MOLPRO : overview and challenges

Bastien Mussard, Julien Toulouse
LCT, ICS
Université Pierre et Marie Curie, Paris, France

János Ángyán
CRM^2
Université de Lorraine, Nancy, France

MOLPRO : overview and challenges

Bastien Mussard, Julien Toulouse
LCT, ICS
Université Pierre et Marie Curie, Paris, France

János Ángyán
CRM^2
Université de Lorraine, Nancy, France

MOLPRO

Molpro : a general-purpose quantum chemistry program package
Werner H.-J., Knowles P. J., Knizia G., Manby F. R., Schütz M.
WIREs Computational Molecular Science **2** 242 (2012)

and User's Manual at
www.molpro.net

Methods and Characteristics

- HF
- DFT (SAPT)
- CI
- CC (MRCC interfaced)
- RPA

- MCSCF
- CASSCF
- MRPT/CASPT
- MRCI

- FCIQMC (NECI)
- DMRG
- *-F12
  - Local-*
  - DF-*

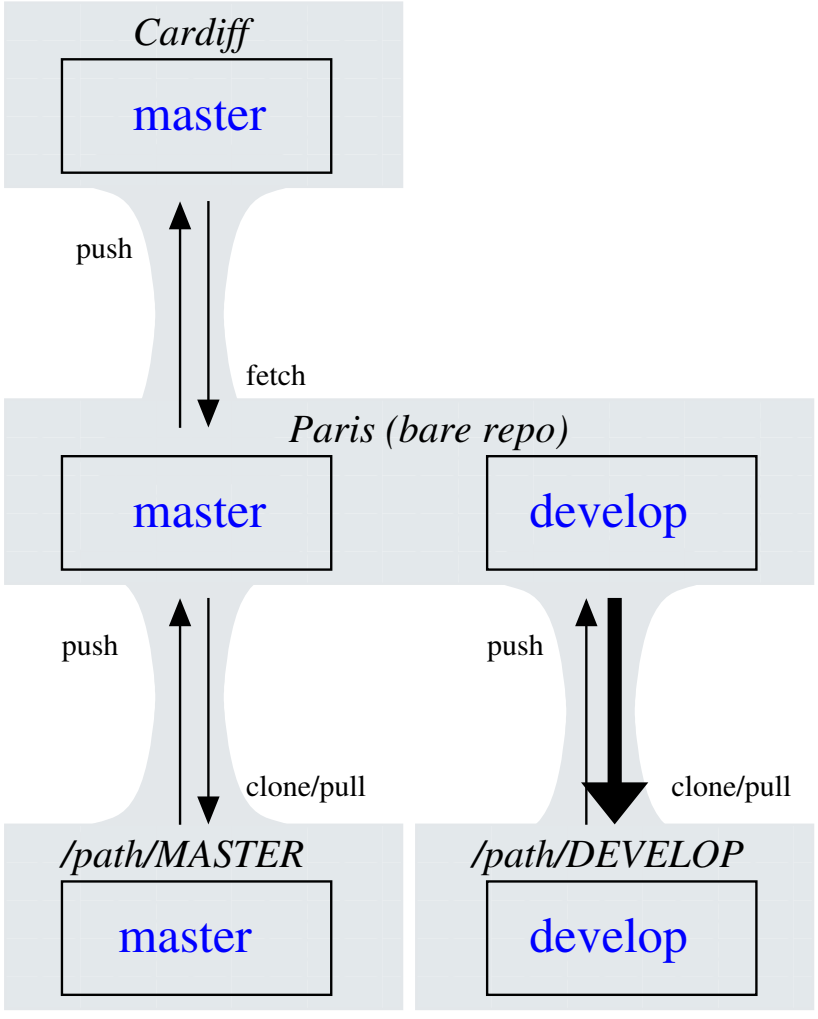
- ~4 000 000 lines of code
- Integral stored on disk or recomputed when needed
- Fast orbital optimisation algorithm
- Gradients and Geometry Optimization
  (analytical or clever numerical)
  Electrical/Vibrational/Magnetical properties

- Density Fitting and very performant Local Approximations
- Parallel implementations (PPIDD on top of GA or MPI, 10^1 cores)

Performance and Range of Applications

DFT	<1min for a 59-atoms molecule with vTz
SAPT	H_2-C_{60} and DNA pairs
DF-LMP2	4000 basis functions
LMP2-F12	200 atoms
CC	approx. 100 atoms
LCCSD(T)	2600 basis functions
LCCSD(T)-F12	100 atoms
MRCI	$O_2Cu_2(NH_3)_6$
	584 basis fcts, 3540 CSFs, 80 correlated electrons
	41 min CPU time per iteration
	exclusive (⚠: en 2012)

Collaborative Development



- Developing a code that is not "ours" (constraint/advantage, bugzilla, ...)
- GIT setup
- Main repository in Cardiff & repository in Paris (has a "develop" branch)

The `rpatddft` code – **very well documented** (in section 17.7)

- RPA correlation energies
- Dynamic polarizabilities
- C_6 dispersion coefficients
- Excitations energies

- Spin-unrestricted versions available
- (with or without) range separation
- Gradients for most versions

Challenges

- {done} Two-electrons integrals transformation (from `MOLPRO`)
- {unproblematic} Use of spatial symmetry

Our main challenges lie with linear algebra
(matrix inversion, matrix power, diagonalisation, ...)

Solutions :

- {new developments} Approximations (Density Fitting, Local, ...)
- {optimal use of existing `MOLPRO` solution} Parallel routines (⚠: 10^1)
- {other parallel solutions}