefficients C_{ij}

2. To compute the coefficients, minimize:

$$\delta\rho_{ij}(\mathbf{r}) = \varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) - \sum_\mu C_{ij}^\mu P_\mu(\mathbf{r})$$

Option a: minimize the norm $(\delta\rho_{ij}\,\delta\rho_{ij})$

$$C_{ij}^\mu = \sum_\nu (ij\nu) S_{\nu\mu}^{-1} \qquad (ij\nu) = \int \varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) P_\nu(\mathbf{r})\, d\mathbf{r}$$

$$S_{\mu\nu} = \int P_\nu(\mathbf{r}) P_\mu(\mathbf{r})\, d\mathbf{r}$$

RI-SVS

Option b: minimize the norm $(\delta\rho_{ij}|\delta\rho_{ij})$

$$C_{ij}^\mu = \sum_\nu (ij|\nu) V_{\nu\mu}^{-1} \qquad (ij|\nu) = \iint \frac{\varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) P_\nu(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\, d\mathbf{r} d\mathbf{r}'$$

$$V_{\mu\nu} = \iint \frac{P_\mu(\mathbf{r}) P_\nu(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$$

RI-V

Method of choice due to accuracy and reliability

$$K_{ij} = \sum_{kl}\sum_{\mu\nu} (ik|\mu) V_{\mu\nu}^{-1} (\nu|jl) D_{kl} = \sum_\mu \sum_{kl} \left(\sum_\nu C_{ik}^\nu V_{\nu\mu}^{1/2} \right) \left(\sum_\nu C_{jl}^\nu V_{\nu\mu}^{1/2} \right) D_{kl}$$

Ren et al. *New J. Phys.* **14** 053020
Ihrig et al. *New J. Phys.*, **17,** 093020
Levchenko et al. *Comp. Phys. Comm.*, **192,** 60

Resolution of identity

$$\varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) = \sum_\mu C_{ij}^\mu P_\mu(\mathbf{r})$$

Four centers becomes two indices. Need to:

1. Select the auxiliary basis functions
2. Define, how to compute the coefficients C_{ij}

1. Construct the auxiliary basis with the products of the orbital basis functions:

a. form on site-products of the radial functions $u_{sk_1l_1}(r)\,u_{sk_2l_2}(r)$
   - (optional) add radial functions to the construction of auxiliary basis only (`for_aux` keyword)

b. remove linear dependencies

c. add angular components with spherical harmonics $Y_{lm}(\phi,\theta)$

atom-centered auxiliary basis

$$\{P_\mu(\mathbf{r})\}$$

Ren et al. *New J. Phys.* **14** 053020
Ihrig et al. *New J. Phys.*, **17,** 093020
Levchenko et al. *Comp. Phys. Comm.,* **192,** 60

Localized resolution of identity

$$\varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) = \sum_\mu C_{ij}^\mu P_\mu(\mathbf{r})$$

atom-centered auxiliary basis

$$\{P_\mu(\mathbf{r})\}$$

There is still a problem: localized basis functions are expanded over all atoms: C_{ij}^{μ} is dense

Fix with localized RI: require $C_{ij}^\mu = 0, \text{ for } \mu \notin \mathcal{P}(IJ)$

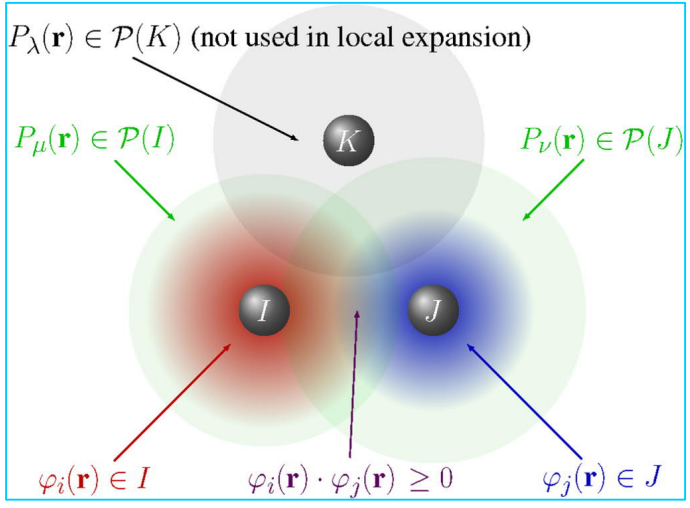
Expand the pair *(i,j)* using only auxiliary functions on the same atoms

$$C_{ij}^\mu = \sum_{\nu \in \mathcal{P}(IJ)} (ij|\nu) L_{\nu\mu}^{IJ}$$

$$L_{\nu\mu}^{IJ} = (V^{IJ})_{\nu\mu}^{-1}$$

RI-LVL

Better scaling implementation of RI-V



Ihrig et al. *New J. Phys.*, **17,** 093020

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`RI_method` defaults to
- `lvl` for HF and hybrid functionals
- `v` for MP2, RPA and GW

Periodic RI

Block-like basis functions $\varphi_{i\mathbf{k}}(\mathbf{r}) = \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} \varphi_i(\mathbf{r}-\mathbf{R})$

In reciprocal space, Fourier transform:

$$K_{ij}(\mathbf{k}) = \frac{1}{N_\mathbf{q}} \sum_{kl,\mathbf{q}} D_{kl}(\mathbf{q}) \iint \frac{\varphi^*_{i\mathbf{k}}(\mathbf{r})\varphi_{k\mathbf{q}}(\mathbf{r})\varphi^*_{l\mathbf{q}}(\mathbf{r}')\varphi_{j\mathbf{k}}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}d\mathbf{r}'$$

$$= \frac{1}{N_\mathbf{q}} \sum_{kl,\mathbf{q}} D_{kl}(\mathbf{q}) \sum_{\nu\mu} C^\mu_{ik}(\mathbf{k},\mathbf{q}) V_{\mu\nu}(\mathbf{k}-\mathbf{q}) C^\nu_{lj}(\mathbf{q},\mathbf{k})$$

$$P^\mathbf{k}_\mu(\mathbf{r}) = \sum_\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} P_\mu(\mathbf{r}-\mathbf{R})$$

$$\varphi^*_{i\mathbf{k}}(\mathbf{r})\varphi^*_{k\mathbf{q}}(\mathbf{r}) = \sum_\mu C^\mu_{ik}(\mathbf{k},\mathbf{q}) P^{\mathbf{q}-\mathbf{k}}_\mu(\mathbf{r})$$ RI-V

Using RI-LVL gives

$$C^\mu_{ik}(\mathbf{k},\mathbf{q}) = C^\mu_{ik}(-\mathbf{k},\mathbf{R}_0) + C^\mu_{ki}(\mathbf{q},\mathbf{R}_0)$$

but the matrix C^μ_{ik} is not sparse… still used for periodic GW

Ren et al. *Phys. Rev. Materials* **5**, 013807

In real space, sum up all periodic images:

$$K_{ij}(\mathbf{k}) = \sum_\mathbf{R} e^{i\mathbf{k}\cdot\mathbf{R}} X_{ij}(\mathbf{R}) \qquad D_{kl}(\mathbf{R}') = \frac{1}{N_\mathbf{q}} \sum_\mathbf{k} D_{kl}(\mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{R}'}$$

$$X_{ij}(\mathbf{R}) = \sum_{kl}\sum_{\mathbf{R}'} D_{kl}(\mathbf{R}') \sum_{\mathbf{R}''} \iint \frac{\varphi_i(\mathbf{r})\varphi_k(\mathbf{r}-\mathbf{R}')\varphi_j(\mathbf{r}'-\mathbf{R})\varphi_l(\mathbf{r}'-\mathbf{R}-\mathbf{R}'')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}d\mathbf{r}'$$

$$= \sum_{k\mathbf{R}'}\sum_{l\mathbf{R}''}\sum_{\mu\mathbf{Q}'}\sum_{\nu\mathbf{Q}''} C^{\mu(\mathbf{Q}')}_{ik(\mathbf{R}')} V_{\mu\nu(\mathbf{R}+\mathbf{Q}''-\mathbf{Q}')} C^{\nu(\mathbf{Q}'')}_{jl(\mathbf{R}'')} D_{kl}(\mathbf{R}+\mathbf{R}''-\mathbf{R}')$$

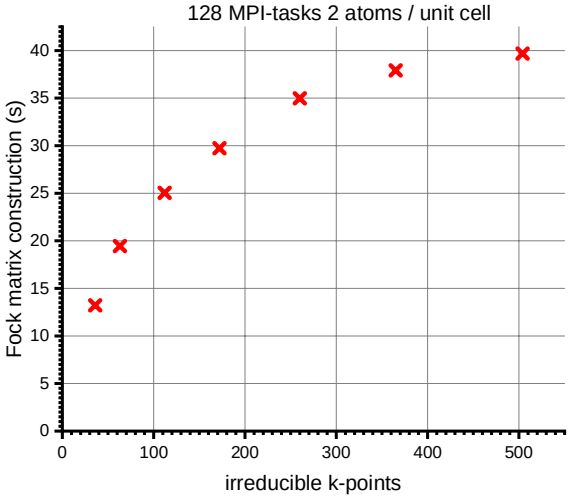
RI-LVL: **Q**' = **0** or **Q**' = **R**' and **Q**'' = **0** or **Q**'' = **R**''

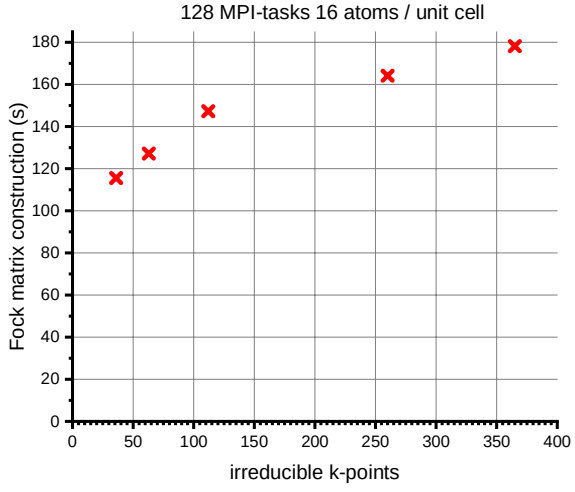
Plus:

- Born-von Karman periodic $V_{\mu\nu}$
- Sparsity of X_{ij} (**R**)
- Screening of small elements in X_{ij} (**R**)

Levchenko et al. *Comp. Phys. Comm.*, **192,** 60

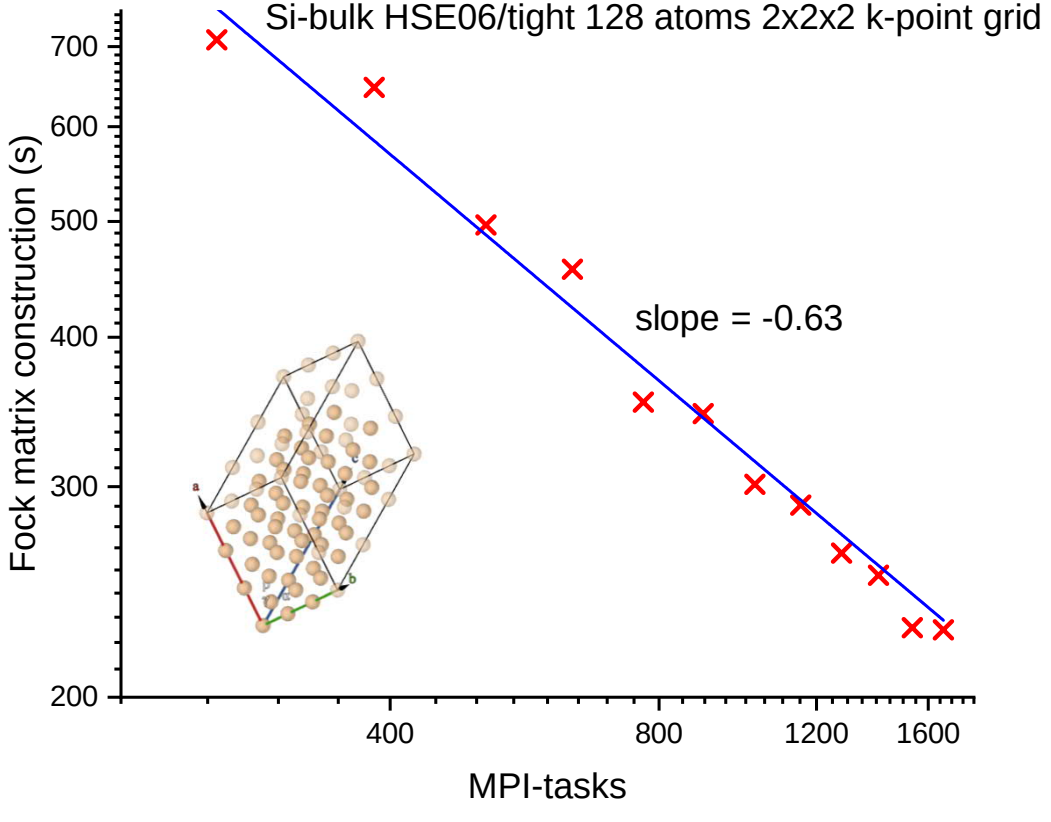
Performance and scalability HSE06



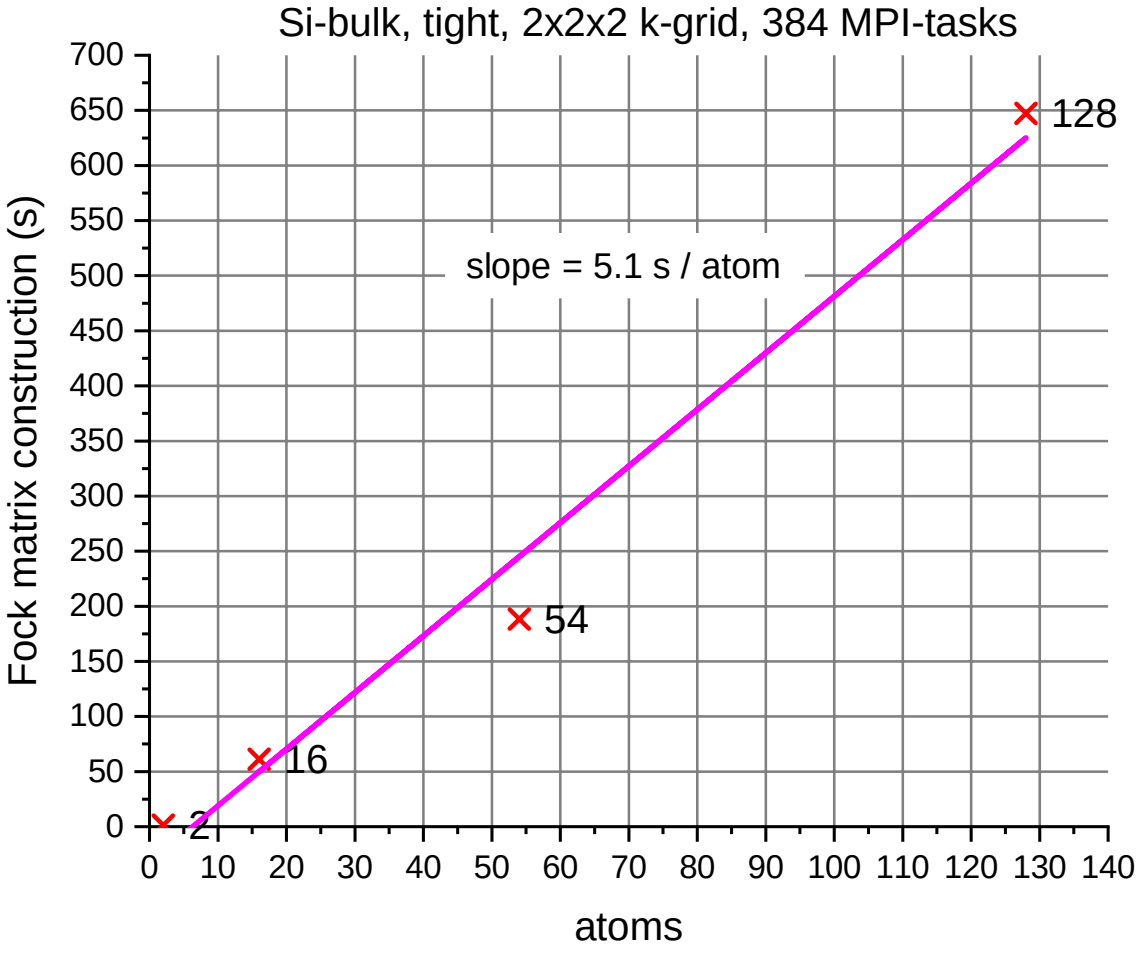


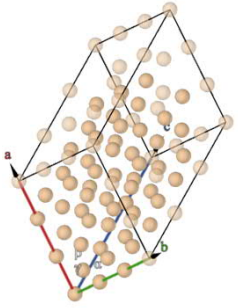
HSE → screened exchange:

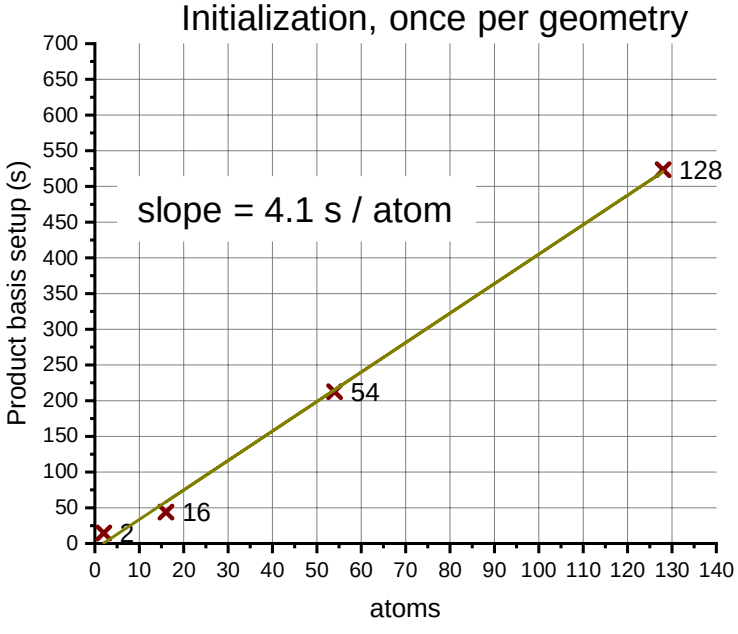
$$(ij|kl) = \iint \frac{\varphi_i^*(\mathbf{r})\varphi_j(\mathbf{r})\,\mathrm{erfc}(\omega|\mathbf{r}-\mathbf{r}'|)\,\varphi_k^*(\mathbf{r}')\varphi_l(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}d\mathbf{r}'$$



Performance and scalability HSE06

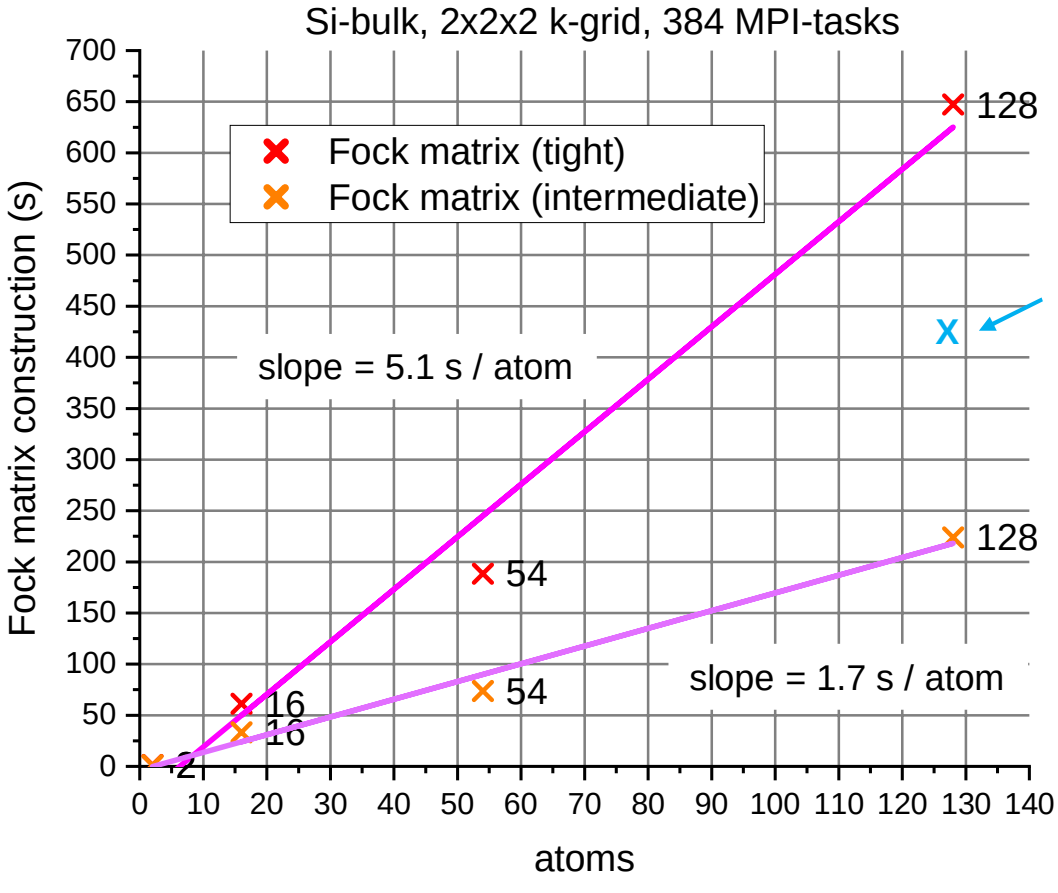


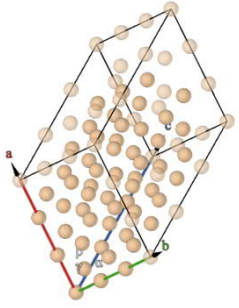




HSE06: tight → intermediate

For practical calculations *intermediate* settings are often sufficient





HF (no HSE-screening), intermediate

Si-tight:

```
hydro 3 d 9
hydro 5 g 9.4
```

Si-intermediate:

```
# hydro 3 d 9
for_aux hydro 5 g 9.4
```

GPU computing

Certain operations can be offloaded to GPUs. The results are best when the operations:

- are vectorizable
- contain no branching statements
- have minimal communication between CPU and GPU

E.g., integration: <u>In each batch:</u>

1. Compute the matrices on CPU

$$M_{jk} = \varphi_j(\mathbf{r}_k)\sqrt{w_k}$$

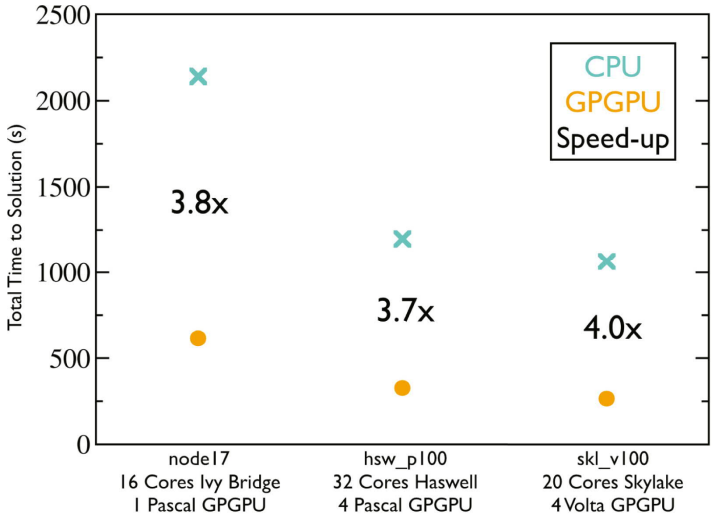
$$N_{ik} = h_{KS}(\mathbf{r}_k)\varphi_i(\mathbf{r}_k)\sqrt{w_k}$$

2. Copy data to GPU
3. Sum up the local contribution on GPU

$$h_{ij,loc} = \sum_k M_{jk} N^*_{ik}$$

works best with local dense storage

<u>When all batches done</u>
copy data to CPU



Si-bulk, 54 atoms / unit cell, tight, single k-point. Time for full calculation including forces and stress tensor.

Huhn et al. *Comp. Phys. Comm.*, **254**, 107314

How to get it? Build a GPU enabled binary and set `use_gpu .true.`

Conclusions

Grid based operations

- O(N) scaling
- Modest prefactor
- Use
  - o `use_local_index` when possible
  - o `load_balancing` when needed
- GPU support available
  - experiment with `points_in_batch`

RI operations

- Also O(N) scaling
- Much larger prefactor and cost of initialization
- Use the default method (RI-V or RI-LVL) or select with `RI_method`
- Test *intermediate* settings
- Experiment with `for_aux` to check the accuracy of the auxiliary basis

Thank you for your attention

Ville Havu

Wednesday, 27 October 2021

Aalto University
School of Science