Targeting Real chemical accuracy at the EXascale

QMC=Chem

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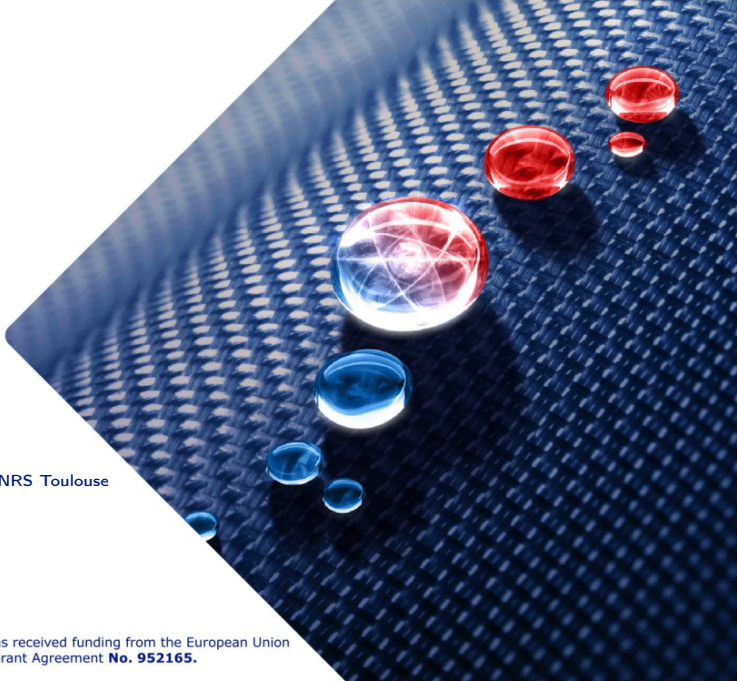

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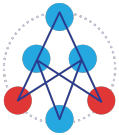


QMC=Chem

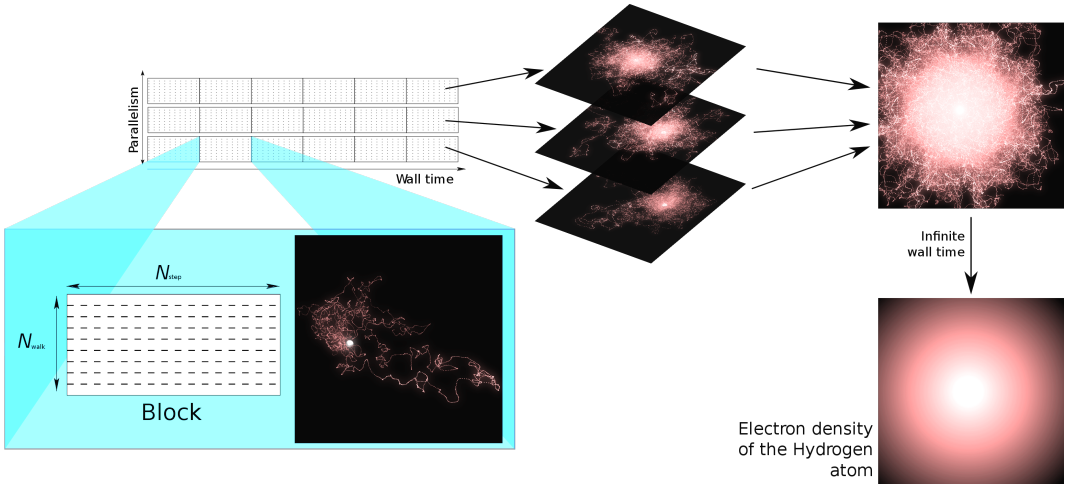
What it can do

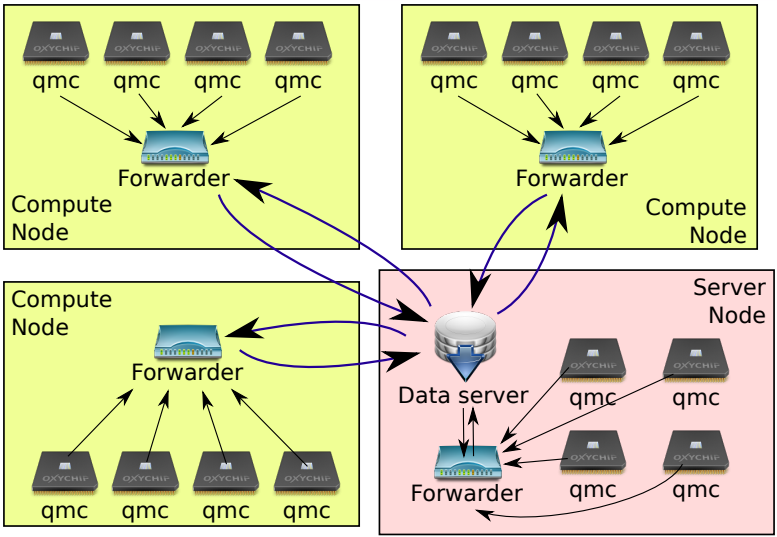
- VMC and DMC of large CI expansions
- Ground and excited states
- All-electron / Pseudo-potentials (DLA)
- Optimization of very large CI expansions
- Massively parallel calculations

What it cannot do

- Periodic systems
- T-moves
- Sophisticated Jastrow factors
- Backflow
- JAGP
- Nuclear forces

Algorithm


Parallelism
Wall time
N_{step}
N_{walk}
Block
Infinite
wall time
Electron density
of the Hydrogen
atom


qmc
qmc
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qmc
Forwarder
Compute
Node
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Forwarder
Compute
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Compute
Node
Forwarder
qmc
qmc
qmc
qmc
Server
Node
Data server
qmc
qmc
Forwarder
qmc
qmc

Programming

- Wave function preparation: Quantum Package
- Computation: Fortran
- Communication + post-processing: OCaml
- ZeroMQ for communication $\Longrightarrow$ HPC systems + Cloud infrastructures
- Fault tolerance + dynamical resource management

Large scale applications

- 0.96 PFlops/s in 2011 on Curie (TGCC/CEA), 76 800 cores
- Distributed HPC+Cloud calculations in 2015 (France Grilles)

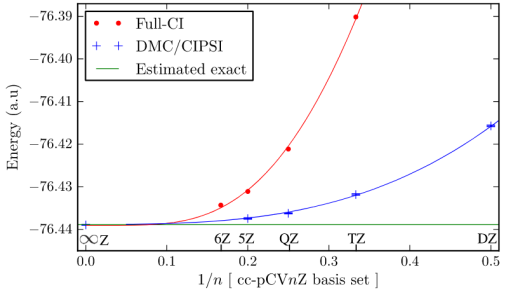

FIG. 1. CBS extrapolation of FCI and DMC/CIPSI energies. Error bars on DMC data are plotted but almost imperceptible.

TABLE III. Comparison of nonrelativistic ground-state total energies of water obtained with the most accurate theoretical methods. Energies in a.u.

Clark *et al.*,[20] DMC (upper bound)	−76.436 8(4)
This work, DMC (upper bound)	−76.437 44(18)
Almora-Dìaz,[27] CISDTQQnSx (upper bound)	−76.434 3
Helgaker *et al.*,[29] R12-CCSD(T)	−76.439(2)
Muller and Kutzelnigg,[30] R12-CCSD(T)	−76.437 3
Almora-Dìaz,[27] FCI + CBS	−76.438 6(9)
Halkier *et al.*,[31] CCSD(T) + CBS	−76.438 6
Bytautas and Ruedenberg,[32] FCI + CBS	−76.439 0(4)
This work, DMC + CBS	−76.438 94(12)
Experimentally derived estimate[25]	−76.438 9

Toward an improved control of the fixed-node error in quantum Monte Carlo: The case of the water molecule
M. Caffarel, T. Applencourt, E. Giner, A. Scemama, *J. Chem. Phys.*, 2016, 144, pp.151103.

References

- QMC=Chem: A Quantum Monte Carlo Program for Large-Scale Simulations in Chemistry at the Petascale Level and beyond
- Quantum Monte Carlo for large chemical systems: Implementing efficient strategies for petascale platforms and beyond
- Quantum Monte Carlo with very large multideterminant wavefunctions
- Optimization of large determinant expansions in quantum Monte Carlo
- Toward an improved control of the fixed-node error in quantum Monte Carlo: The case of the water molecule
- Deterministic construction of nodal surfaces within quantum Monte Carlo: the case of FeS
- Excitation energies from diffusion Monte Carlo using selected configuration interaction nodes
- Taming the fixed-node error in diffusion Monte Carlo via range separation