Some aspects of multi-reference methods: size consistency and coupling between different correlation effects

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Single reference systems

Weakly correlated systems

- **Qualitatively** :
  - $|\Psi\rangle \approx |\mathbf{HF}\rangle$
  - Closed-shell grd. states
  - Large HOMO-LUMO gap
- **Dynamical correlation**
  - Short-range ($\approx$ cusp)
  - Long-range ($\approx$ VdW)
- e-e **correlation is weak** :
  - Perturbation
  - Coupled Cluster
- **Size extensivity**
  - Closed-shell systems
  - Large system

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Single-reference (SR) methods

- **Perturbative expansion** :
  - Rayleigh-Schroedinger
  - $|\Psi^{(0)}\rangle = |\mathbf{HF}\rangle$ (MPn)
  - Useful guide ! !
- **Important applications** :
  - Linked-Cluster Thm.
  - Size-extensivity
  - Coupled-Cluster
  - **CCSD(T)**
- **Nowadays developments** :
  - Bigger system (locality of e-e corr.)
  - Basis-set error (f_{12}, DFT-WFT)

Qualitative description of MR systems

- **Relatively few strongly correlated electrons**
  - Bond breakings
  - Magnetic systems

- **But rapidly large expansion for $|\Psi^{(0)}\rangle$!**

$$|\Psi^{(0)}\rangle = \sum_{\mathrm{I}=1}^{10^3-10^6} c_{\mathrm{I}} \, |\mathrm{I}\rangle$$

- **The ratios $\frac{c_{\mathrm{I}}}{c_{\mathrm{J}}}$ drive most of the physical properties**

- **Between the $|\mathrm{I}\rangle$ and $|\mathrm{J}\rangle$**
  - Large interactions
  - Energetic degeneracies
  - $\frac{\langle \mathrm{J}|H|\mathrm{I}\rangle}{\Delta E_{\mathrm{IJ}}} \gg 1$

- **Non perturbative**

Quantitative description : the physics beyond $|\Psi^{(0)}\rangle$

$$|\Psi\rangle = |\Psi^{(0)}\rangle \; + \sum_i \; c_i \; |\phi_i\rangle$$

- **Standard dynamical correlation** (cusp, VdW)
- **Differential correlation effects**
  - The $|\mathrm{I}\rangle$ are different
  - Correlation effects depend on $|\mathrm{I}\rangle$
- **Change** $|\Psi^{(0)}\rangle$
  - Affects the $\langle \mathrm{J}|H|\mathrm{I}\rangle$ and ΔE_{IJ}
  - Renormalization of H
- **Size consistency**
  - Able to break bonds
  - Treat open shell systems

The questions that must be answered for our MR methods

$$|\Psi\rangle = |\Psi^{(0)}\rangle + \sum_i c_i \, |\phi_i\rangle$$

- How do we **compute the energy** ?
- What **choice for** $|\Psi^{(0)}\rangle$?
- What **choice for the** $|\phi_i\rangle$?
- How do we **determine the** c_i ?

Requirements for a good MR method

- **"Truly MR"**
  - Same status for all $|\mathrm{I}\rangle$ in $|\Psi^{(0)}\rangle$
- **Correct treatment of dynamic correlation**
  - No divergences
  - Accurate
- **Treat the coupling static / dynamical correlation**
  - Building an effective Hamiltonian $\tilde{H}$ within $\{|\mathrm{I}\rangle\}$
    $\tilde{H} = \sum_{\mathrm{I,J}} \left(H_{\mathrm{IJ}} + \tilde{O}_{\mathrm{IJ}}\right) |\mathrm{I}\rangle\langle\mathrm{J}|$
  - Diagonalize $\tilde{H}$ can change $|\Psi^{(0)}\rangle$
- **Size-consistent**
  - $E(A \cdots B) = E(A) + E(B)$
  - Correct even for open-shell sub-systems A and B
- **Lowest computational cost ..**

How to compute the energy ... ?

- **Variational calculations**
- **Projection technique**

To be variational or not, that is the question ...

Variational calculations : **CI calculations**

- **Average value** of H on $|\Psi\rangle$:

$$E_{\Psi}^{\mathrm{Var}} = \frac{\langle\Psi|H|\Psi\rangle}{\langle\Psi|\Psi\rangle} = \frac{\sum_{\mathrm{IJ}} c_{\mathrm{J}} \langle \mathrm{J}|H|\mathrm{I}\rangle c_{\mathrm{I}}}{\langle\Psi|\Psi\rangle}$$

- **Upper bound to the FCI energy : ☺**
  - no divergences : can treat strong correlation
  - $E = \min_{\Psi} E_{\Psi}^{\mathrm{Var}}$
  - easy to solve (Lanczos, Davidson)
- **Space is not closed : ☹**
  - always exist some $|\mu\rangle$ such that $\langle\mu|H|\Psi\rangle \neq 0$
  - linear parametrization are required
  - size consistency issues

To be variational or not, that is the question ...

Non-Variational calculations : **CC, PT, FCI-QMC**

- Suppose that $H|\Psi\rangle = E|\Psi\rangle$ **is valid** with :

$$|\Psi\rangle = |\Psi^{(0)}\rangle + \sum_{i \in \text{FOIS}} c_i |\phi_i\rangle + |\mathcal{R}\rangle$$

  with FOIS $\Leftrightarrow \langle\Psi^{(0)}|H|\phi_i\rangle \neq 0$ and $\langle\Psi^{(0)}|H|\mathcal{R}\rangle = 0$

- Non variational $\Leftrightarrow$ **projection** on the **reference WF** $\langle\Psi^{(0)}|$:

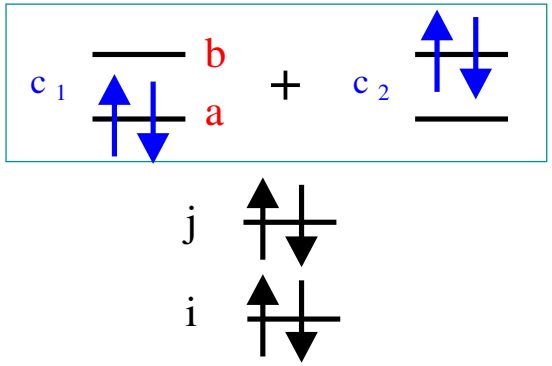
$$\begin{aligned} E_\Psi^{\text{Proj}} &= \langle\Psi^{(0)}|H|\Psi\rangle \\ &= \underbrace{\langle\Psi^{(0)}|H|\Psi^{(0)}\rangle}_{E^{\text{Var}}_{\Psi^{(0)}}} + \sum_{i \in \text{FOIS}} c_i \langle\Psi^{(0)}|H|\phi_i\rangle \end{aligned}$$

- **not necessary an upper bound** ☹
- **Variational for** $|\Psi^{(0)}\rangle$
  $\Rightarrow$ good for the strongly correlated electrons !
- Only needs the **coefficient of the FOIS** ☺
  - Much easier to close the space ☺
  - Size consistency ☺

The space in which we are going to work

- **The zeroth-order wave function : CAS-CI**
  - All determinants within n e and m orbitals

$$|\Psi^{(0)}\rangle = \sum_{\mathrm{I}} c_{\mathrm{I}} |\mathrm{I}\rangle$$



- **Size extensive ☺**
  - If active space is correctly chosen

$$E^{(0)}(A \cdots B) = E_A^{(0)} + E_B^{(0)}$$

  - Also works for open-shell systems A and B

How do we determine c_i ?

Rayleigh-Schroedinger Perturbation Theory

- Assume a **partitioning of** H

$$H = H^{(0)} + V$$

- and $H^{(0)}$ **being diagonal on the** $|\phi_i\rangle$ **and** $|\Psi^{(0)}\rangle$

$$H^{(0)}|\Psi^{(0)}\rangle = E^{(0)}|\Psi^{(0)}\rangle$$
$$H^{(0)}|\phi_i\rangle = E_i^{(0)}|\phi_i\rangle$$

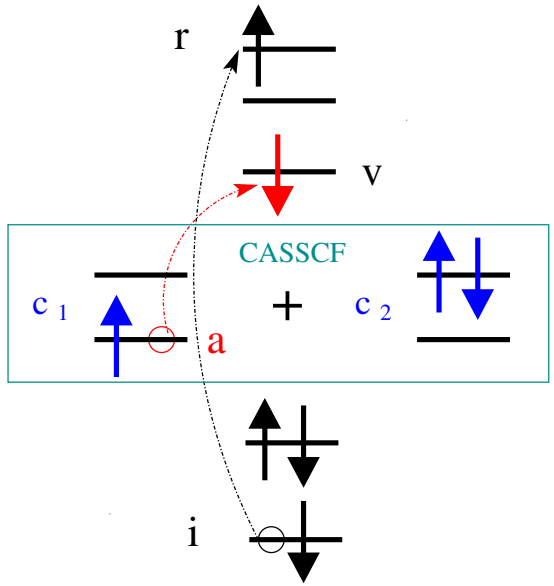
- Then the **coefficient** c_i **at first order is simply** :

$$c_i^{(1)} = \frac{\langle\Psi^{(0)}|H|\phi_i\rangle}{E^{(0)} - E_i^{(0)}}$$

Choice of the $|\phi_i\rangle$

- **The $|\phi_i\rangle$: connected to $|\Psi^{(0)}\rangle$**

$$|\phi_i\rangle \text{ such that } \langle\phi_i|H|\Psi^{(0)}\rangle \neq 0$$



- **Singles and doubles exc. on top of $|\Psi^{(0)}\rangle$**

$$|\Psi\rangle = |\Psi^{(0)}\rangle + \underbrace{\sum_i c_i \, |\phi_i\rangle}_{\text{singles and doubles exc.}}$$

- **In SR methods $|\phi_i\rangle$ are Slater determinants**

$$|\phi_i\rangle = a_a^\dagger a_b^\dagger a_i a_j |\mathbf{HF}\rangle$$

- In MR methods it is more complicated ..

Choice of the $|\phi_i\rangle$ in MR method

- **Linear combinations** (Internally-contracted)

$$|\phi_i\rangle = a_a^\dagger a_b^\dagger a_i a_j \; |\Psi^{(0)}\rangle = \sum_{\mathrm{I}} \; c_{\mathrm{I}} \; a_a^\dagger a_b^\dagger a_i a_j \; |\mathrm{I}\rangle$$

- **Single determinants** (Externally-uncontracted)

$$|\phi_i\rangle = a_a^\dagger a_b^\dagger a_i a_j \; |\phi_i\rangle$$

- **Key questions** :
  - Size-extensivity
  - Changing $|\Psi^{(0)}\rangle \Leftrightarrow$ building $\tilde{H}$

The size-extensivity

MRPT2 using Linear combinations

- **CASPT2**
  - Quite accurate (but empirical ..)
  - **Empirical parameters** (IP-EA shifts, imaginary shifts ...) ☹
  - $H^{(0)}$ is a generalized Fock operator
  - **One body operator ⇔ Not size consistent** ☹
- **NEVPT2**
  - Quite accurate
  - **No empirical parameters** ☺
  - $H^{(0)}$ is hybrid : the Dyall Hamiltonian

$$\hat{H}_D = \hat{F}_{\text{core}} + \hat{F}_{\text{virtuals}} + \sum_{a,b,c,d} (ab|cd) a_b^\dagger a_d^\dagger a_c a_a$$

  - **Two body operator in the active space + Linear combination ⇒ size consistent ! !** ☺

The size-extensivity

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  - **Two body operator in the active space + Linear combination $\Rightarrow$ size consistent ! !** ☺

But really hard to build $\tilde{H}$ ... ☹

Building of $\tilde{H}$: the Shifted-B_k

MRPT2 using Slater determinants

- The **first order coefficient** of $|\phi_i\rangle$:

$$c_i^{(1)} = \frac{\langle \phi_i | H | \Psi^{(0)} \rangle}{E^{(0)} - E_i^{(0)}} = \sum_{\mathrm{I}} c_{\mathrm{I}} \frac{\langle \phi_i | H | \mathrm{I} \rangle}{E^{(0)} - E_i^{(0)}}$$

- The contribution of $|\phi_i\rangle$ to the **energy at second order** :

$$e_i^{(2)} = c_i^{(1)} \langle \Psi^{(0)} | H | \phi_i \rangle = \frac{\langle \Psi^{(0)} | H | \phi_i \rangle^2}{E^{(0)} - E_i^{(0)}}$$

Building of $\tilde{H}$

MRPT2 using Slater determinants : the Shifted-B_k

- $e_i^{(2)}$ can be reinterpreted as an **expectation value** of a new operator :

$$e_i^{(2)} = \sum_{\mathrm{IJ}} c_{\mathrm{J}} \frac{\langle \mathrm{J}|H|\phi_i\rangle\langle\phi_i|H|\mathrm{I}\rangle}{E^{(0)} - E_i^{(0)}} \; c_{\mathrm{I}} = \langle\Psi^{(0)}|\tilde{O}_i|\Psi^{(0)}\rangle$$

$$\langle \mathrm{J}|\tilde{O}_i|\mathrm{I}\rangle = \frac{\langle \mathrm{J}|H|\phi_i\rangle\langle\phi_i|H|\mathrm{I}\rangle}{E^{(0)} - E_i^{(0)}}$$

- And so the total dressed $\tilde{H}$ is simply :
(Shavitt, 1968 ; Davidson, 1983, Nakano, 1993)

$$\langle \mathrm{J}|\tilde{H}|\mathrm{I}\rangle = \langle \mathrm{J}|H|\mathrm{I}\rangle + \sum_i \frac{\langle \mathrm{J}|H|\phi_i\rangle\langle\phi_i|H|\mathrm{I}\rangle}{E^{(0)} - E_i^{(0)}}$$

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Now we can couple the $|\Psi^{(0)}\rangle$ and the $|\phi_i\rangle$ through $\langle \mathrm{J}|\tilde{H}|\mathrm{I}\rangle$! ! ☺

Building of $\tilde{H}$

MRPT2 using Slater determinants : the Shifted-B_k

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Now we can couple the $|\Psi^{(0)}\rangle$ and the $|\phi_i\rangle$ through $\langle \mathrm{J}|\tilde{H}|\mathrm{I}\rangle$! ! ☺

But we have a size consistency issue now ... ☹

Why a size consistency issue ....

The problem of Slater determinants ...

- The problem comes from **the energy denominators**

$$\Delta E_i^{(0)} = E^{(0)} - E_i^{(0)}$$

- Let's assume a Epstein-Nesbet H_0

$$E^{(0)} = \langle \Psi^{(0)} | H | \Psi^{(0)} \rangle$$
$$E_i^{(0)} = \langle \phi_i | H | \phi_i \rangle$$

- **This comparaison is unfair ! !**
  - $E^{(0)}$ contains correlation effects ☺
  - $E_i^{(0)}$ does not ! ☹
- Leads to **non separable correlated energies ...**

$$E(A \cdots B) \neq E(A) + E(B)$$

Some mumerical test of separability

TABLE – Total energies (a. u.) for the numerical separability check on $F_2 \ldots FH$.

	CASSCF	Shifted-B_k
F_2	-198.746157368569	-199.122170300
FH	-100.031754985880	-100.289784498
F_2 + FH	-298.777912354448	-299.411954798
$F_2 \ldots$ FH	-298.777912354443	-299.396752116
Absolute error (a.u.)	5.0×10^{-12}	1.5×10^{-2}
Relative error	1.7×10^{-14}	5.1×10^{-5}

Alternatives

- Proposal by **Lindgren** (QD-PT, 1974)

$$\Delta E_i^{(0)} = E_{\rm I}^{(0)} - E_i^{(0)} = \langle {\rm I}|H|{\rm I}\rangle - \langle \phi_i|H|\phi_i\rangle$$

  $\Rightarrow$ **systematically diverges ! !** ☹

- Proposal by **Heully *et. al*** (H_{int}, 1996)

$$\Delta E_{{\rm I}i}^{(0)} = \langle {\rm I}|H|{\rm I}\rangle - \langle \phi_i|H|\phi_i\rangle + \delta_{{\rm I}i}$$

  $\Rightarrow$ **Numerically instable** ☹

- Related proposal by **Mukherjee *et. al*** (Mk-MRPT2, 1999)
  $\Rightarrow$ **Numerically instable** ☹

Proposal of a solution (Giner *et. al*, 2017)

- Taking zeroth-order energies of **multi-reference wave functions**

$$\Delta E^{(0)}_{\mathrm{I}i} = \langle \Psi^{(0)} | H | \Psi^{(0)} \rangle - \frac{\langle \Psi^{(0)} | \left(\hat{T}_{\mathrm{I}i} \right)^{\dagger} \; H \; \hat{T}_{\mathrm{I}i} | \Psi^{(0)} \rangle}{\langle \Psi^{(0)} | \left(\hat{T}_{\mathrm{I}i} \right)^{\dagger} \; \hat{T}_{\mathrm{I}i} | \Psi^{(0)} \rangle}$$

- Leads to **strictly separable energies !** ☺
- Linked to MR-CC formalism

Some mumerical proof of separability

TABLE – Total energies (a. u.) for the numerical separability check on $F_2 \ldots FH$.

	CASSCF	JM-HeffPT2
F_2	-198.746157368569	-199.085305155169
FH	-100.031754985880	-100.262424667296
F_2 + FH	-298.777912354448	-299.347729822466
$F_2 \ldots$ FH	-298.777912354443	-299.347729822462
Absolute error (a.u.)	5.0×10^{-12}	4.4×10^{-12}
Relative error	1.7×10^{-14}	1.4×10^{-14}

Summary

What we briefly saw ...

- No definitive answer unlike SR methods
- Still room for improvements in MR methods
  - Coupling betweem static / dynamic
  - Size extensivity
- Requires flexible formalisms (and codes ! !)

Main developpements

- Flexible MRCC / MRPT
- Coupling MR-WFT with range-separated DFT
- Using local orbitals

Size extensivity and size consistency

Why worrying so much about that ?

- Energy is an **extensive property** :
  - scales linearly with the number of **non interacting systems**
  - **Correlation energy** also does
- Such a property requires a **product wave function**

$$E(A \cdots B) = E(A) + E(B) \Leftrightarrow |\Psi_{A+B}\rangle = |\Psi_A\rangle \times |\Psi_B\rangle$$

- Consider two wave function $|\Psi_A\rangle$ and $|\Psi_B\rangle$ and their product

$$|\Psi_A\rangle = \sum_{\mathrm{I}_A}^{N_A} c_{\mathrm{I}_A} |\mathrm{I}_A\rangle, \quad |\Psi_B\rangle = \sum_{\mathrm{I}_B}^{N_B} c_{\mathrm{I}_B} |\mathrm{I}_B\rangle$$

$$|\Psi_{A+B}\rangle = \sum_{\mathrm{I}_A}^{N_A} \sum_{\mathrm{I}_A}^{N_B} c_{\mathrm{I}_B} c_{\mathrm{I}_A} |\mathrm{I}_A\rangle \times |\mathrm{I}_B\rangle$$

Size extensivity and size consistency

Variational methods

- In a variational calculation one has an **expectation value**

$$E_\Psi = \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle} = \sum_{\mathrm{I}}^{N_\mathrm{I}} \sum_{\mathrm{J}}^{N_\mathrm{I}} c_\mathrm{I} c_\mathrm{J} \langle \mathrm{J} | H | \mathrm{I} \rangle$$

- It is typically a $(N_\mathrm{I})^2$ calculations
- $|\Psi_{A+B}\rangle = |\Psi_A\rangle \times |\Psi_B\rangle$ contains $N_A \times N_B$ Slater determinants
- Typically for practical QM calculation, $N_{A/B} \approx 10^6$
- Computing the variational energy of $|\Psi_{A+B}\rangle$ requires :
  - the storage of typically 10^{12} Slater determinants ☹
  - 10^{24} floating points operations ☹
- Doesn't looks afordable ....

Size extensivity and size consistency

Projection methods

- Now these wave functions are written as :

$$|\Psi_A\rangle = |\Psi_A^{(0)}\rangle + \sum_{i=S,D}^{N_{SD}^A} c_i^A |i_A\rangle + |\mathcal{R}_A\rangle, \quad |\Psi_B\rangle = |\Psi_B^{(0)}\rangle + \sum_{i=S,D}^{N_{SD}^B} c_i^B |i_B\rangle + |\mathcal{R}_B\rangle$$

with $\langle \Psi^{(0)} | H | \mathcal{R}_A \rangle = 0$

- The energy can be computed by **projection** of $\langle \Psi_{A/B}^{(0)}|$:

$$\begin{aligned} E_A = \langle \Psi_A^{(0)} | H | \Psi_A \rangle = & \langle \Psi_A^{(0)} | H | \Psi_A^{(0)} \rangle \\ & + \langle \Psi_A^{(0)} | H \left(\sum_{i=S,D}^{N_{SD}^A} c_i^A |i_A\rangle \right) \\ & \cancel{\langle \Psi_A^{(0)} | H | \mathcal{R}_A \rangle} \end{aligned}$$

- Which is

$$E_A = E_{\Psi_A^{(0)}} + E_{\text{corr}}^A$$

Size extensivity and size consistency

Projection methods

- The wave function of $A \cdots B$ can also be written as :

$$
\begin{aligned}
|\Psi_{A+B}\rangle =& |\Psi_A^{(0)}\rangle \times |\Psi_B^{(0)}\rangle \\
&+ |\Psi_A^{(0)}\rangle \times \left(\sum_{i=S,D}^{N_{SD}^B} c_i^B |i_B\rangle \right) + |\Psi_B^{(0)}\rangle \times \left(\sum_{i=S,D}^{N_{SD}^A} c_i^A |i_A\rangle \right) \\
&+ \left(\sum_{i=S,D}^{N_{SD}^B} c_i^B |i_B\rangle \right) \times \left(\sum_{i=S,D}^{N_{SD}^A} c_i^A |i_A\rangle \right) \\
&+ \left(\sum_{i=S,D}^{N_{SD}^B} c_i^B |i_B\rangle \right) \times |\mathcal{R}_A\rangle + \left(\sum_{i=S,D}^{N_{SD}^A} c_i^A |i_A\rangle \right) \times |\mathcal{R}_B\rangle
\end{aligned}
$$

- The zeroth-order wave function for $A \cdots B$ is

$$
|\Psi_{A+B}^{(0)}\rangle = |\Psi_A^{(0)}\rangle \times |\Psi_B^{(0)}\rangle
$$

Size extensivity and size consistency

Link with variational / projected method

- If we compute the energy by projection on $\langle \Psi^{(0)}_{A+B}|$ it gives

$$\langle \Psi^{(0)}_{A+B}|H|\Psi_{A+B}\rangle = \left(\langle \Psi^{(0)}_{A}| \times \langle \Psi^{(0)}_{B}|\right) H \left(|\Psi^{(0)}_{A}\rangle \times |\Psi^{(0)}_{B}\rangle\right) (= E_{\Psi^{(0)}_A} + E_{\Psi^{(0)}_B})$$

$$+ \left(\langle \Psi^{(0)}_{A}| \times \langle \Psi^{(0)}_{B}|\right) H|\Psi^{(0)}_{A}\rangle \times \left(\sum_{i=S,D}^{N^B_{SD}} c^B_i |i_B\rangle\right) (= E^B_{\rm corr})$$

$$+ \left(\langle \Psi^{(0)}_{A}| \times \langle \Psi^{(0)}_{B}|\right) H|\Psi^{(0)}_{B}\rangle \times \left(\sum_{i=S,D}^{N^A_{SD}} c^A_i |i_A\rangle\right) (= E^A_{\rm corr})$$

$$+ \left(\langle \Psi^{(0)}_{A}| \times \langle \Psi^{(0)}_{B}|\right) H \left(\sum_{i=S,D}^{N^B_{SD}} c^B_i |i_B\rangle\right) \times \left(\sum_{i=S,D}^{N^A_{SD}} c^A_i |i_A\rangle\right) (= 0)$$

- That's a $N^A_{SD} + N^B_{SD}$ CPU operation and storage ! ! ☺