A brief introduction to coupled cluster methods

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ABINIT School 2024
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Outline

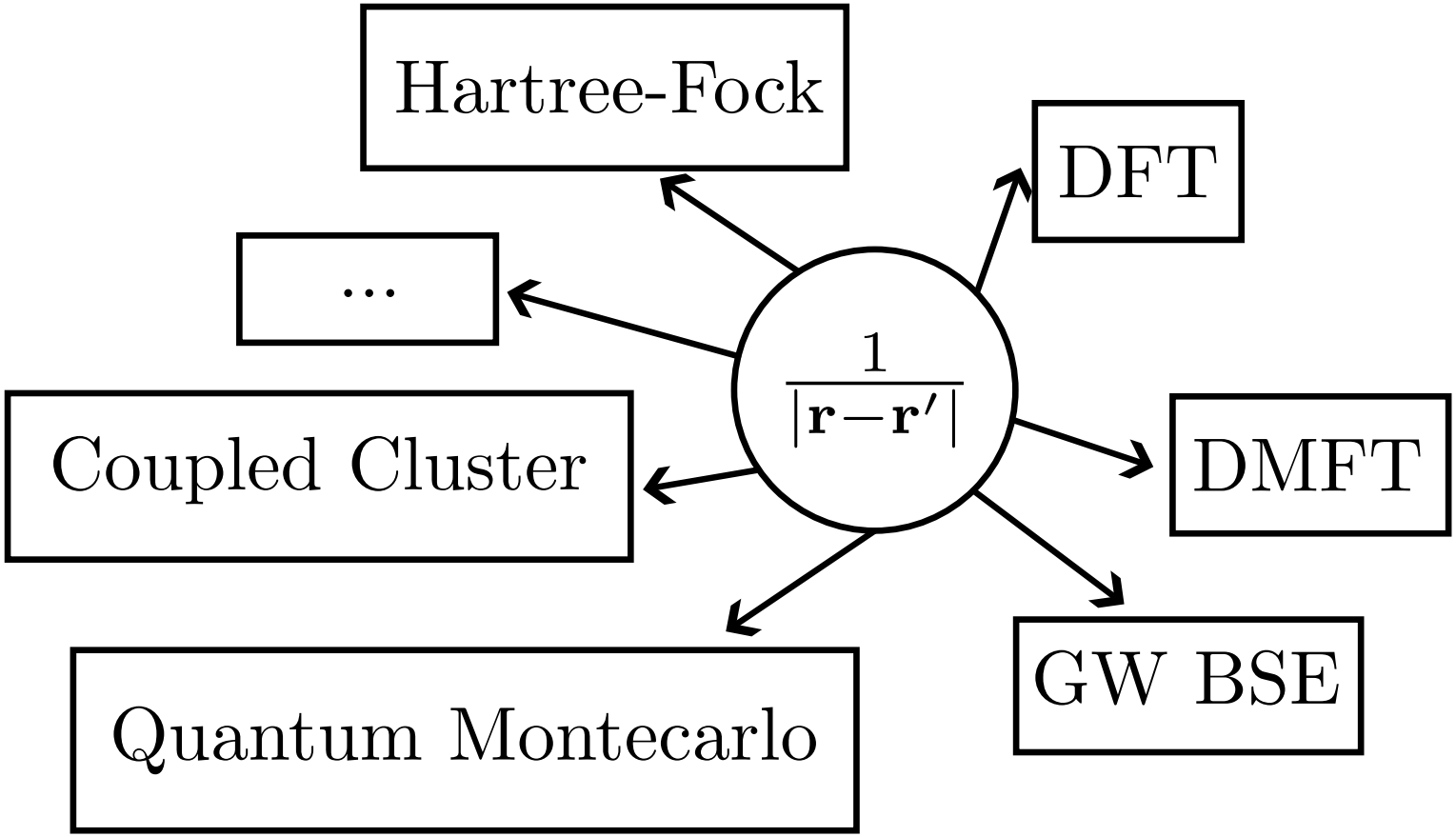
- Wavefunction basics
- The coupled cluster *Ansatz*
- Motivation: atomization energy errors with CCSD(T)
- Our code and its interfaces
- This afternoon: more information about the interface with `ABINIT`

What unites us all here?

What unites us all here?

$$\frac{1}{|\mathbf{r} - \mathbf{r}'|}$$

What unites us all here?



Hartree-Fock I

- Orbital basis $\{\varphi_1, \ldots, \varphi_N\}$
- Take one slater determinant

$$|\Psi\rangle = |\varphi_1 \cdots \varphi_N\rangle$$

$$\Psi(\mathbf{r}_1, \ldots, \mathbf{r}_N) = \frac{1}{\sqrt{N!}} \det \begin{pmatrix} \varphi_1(\mathbf{r}_1) & \cdots & \varphi_1(\mathbf{r}_N) \\ \vdots & \ldots & \vdots \\ \varphi_N(\mathbf{r}_1) & \cdots & \varphi_N(\mathbf{r}_N) \end{pmatrix}$$

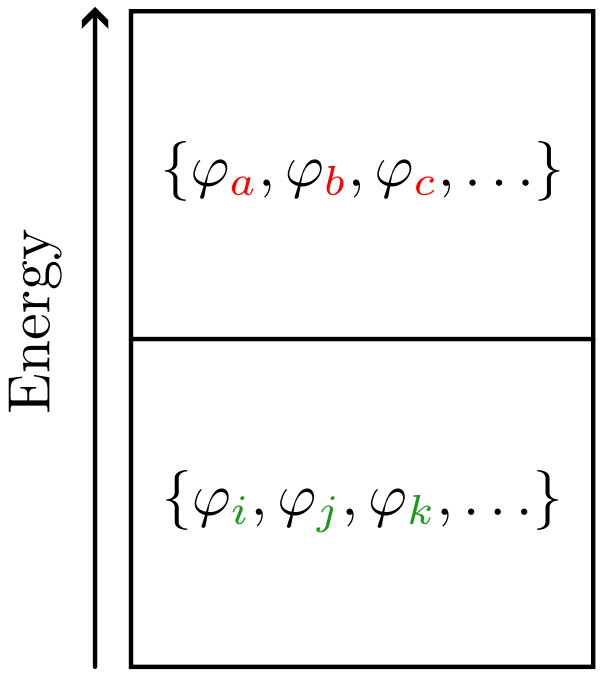
- Compute the energy of this determinant

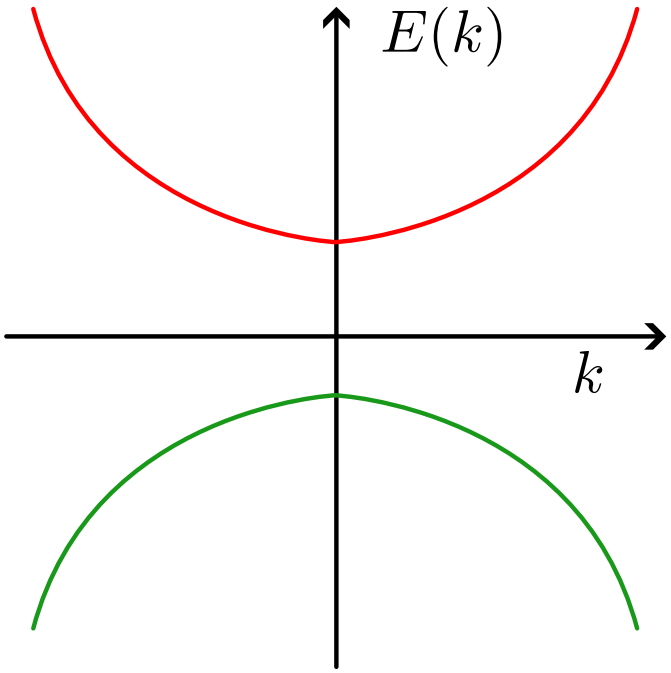
$$\begin{aligned} E &= \langle \Psi | \hat{H} | \Psi \rangle \\ &= \int \Psi^*(\mathbf{r}_1 \ldots) \hat{H}(\mathbf{r}_1 \ldots) \Psi(\mathbf{r}_1 \ldots) \, \mathrm{d}\mathbf{r}_1 \ldots \mathrm{d}\mathbf{r}_N \end{aligned}$$

- Minimize the energy in terms of the orbitals

$$E_{\mathrm{HF}} = \min_{\varphi_i} E$$

Hartree-Fock II





- Solving the self-consistent equations leads to
  occupied orbitals/bands $\{\varphi_i, \varphi_j, \varphi_k, \ldots\}$
  virtual orbitals/bands $\{\varphi_a, \varphi_b, \varphi_c, \ldots\}$

Hartree-Fock III

- A problem with HF: lack of correlation.
  Two electrons up and down spins

$$\text{orbitals} \rightarrow \{\varphi(\mathbf{r})\,|\alpha\rangle\,, \varphi(\mathbf{r})\,|\beta\rangle\}$$

- Slater determinant

$$\begin{aligned}\Psi(\mathbf{r}_1, \mathbf{r}_2) &= \frac{1}{\sqrt{2}} \det \begin{pmatrix} \varphi(\mathbf{r}_1)\,|\alpha\rangle & \varphi(\mathbf{r}_2)\,|\alpha\rangle \\ \varphi(\mathbf{r}_1)\,|\beta\rangle & \varphi(\mathbf{r}_2)\,|\beta\rangle \end{pmatrix} \\ &= \frac{1}{\sqrt{2}} \varphi(\mathbf{r}_1)\varphi(\mathbf{r}_2)(|\alpha\rangle\,|\beta\rangle - |\beta\rangle\,|\alpha\rangle)\end{aligned}$$

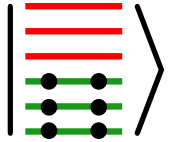
- Probability density is separable

$$|\Psi(\mathbf{r}_1, \mathbf{r}_2)|^2 = |\varphi(\mathbf{r}_1)|^2 \times |\varphi(\mathbf{r}_2)|^2$$

Adding a bit more correlation I

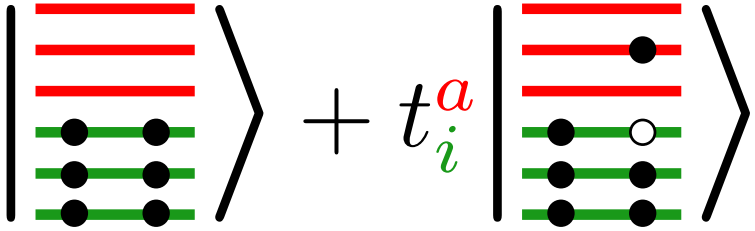
- One determinant is not enough, everything beyond having one determinant is called *Correlation Energy*
- Hartree-Fock determinant

$$|\mathrm{HF}\rangle = |\varphi_i \varphi_j \varphi_k \cdots\rangle =$$



- Add one singles excitation:

$$|\Psi_i^a\rangle = (1 + t_i^a \hat{c}_a^\dagger \hat{c}_i)\,|\mathrm{HF}\rangle$$



$$|\mathrm{HF}\rangle \quad + t_i^a \hat{c}_a^\dagger \hat{c}_i\,|\mathrm{HF}\rangle$$

- $|\Psi_i^a\rangle$ adds some correlation depending on a parameter t_i^a.

Adding a bit more correlation II

- Add all one singles excitations one after the other

$$
\begin{aligned}
|\Psi_{\mathrm{S}}\rangle &= (1 + t_i^a \hat{c}_a^\dagger \hat{c}_i)(1 + t_j^b \hat{c}_b^\dagger \hat{c}_j) \cdots |\mathrm{HF}\rangle \\
&= \prod_{\substack{i \in \text{Occupied} \\ a \in \text{Virtual}}} (1 + t_i^a \hat{c}_a^\dagger \hat{c}_i) \, |\mathrm{HF}\rangle \\
&= |\mathrm{HF}\rangle + \sum_{ia} t_i^a \hat{c}_a^\dagger \hat{c}_i \, |\mathrm{HF}\rangle + \sum_{ijab} t_i^a t_j^b \hat{c}_a^\dagger \hat{c}_i \hat{c}_b^\dagger \hat{c}_j \, |\mathrm{HF}\rangle + \cdots
\end{aligned}
$$

- But

$$
\hat{c}_a^\dagger \hat{c}_i \hat{c}_a^\dagger \hat{c}_i = 0 \quad \Longrightarrow \quad 1 + \hat{c}_a^\dagger \hat{c}_i = 1 + \hat{c}_a^\dagger \hat{c}_i + \frac{1}{2!} \left(\hat{c}_a^\dagger \hat{c}_i \right)^2 + \cdots = e^{\hat{c}_a^\dagger \hat{c}_i}
$$

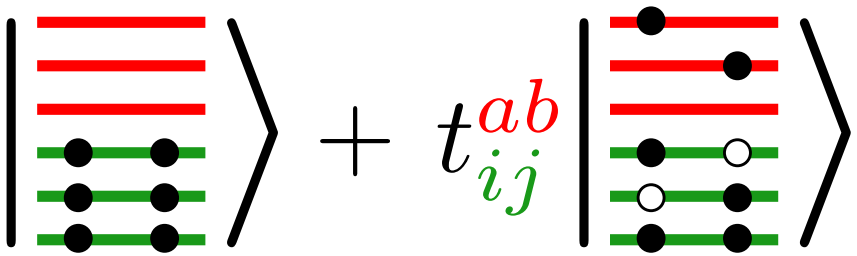
- Therefore

$$
|\Psi_{\mathrm{S}}\rangle = e^{\sum_{ai} t_i^a \hat{c}_a^\dagger \hat{c}_i} \, |\mathrm{HF}\rangle
$$

Adding a bit more correlation III

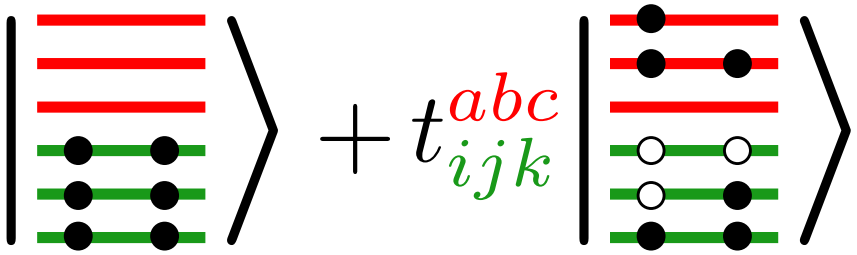
- One can also add pure two body excitations

$$|\Psi_{ij}^{ab}\rangle = (1 + t_{ij}^{ab}\hat{c}_a^\dagger\hat{c}_b^\dagger\hat{c}_j\hat{c}_i)\,|\mathrm{HF}\rangle$$



- Three body excitations

$$|\Psi_{ijk}^{abc}\rangle = (1 + t_{ijk}^{abc}\hat{c}_a^\dagger\hat{c}_b^\dagger\hat{c}_c^\dagger\hat{c}_k\hat{c}_j\hat{c}_i)\,|\mathrm{HF}\rangle$$



Coupled Cluster *Ansatz*

- Following the same procedure

$$|\Psi_{\mathrm{CC}}\rangle = \prod_{\substack{i \in \text{Occupied} \\ a \in \text{Virtual}}} (1 + t_i^a \hat{c}_a^\dagger \hat{c}_i) \prod_{a<b, i<j} (1 + t_{ij}^{ab} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_j \hat{c}_i) \cdots |\mathrm{HF}\rangle$$

$$= e^{\sum_{ai} t_i^a \hat{c}_a^\dagger \hat{c}_i + \sum_{abij} \frac{1}{4} t_{ij}^{ab} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_j \hat{c}_i + \cdots} |\mathrm{HF}\rangle$$

$$= e^{\hat{T}} |\mathrm{HF}\rangle$$

- The cluster operator

$$\hat{T} = \sum_{ai} t_i^a \hat{c}_a^\dagger \hat{c}_i + \sum_{abij} \frac{1}{2!^2} t_{ij}^{ab} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_j \hat{c}_i + \sum_{abcijk} \frac{1}{3!^2} t_{ijk}^{abc} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_c^\dagger \hat{c}_k \hat{c}_j \hat{c}_i + \cdots$$

N_{el}	t_{ij}^{ab} (GB)	t_{ijk}^{abc} (TB)
50	0.7	5
100	10.5	383
150	53.0	4370

Wave function correlation vs DFT correlation I

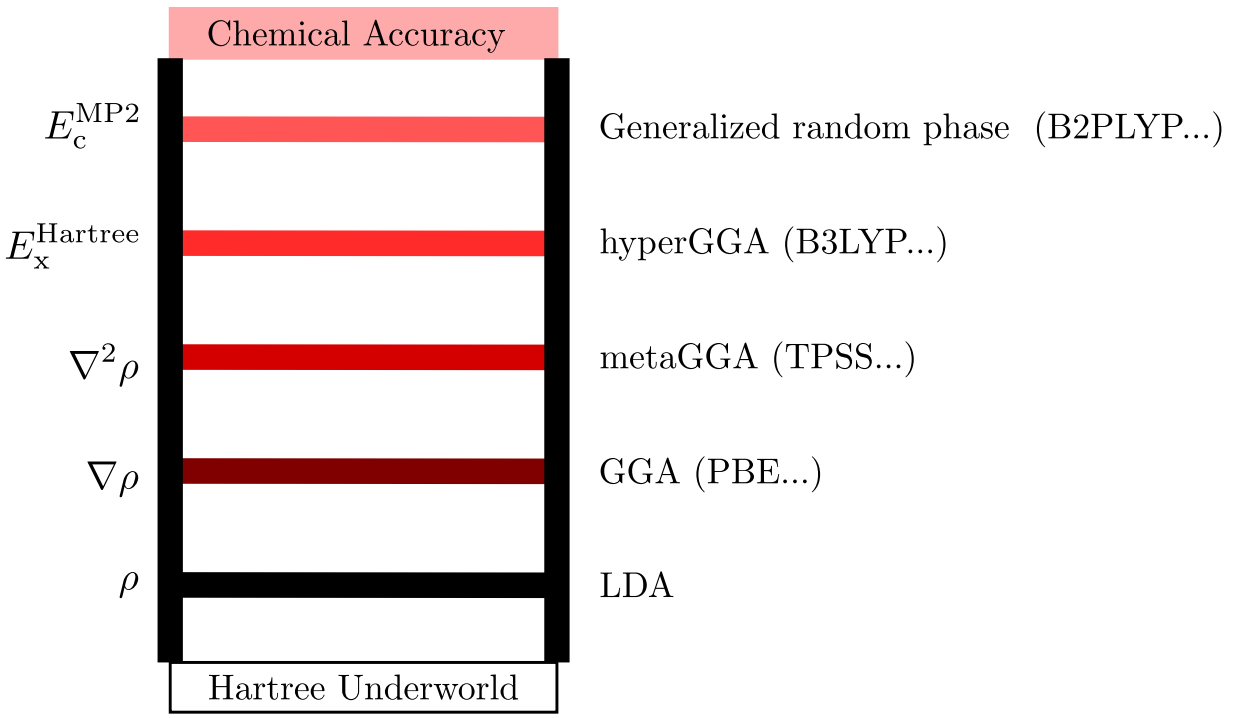
$$\left\{-\nabla^2 + v_{\text{ext}}(\mathbf{r}) + \int \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\mathrm{d}\mathbf{r}' + v_{\text{xc}}[n(\mathbf{r})]\right\}\varphi_n(\mathbf{r}) = \epsilon_n \varphi_n(\mathbf{r})$$

versus

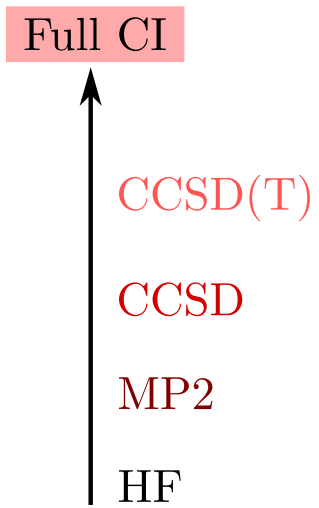
$$|\Psi\rangle = e^{\hat{T}}\,|\text{HF}\rangle = (1+\hat{C})\,|\text{HF}\rangle$$

Wave function correlation vs DFT correlation II

- ρ



- $|\Psi\rangle$



The Electronic Hamiltonian

- First quantization

$$\hat{H} = \underbrace{\sum_i \frac{1}{2}\hat{\mathbf{p}}_i^2}_{\text{El. Kinetic Energy}} - \underbrace{\sum_{i,J} \frac{Z_J}{|\mathbf{r}_i - \mathbf{r}_J|}}_{\text{Nuclei interaction}} + \underbrace{\frac{1}{2}\sum_{i \neq j} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}}_{\text{Electron-electron}}$$

- Second quantization

$$\hat{H} = \sum_{p,q} h_q^p \hat{c}_p^\dagger \hat{c}_q + \sum_{p,q,r,s} \frac{1}{4} V_{rs}^{pq} \hat{c}_p^\dagger \hat{c}_q^\dagger \hat{c}_s \hat{c}_r$$

where p, q, r, s can be either occupied $(i, j, k, \ldots)$ or virtual $(a, b, c, \ldots)$

- Coulomb integrals V_{rs}^{pq}

$$V_{rs}^{pq} = \int \frac{\varphi_p^*(\mathbf{r}_1)\varphi_r(\mathbf{r}_1)\varphi_q^*(\mathbf{r}_2)\varphi_s(\mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} \mathrm{d}\mathbf{r}_1 \mathrm{d}\mathbf{r}_2$$

Projection method for coupled cluster I

- insert ansatz into stationary Schrödinger equation

$$\hat{H} e^{\hat{T}} |\mathrm{HF}\rangle = E\, e^{\hat{T}} |\mathrm{HF}\rangle$$
$$e^{-\hat{T}} \hat{H} e^{\hat{T}} |\mathrm{HF}\rangle = e^{-\hat{T}} E\, e^{\hat{T}} |\mathrm{HF}\rangle = E\, |\mathrm{HF}\rangle$$

- project onto GS Slater determinant for correlated energy

$$\langle \mathrm{HF}| e^{-\hat{T}} \hat{H}\, e^{\hat{T}} |\mathrm{HF}\rangle = \langle \mathrm{HF}| E\, |\mathrm{HF}\rangle = E$$

- Orbital basis

  occupied $\{\varphi_i, \varphi_j, \ldots\}$
  virtual $\{\varphi_a, \varphi_b, \ldots\}$

- Determinant basis

  singles $\left\{ |{}^{a}_{i}\rangle = \hat{c}_a^\dagger \hat{c}_i\, |\mathrm{HF}\rangle \right\}$
  doubles $\left\{ |{}^{ab}_{ij}\rangle = \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_j \hat{c}_i\, |\mathrm{HF}\rangle \right\}$

Projection method for coupled cluster II

$$e^{-\hat{T}}\hat{H}e^{\hat{T}}\left|\mathrm{HF}\right\rangle = e^{-\hat{T}}E\,e^{\hat{T}}\left|\mathrm{HF}\right\rangle = E\left|\mathrm{HF}\right\rangle$$

- project onto determinant basis elements for coefficient equations

$$\left\langle{}^{a}_{i}\right|e^{-\hat{T}}\hat{H}\,e^{\hat{T}}\left|\mathrm{HF}\right\rangle = \left\langle{}^{a}_{i}\right|E\left|\mathrm{HF}\right\rangle = 0$$
$$\left\langle{}^{ab}_{ij}\right|e^{-\hat{T}}\hat{H}\,e^{\hat{T}}\left|\mathrm{HF}\right\rangle = \left\langle{}^{ab}_{ij}\right|E\left|\mathrm{HF}\right\rangle = 0$$

Coupled cluster equations (CCSD)

- The Schrödinger equation is non-linear now

$$\hat{H}e^{\hat{T}}|\mathrm{HF}\rangle = E_{\mathrm{CC}}e^{\hat{T}}|\mathrm{HF}\rangle \quad \Rightarrow \quad \underbrace{e^{-\hat{T}}\hat{H}e^{\hat{T}}}_{\bar{H}}|\mathrm{HF}\rangle = E_{\mathrm{CC}}|\mathrm{HF}\rangle$$

- Solve it projectively

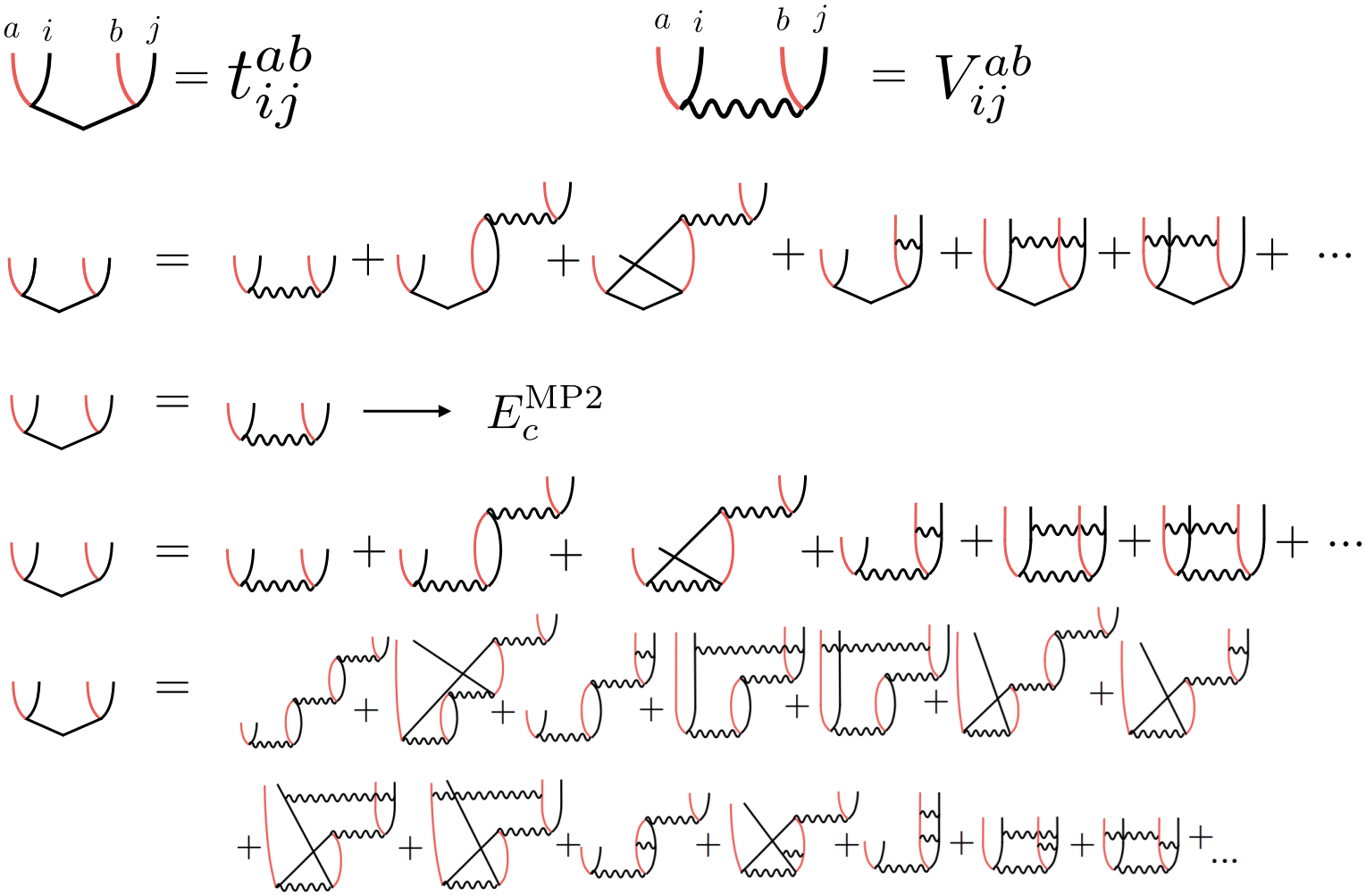
$$\begin{aligned}
E_{\mathrm{CC}} &= \langle \mathrm{HF}|\bar{H}|\mathrm{HF}\rangle &&= \textstyle\sum_{i,j,a,b} \frac{1}{4} V_{ab}^{ij} T_{ij}^{ab} + \dots \\
0 &= \langle {}_{i}^{a}|\, \bar{H}|\mathrm{HF}\rangle &&= \textstyle\sum_{b} f_{b}^{a} t_{i}^{b} - \sum_{j} f_{i}^{j} t_{j}^{a} + \dots \\
0 &= \langle {}_{ij}^{ab}|\, \bar{H}|\mathrm{HF}\rangle &&= \textstyle\sum_{c} f_{c}^{b} t_{ij}^{ac} + \sum_{ab} V_{cd}^{ab} t_{ij}^{cd} + \dots
\end{aligned}$$

- Start with an intial guess of $t_{ij}^{ab} = V_{ij}^{ab}$ and iterate (and hope for the best)
- Scaling $\mathcal{O}(N^6)$

Example of t_{ij}^{cd} equations (sum over repeated indices)

$$
\begin{aligned}
t_{ij}^{cd} = {} & V_{ij}^{cd} - t_m^c V_{ij}^{md} + t_m^d V_{ij}^{mc} + t_j^e V_{ie}^{cd} - t_i^e V_{je}^{cd} - f_i^m t_{mj}^{cd} \\
& + f_j^m t_{mi}^{cd} - f_e^d t_{ij}^{ec} + f_e^c t_{ij}^{ed} + \frac{1}{2} t_{mn}^{cd} V_{ij}^{mn} + t_{nj}^{ec} V_{ie}^{nd} - t_{nj}^{ed} V_{ie}^{nc} - t_{ni}^{ec} V_{je}^{nd} + t_{ni}^{ed} V_{je}^{nc} + \frac{1}{2} t_{ij}^{ef} V_{ef}^{cd} \\
& + t_m^c t_n^d V_{ij}^{mn} - t_j^e t_n^c V_{ie}^{nd} + t_j^e t_n^d V_{ie}^{nc} + t_i^e t_n^c V_{je}^{nd} \\
& - t_i^e t_n^d V_{je}^{nc} + t_i^e t_j^f V_{ef}^{cd} - f_f^m t_{mj}^{cd} t_i^f + f_f^m t_{mi}^{cd} t_j^f + f_f^m t_{ij}^{fc} t_m^d - f_f^m t_{ij}^{fd} t_m^c \\
& + \frac{1}{2} t_{mn}^{cd} t_j^g V_{ig}^{mn} - \frac{1}{2} t_{mn}^{cd} t_i^g V_{jg}^{mn} - t_{nj}^{ec} t_o^d V_{ie}^{no} + t_{nj}^{ed} t_o^c V_{ie}^{no} + t_{ni}^{ec} t_o^d V_{je}^{no} - t_{ni}^{ed} t_o^c V_{je}^{no} \\
& - t_{mj}^{cd} t_o^f V_{if}^{mo} + t_{mi}^{cd} t_o^f V_{jf}^{mo} - t_{nj}^{ec} t_i^g V_{eg}^{nd} + t_{nj}^{ed} t_i^g V_{eg}^{nc} + t_{ni}^{ec} t_j^g V_{eg}^{nd} - t_{ni}^{ed} t_j^g V_{eg}^{nc} - \frac{1}{2} t_{ij}^{ef} t_o^c V_{ef}^{od} \\
& + \frac{1}{2} t_{ij}^{ef} t_o^d V_{ef}^{oc} + t_{ij}^{ec} t_o^f V_{ef}^{od} - t_{ij}^{ed} t_o^f V_{ef}^{oc} + \frac{1}{2} t_{ij}^{ed} t_{op}^{fc} V_{ef}^{op} \\
& - \frac{1}{2} t_{ij}^{ec} t_{op}^{fd} V_{ef}^{op} + \frac{1}{4} t_{ij}^{ef} t_{op}^{cd} V_{ef}^{op} - \frac{1}{2} t_{mi}^{cd} t_{pj}^{fg} V_{fg}^{mp} + \frac{1}{2} t_{mj}^{cd} t_{pi}^{fg} V_{fg}^{mp} - t_{ni}^{ed} t_{pj}^{gc} V_{eg}^{np} \\
& + t_{ni}^{ec} t_{pj}^{gd} V_{eg}^{np} + t_j^e t_n^c t_o^d V_{ie}^{no} - t_i^e t_n^c t_o^d V_{je}^{no} - t_i^e t_j^f t_o^c V_{ef}^{od} \\
& + t_i^e t_j^f t_o^d V_{ef}^{oc} + \frac{1}{2} t_i^e t_j^f t_{op}^{cd} V_{ef}^{op} + t_i^e t_n^d t_{pj}^{gc} V_{eg}^{np} - t_i^e t_n^c t_{pj}^{gd} V_{eg}^{np} \\
& - t_j^e t_n^d t_{pi}^{gc} V_{eg}^{np} + t_j^e t_n^c t_{pi}^{gd} V_{eg}^{np} + t_i^e t_o^f t_{pj}^{cd} V_{ef}^{op} - t_j^e t_o^f t_{pi}^{cd} V_{ef}^{op} \\
& + \frac{1}{2} t_m^c t_n^d t_{ij}^{gh} V_{gh}^{mn} - t_m^d t_o^f t_{ij}^{hc} V_{fh}^{mo} + t_m^c t_o^f t_{ij}^{hd} V_{fh}^{mo} + t_i^e t_j^f t_o^c t_p^d V_{ef}^{op}
\end{aligned}
$$

Diagramatic fractal structure



Amplitudes with k-points

- Bloch orbitals $\varphi_i \to \varphi_{i\mathbf{k}_i}$
- Translational symmetry reduces memory constraints for integrals (block sparsity)

$$V_{r\mathbf{k}_r\ s\mathbf{k}_s}^{p\mathbf{k}_p\ q\mathbf{k}_q}\delta(\mathbf{k}_p + \mathbf{k}_q = \mathbf{k}_r + \mathbf{k}_s + \mathbf{G})$$

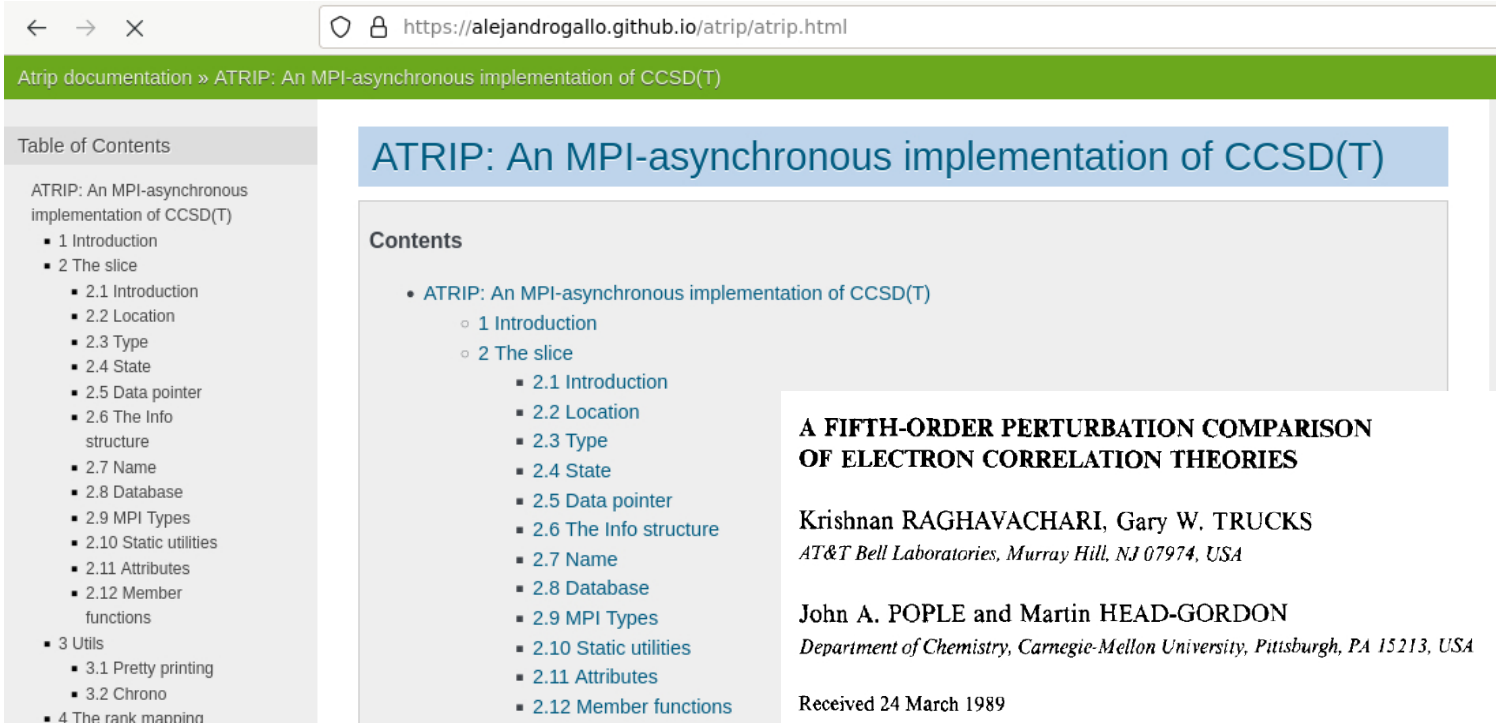
  with $\mathbf{G}$ a reciprocal lattice vector
- $t_{i\mathbf{k}_i\ j\mathbf{k}_j}^{a\mathbf{k}_a\ b\mathbf{k}_b}$ inherits block sparse structure
- Adapt equations to obey momentum conservation
- Adapt underlying tensor libraries to efficiently deal with block sparsity

CCSD(T)

- CCSDT too expensive for most electron sizes ($\mathcal{O}(N^8)$)

N_{el}	t_{ij}^{ab} (GB)	t_{ijk}^{abc} (TB)
50	0.7	5
100	10.5	383
150	53.0	4370

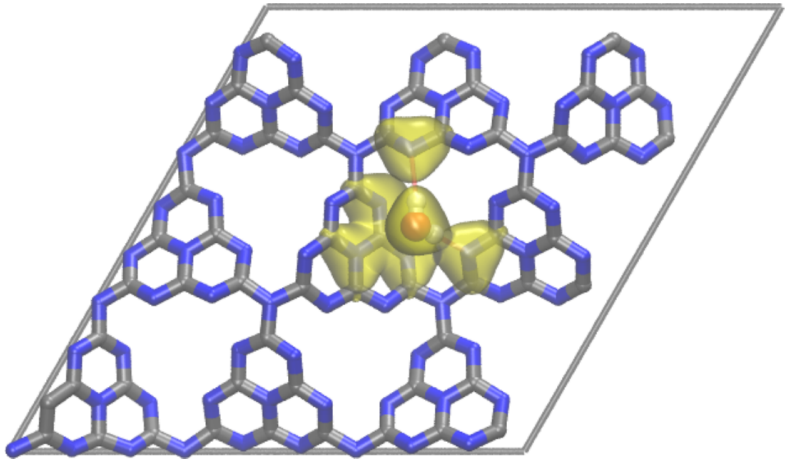
- Add triples correlation in a perturbative, *one-shot* fashion
- Exascale (GPU & CPU) implementation `ATRIP` $\mathcal{O}(N^7)$



CCSD(T): atomization energy errors

$$H\Psi(\mathbf{r}_1, ..., \mathbf{r}_N) = E\Psi(\mathbf{r}_1, ...\mathbf{r}_N)$$

CC4S ATRIP



Atomization energy errors

ΔE	PBE	SCAN	CCSD(T)
N2	-65.04	35.41	0.47
H2	20.51	7.79	0.00
F2	-58.41	-1.46	2.11
O2	-96.94	-28.38	2.39
CO	-39.38	26.48	-0.06
C2H2	-40.12	16.69	-0.80
CCH	-46.58	-0.03	2.84
CH2	-15.49	-25.29	0.12
CO2	-109.58	-4.93	-0.38
H2O2	-55.08	13.15	0.58
H2O	-5.03	19.28	0.02
HCO	-67.33	-9.42	0.41
HF	-1.59	14.94	0.05
NO	-82.85	6.60	1.53
OH	-11.08	-4.56	0.35
HNO	-74.63	25.14	1.09
CN	-69.55	13.10	6.57
HCN	-55.09	24.25	-0.17
CF	-51.94	-10.92	0.40
NH2	-25.49	-9.97	0.54
NH	-22.63	-11.15	0.51
rms	56.5	17.5	1.8

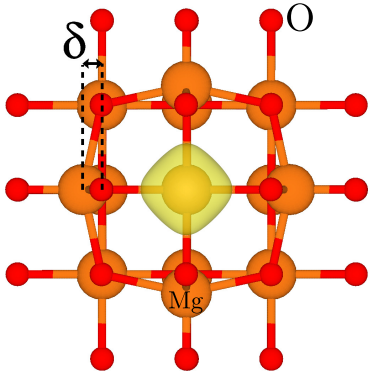
Room temp. 2.5 kJ/mol

Bonus: excited states for F-centers in MgO, SrO, CaO[1]

- Equation of motion formalism, (diagonalize $e^{-\hat{T}}\hat{H}e^{\hat{T}}$)

$$|R_n\rangle = \hat{R}_n e^{\hat{T}} |\mathrm{HF}\rangle \quad \hat{H}|R_n\rangle = E_n |R_n\rangle$$

- Absorption and emission energies



System	Method	Absorption (eV)	Emission (eV)
CaO	EOM-CCSD@HF	3.13	1.93
	Experiment	3.02	1.93
	Experiment	3.1	2.05 – 2.01
	TDDFT@B3LYP	3.52	2.1
	G_0W_0@PBE	3.20	
SrO	EOM-CCSD@HF	2.34	1.2
	Experiment	2.4	
MgO	EOM-CCSD@HF	5.28	3.66
	Experiment	5.0	2.4
	QMC	5.0(1)	3.8(1)
	CASPT2	5.44	4.09
	G_0W_0@LDA0-BSE	4.95	3.4

[1] A. Gallo et al. JCP, 154, 234103, 2021

./thank-you\!

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NOMAD
NOVEL MATERIALS DISCOVERY

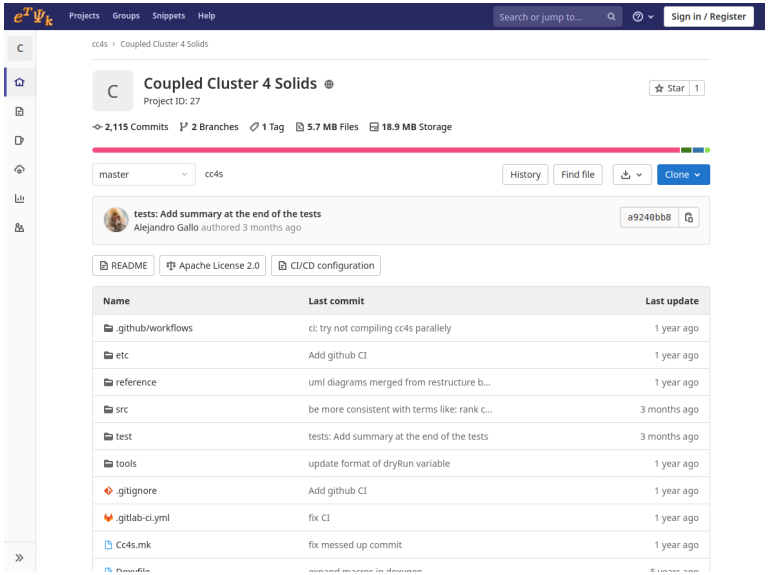
Hands-on session with the `ABINIT` interface

- We will calculate the atomization energy of `Be` as a test to show the workflow

  ABINIT DFT
  CC4S CCSD
  CC4S CCSD(T)

./CC4S/

- Massively parallel `C++` code for coupled cluster for solids.
- Interfaces with other codes exist, more on this later
- © `APACHEv2` license
- `github:cc4s/cc4s`



./CC4S/docs

- Comprehensive documentation
- https://manuals.cc4s.org/user-manual

Cc4s | Home | Sitemap | Algorithms | Objects

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   3.6. amplitudesConvergence
   3.7. initialAmplitudes
   3.8. mixer
      3.8.1. DiisMixer
      3.8.2. LinearMixer
4. Algorithm output
   4.1. Sample `stdout`
5. Sample `yaml` output
6. Computational complexity
7. Theory
   7.1. Coupled-Cluster Singles Doubles (`Ccsd`)
   7.2. Direct-Ring Couple-Cluster Doubles (`Drccd`)
8. Literature

CoupledCluster

1. Brief description

This algorithm solves the CCSD amplitude equations using the Coulomb integrals and Hartree–Fock eigenenergies.

2. Algorithm call

A typical input file snippet to call the `CoupledCluster` algorithm is given below.

```
- name: CoupledCluster
  in:
    method: Ccsd
    integralsSliceSize: 100
    slicedEigenEnergies: EigenEnergies
    coulombIntegrals: CoulombIntegrals
    slicedCoulombVertex: CoulombVertex
    maxIterations: 20
    energyConvergence: 1.0E-4
    amplitudesConvergence: 1.0E-4
    mixer:
      type: DiisMixer
      maxResidua: 5
    #or for instance a linear mixer
    #mixer:
    #  type: LinearMixer
    #  ratio: 1.0
  out:
    amplitudes: Amplitudes
```

3. Algorithm input

Table 1: Input keywords

Keyword	Value
`method`	Type of coupled cluster algorithm
`linearized`	Use linearized or fully non-linear coupled cluster
`integralsSliceSize`	Integral slice size

./CC4S/libs/CTF

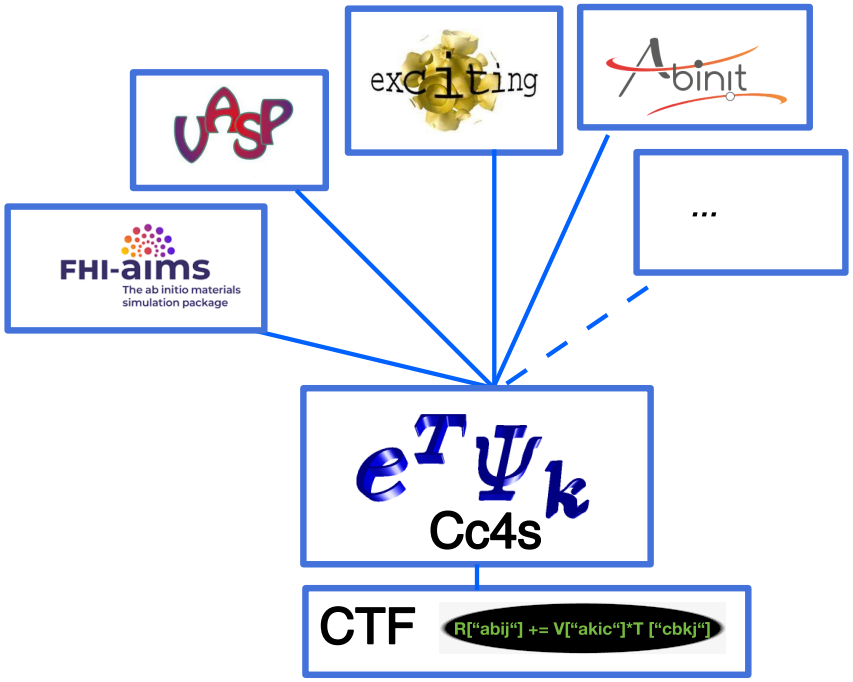
- Cyclops tensor framework
  `https://github.com/cyclops-community/ctf`
- Easily write tensor contractions massive parallelly

$$r_{ij}^{ab} = \sum_{ckdl} t_{ik}^{ac} V_{cd}^{lk} t_{lj}^{db}$$

turns into MPI C++ as

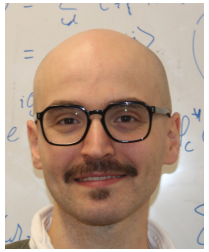
```
Tensor<double> r(4, ....), ...;

r["abij"] = t["acik"]
          * V["lkcd"]
          * t["dblj"];
```

./interfaces/


VASP
exciting
Abinit
...
FHI-aims
The ab initio materials simulation package
Cc4s
CTF
R["abij"] += V["akic"]*T ["cbkj"]

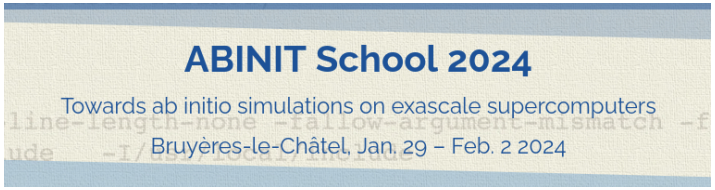
./interfaces/abinit

M. Giantomassi



A. Gallo

```
##########################################
# Dataset 2: CC4S interface
##########################################
optdriver2  6          # Activate GWR code
gwr_task2 "CC4S"       # Enter Abinit -> CC4S interface
getden2     1
nband2     100         # Number of (occ + empty) bands
ecuteps2   4
```



- Decemeber 2023 visit organized by X. Gonze in Louvain-la-Neuve
- Hands-on session in ABINIT school 2024

./interfaces/exciting

exciting

a full-potential all-electron package implementing linearized augmented planewave methods

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FHI-aims
The ab initio materials simulation package

JOSS
The Journal of Open Source Software

Interface to high-performance periodic coupled-cluster theory calculations with atom-centered, localized basis functions

Evgeny Moerman[1], **Felix Hummel**[2], **Andreas Grüneis**[2], **Andreas Irmler**[2], **and Matthias Scheffler**[1]

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DOI: 10.21105/joss.04040

Ingredients coming from ABINIT: Coulomb Vertex

- CC4S uses a *cholesky decomposition* of the Coulomb Integrals:

$$
\begin{aligned}
V_{rs}^{pq} &= \int \frac{\varphi_p^*(\mathbf{r}_1)\varphi_r(\mathbf{r}_1)\varphi_q^*(\mathbf{r}_2)\varphi_s(\mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} \mathrm{d}\mathbf{r}_1 \mathrm{d}\mathbf{r}_2 \\
&= \sum_G \bar{\Gamma}_r^{pG}\ \Gamma_s^{qG}
\end{aligned}
$$

where

$$
\Gamma_r^{pG} = \int_\Omega \mathrm{d}\mathbf{r} \sqrt{\frac{4\pi}{\mathbf{G}^2}}\, e^{i\mathbf{r}\cdot\mathbf{G}}\, \psi^*{}_p(\mathbf{r})\, \psi_r(\mathbf{r})
$$

- The Γ tensor is given by the files `CoulombVertex.yaml` and `CoulombVertex.elements`.
- Every tensor has a description in the `yaml` file, and data in the `.elements` file.

Ingredients coming from ABINIT: Eigenenergies ϵ_i

- At the moment KS eigenenergies supported, future work will enhance the Fock matrix routines in ABINIT in order to improve the hybrids and the CC4S interface.

ABINIT input file: SCF run

```
# Number of datasets: 2-step calculation
ndtset 2

ngkpt     1 1 1              # Number of grid points for k-point generation
nshiftk   1
shiftk    0 0 0              # Mesh centered on gamma
istwfk    *1                 # Required (for the time being)

# Definition of the plane-wave basis set
ecut 42.0                     # This should be used to reach convergence.

####################
# Dataset 1: SCF run
####################
nband1  48                   # Number of bands
tolvrs1  1.0d-8              # Tolerance to stop SCF cycles
```

ABINIT input file: CC4S

- It uses the GWR subsystem
- $N_{\mathrm{v}} + N_{\mathrm{o}} = 100$

```
#########################################
# Dataset 2: CC4S interface
#########################################
optdriver2  6          # Activate GWR code
gwr_task2 "CC4S"       # Enter Abinit -> CC4S interface
# gwr_task2 "CC4S_FULL" # Full DIAGO total number of states
getden2     1
nband2     100        # Number of (occ + empty) bands
ecuteps2   4
```

./CC4S/cc4s.in

- It uses also `yaml` as the input file format
- `CC4S` is quite verbose, like an *assembly* language
- Simple format

```
- name: NameOfTheAlgorithm
  in:
    # input parameters
    nelec: 42              #  Fake examples
    encut: 424242
  out:
    # output parameters
    density: Rho
```

./CC4S/Read

- The `Read` step reads in tensors
- Read Γ_r^{Gp}

```
- name: Read
  in:
    fileName: "CoulombVertex.yaml"
  out:
    destination: CoulombVertex
```

- Read eigenspektrum

```
- name: Read
  in:
    fileName: "EigenEnergies.yaml"
  out:
    destination: EigenEnergies
```

./CC4S/ holes and particles

- turn $\epsilon_p \rightarrow \{\epsilon_i, \epsilon_a\}$

```
- name: DefineHolesAndParticles
  in:
    eigenEnergies: EigenEnergies
  out:
    slicedEigenEnergies: EigenEnergies
```

- turn $\Gamma_{pGq} \rightarrow \{\Gamma_{pGq}, \Gamma_{aGb}, \Gamma_{aGi}, \Gamma_{iGj}\}$

```
- name: SliceOperator
  in:
    slicedEigenEnergies: EigenEnergies
    operator: CoulombVertex
  out:
    slicedOperator: CoulombVertex
```

./CC4S/coulomb-integrals

- Compute *lazy* Coulomb integrals $V_{rs}^{pq}(\Gamma)$

```
- name: VertexCoulombIntegrals
  in:
    slicedCoulombVertex: CoulombVertex
  out:
    coulombIntegrals: CoulombIntegrals
```

./CC4S/CCSD

```
- name: CoupledCluster
  in:
    method: Ccsd
    slicedEigenEnergies: EigenEnergies
    coulombIntegrals: CoulombIntegrals
    slicedCoulombVertex: CoulombVertex
    integralsSliceSize: 100
    maxIterations: 100
    energyConvergence: 1.0E-5
    amplitudesConvergence: 1.0E-5
    mixer:
      type: DiisMixer
      maxResidua: 12
  out:
    energy: CcsdEnergy
    amplitudes: Amplitudes
```

- Store the t_i^a, t_{ij}^{ab} in the tensor `Amplitudes`, you could then write out this tensor for inspection or postprocessing

./CC4S/CCSD(T)

- Use the CCSD amplitudes of previous step

```
- name: PerturbativeTriples
  in:
    slicedEigenEnergies: EigenEnergies
    amplitudes: Amplitudes
    coulombIntegrals: CoulombIntegrals
```

./CC4S/compile

```
git clone https://github.com/cc4s/cc4s
cd cc4s
```

or use `Makefile` configurations

- `./etc/config/gcc-oblas-ompi.mk`
- `./etc/config/icc-mkl-impi.mk`
- `./etc/config/icc-mkl-ompi.mk`
- You can edit this example files and do

```
# make external libraries
make CONFIG=gcc-oblas-ompi extern
make CONFIG=gcc-oblas-ompi
```

./CC4S/make-configuration/gcc-openBLAS-openmpi

```
include Cc4s.mk
include Extern.mk
include etc/make/blas.mk

# compiler and linker
CXX = mpicxx

# general and language options (for preprocessing, compiling and linking)
CXXFLAGS += \
-fopenmp -std=c++11 \
-Wall -pedantic --all-warnings -fmax-errors=3 \
-Wno-vla \
-Wno-int-in-bool-context

# optimization options (only for compiling and linking)
CXXFLAGS += -Ofast -march=native -fno-lto

CTF_CONFIG_FLAGS = CXX=${CXX} \
                   AR=gcc-ar \
                   CXXFLAGS="-Ofast -std=c++11 -march=native -fno-lto" \
                   LIBS="-L$(BLAS_PATH)" \
                   --no-dynamic \
                   --without-scalapack

LDFLAGS += \
-Wl,-Bstatic \
${STATIC_LIBS} \
-lgfortran -lquadmath \
-Wl,-Bdynamic \
${DYNAMIC_LIBS} \
```

LIVE DEMO
I ALSO LIKE TO LIVE DANGEROUSLY
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