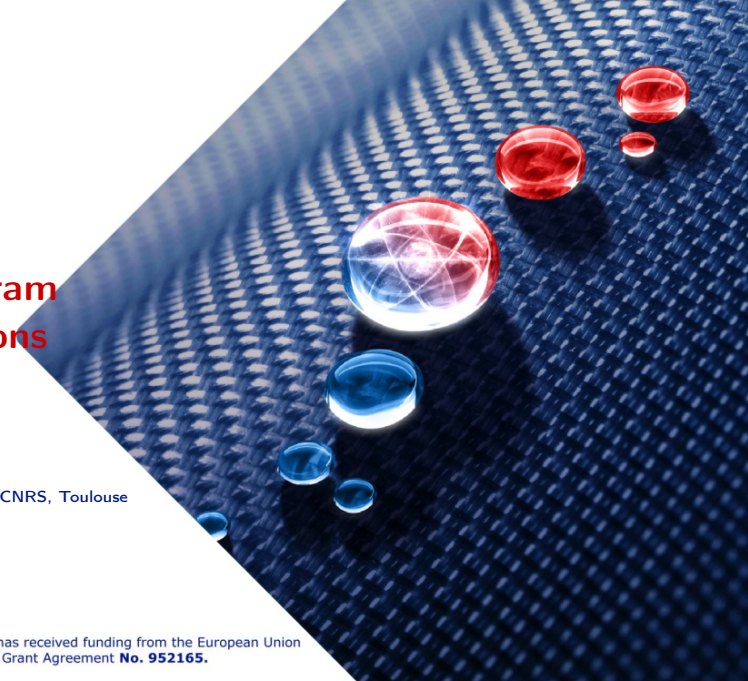
Targeting Real chemical accuracy at the EXascale

QMC=CHEM: a QMC program for large-scale simulations

Abdallah Ammar, Anthony Scemama

20/2/2024

Lab. Chimie et Physique Quantiques, IRSAMC, UPS/CNRS, Toulouse (France)



Targeting Real Chemical Accuracy at the Exascale project has received funding from the European Union Horizoon 2020 research and innovation programme under Grant Agreement **No. 952165.**

QMC=Chem

↬ original authors: Anthony Scemama and Michel Caffarel, Toulouse

- ➢ **VMC**, **DMC**, ... for CI-Jastrow wavefunction[1] ($\sim 10^6$ Slater determinants)
- ➢ very good single-core performance (`qmckl`[2]: optmimization with the William Jalby group, Versailles)
- ➢ full fault-tolerance implementation
- ➢ benchmarked on 76 800 CPUs on Joliot-Curie: 960 TFlops/s ($>$ 30% of peak performance)
- ➢ **CIPSI** (E. Giner talk) WF + FN-**DMC** (`trexio`[3]) lowest upper bound for the ground-state energy of H_2O[4]

[1] A. Scemama *et al*, *J. Comput. Chem.* **37** (2016)

[2] `https://github.com/TREX-CoE/qmckl`

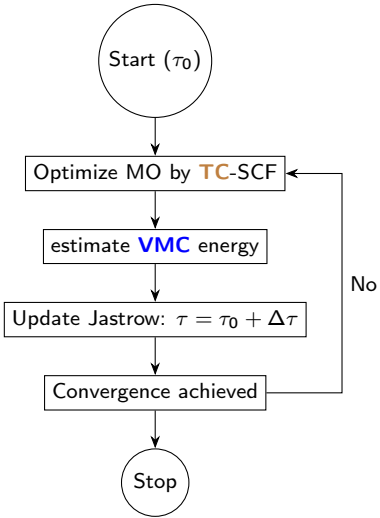
[3] `https://github.com/TREX-CoE/trexio`

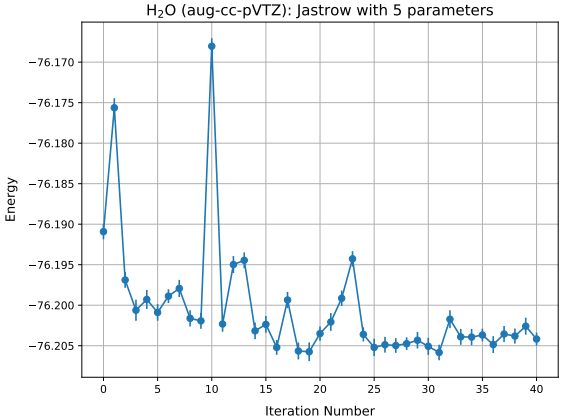
[4] M Caffarel *et al*, *J. Chem. Phys.* **144** (2016)

- ↣ CI-Jastrow is a powerful Ansatz: $\Psi = \Phi_{\text{CI}} \times e^{+\tau}$
- 🎯 **Goal**: Develop efficient algorithms to provide highly accurate and compact wavefunctions
- ↣ A fully stochastic optimization (MO, linear coefficients and Jastrow) is very expensive and tedious in practice
- ↣ **TC** theory to optimize CI-part **deterministically** and **VMC** to optimize the Jastrow **stochastically**

$$\begin{aligned}\hat{H}_{\text{TC}} &\equiv e^{-\hat{\tau}}\, \hat{H}\, e^{+\hat{\tau}} \\ &= \hat{H} + \hat{K}_{12} + \hat{L}_{123}\end{aligned}$$

➙ Slater-Jastrow Ansatz: $\Psi = \Phi \times e^{+\tau}$





Thank you for your attention

https://github.com/TREX-CoE/qmcchem2

Targeting Real chemical accuracy at the EXascale

This project has received funding from the European Union's Horizon 2020 - Research and Innovation program - under grant agreement no. 952165.