I.
A priori modeling of chemical reactions on a Grid based virtual laboratory

Towards standard representations of data for molecular chemistry

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Outline

Introduction

Elementary exchange reactions

qGEMS, the workflow

The Q5Cost and D5Cost data models

A prototype reaction: $H + H_2$

Outline

Modeling chemistry

Simulating processes on a molecular basis

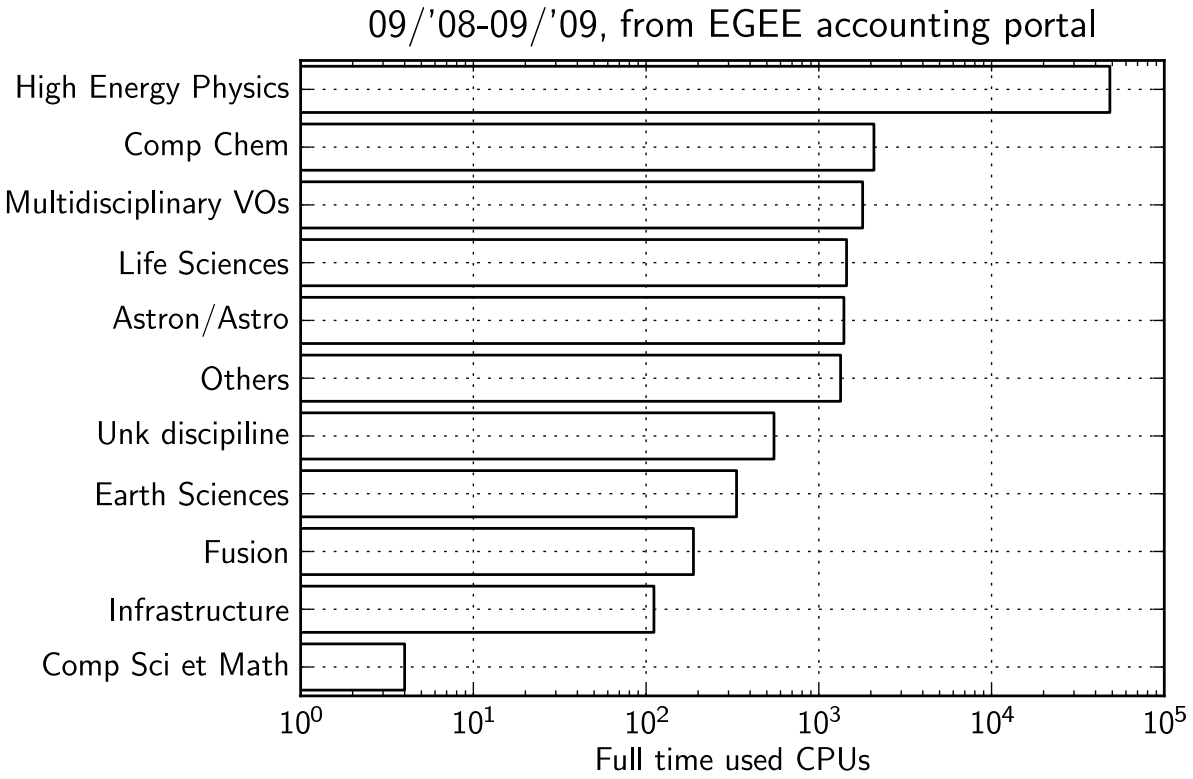
- modeling natural phenomena
- designing new materials
- mastering new technologies

. . . requires

- assembling various pieces of software
- converging different competences
- a world spread virtual laboratory

. . . on the Grid:

A self-introducing picture



The Grid Enabled Molecular Simulator

GEMS is

A grid based realistic simulator that can act as a molecular science engine in complex multiscale chemical contexts.

The recipe

- software: a suite of codes
- interoperability: standards and tools
- a director: workflow management
- a factory: Grid, the modern paradigm of HTC

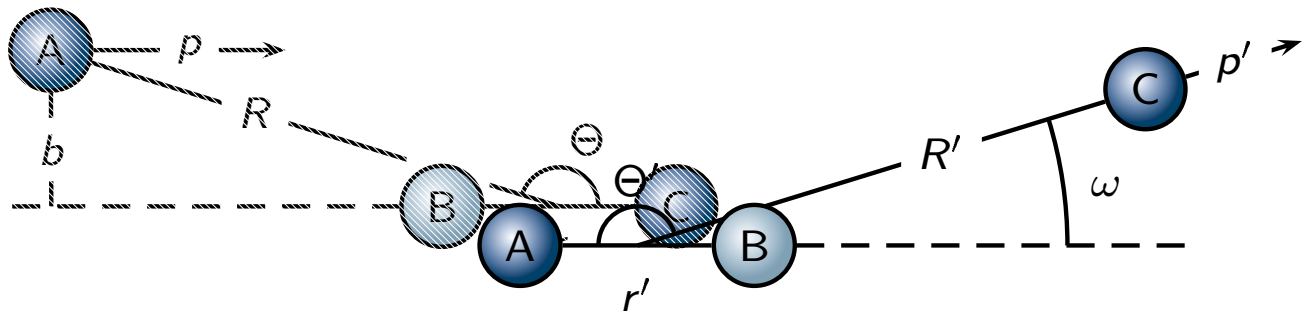
Let's start with a few atoms...

Outline

A + BC, the quantum view

A reactive collision

$$A + BC\ (v,j) \rightarrow AB\ (v',j') + C$$



The B-O "equation of motion"

$$i\hbar\frac{\partial\psi(\mathbf{w},t)}{\partial t} = \left[\hat{T}_{\mathbf{w}} + V(\mathbf{w})\right]\psi(\mathbf{w},t)$$

A + BC, solution methods

If $\hat{H} \neq f(t)$, then either (TD methods)

$$\psi(\mathbf{w}, t+\tau) = e^{-\frac{i\hat{H}\tau}{\hbar}}\psi(\mathbf{w}, t)$$

Or simply (TI methods)

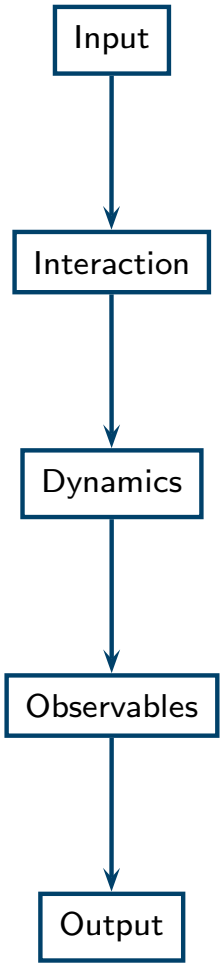
$$\hat{H}\psi(\mathbf{w}) = E\psi(\mathbf{w})$$

From an analysis on ψ

The detailed scattering matrix elements $S^J_{cv'j'k',avjk}(E)$

Outline

Overview



Interaction

- evaluate $V(\mathbf{w})$

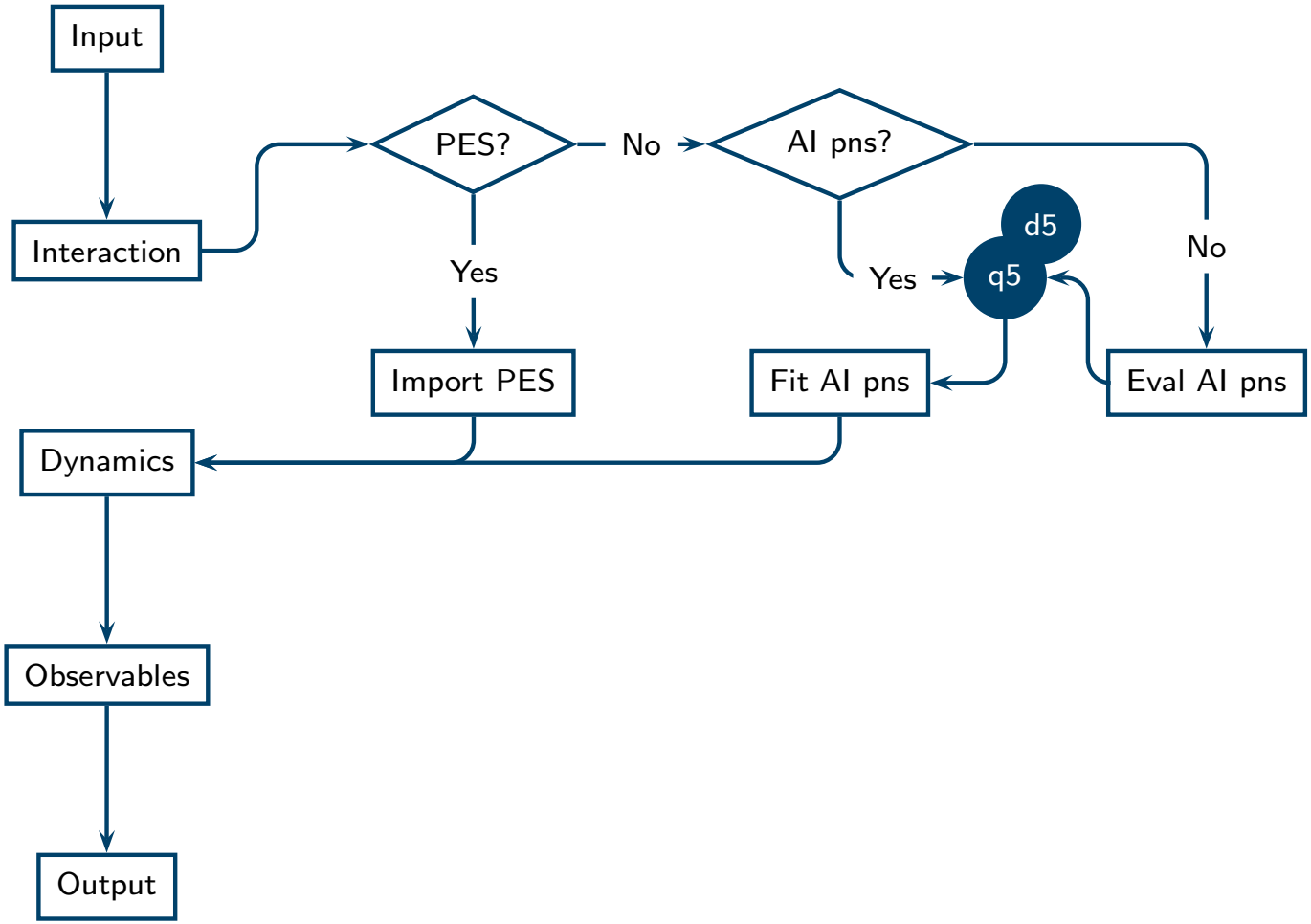
Dynamics

- solve the equation of motion

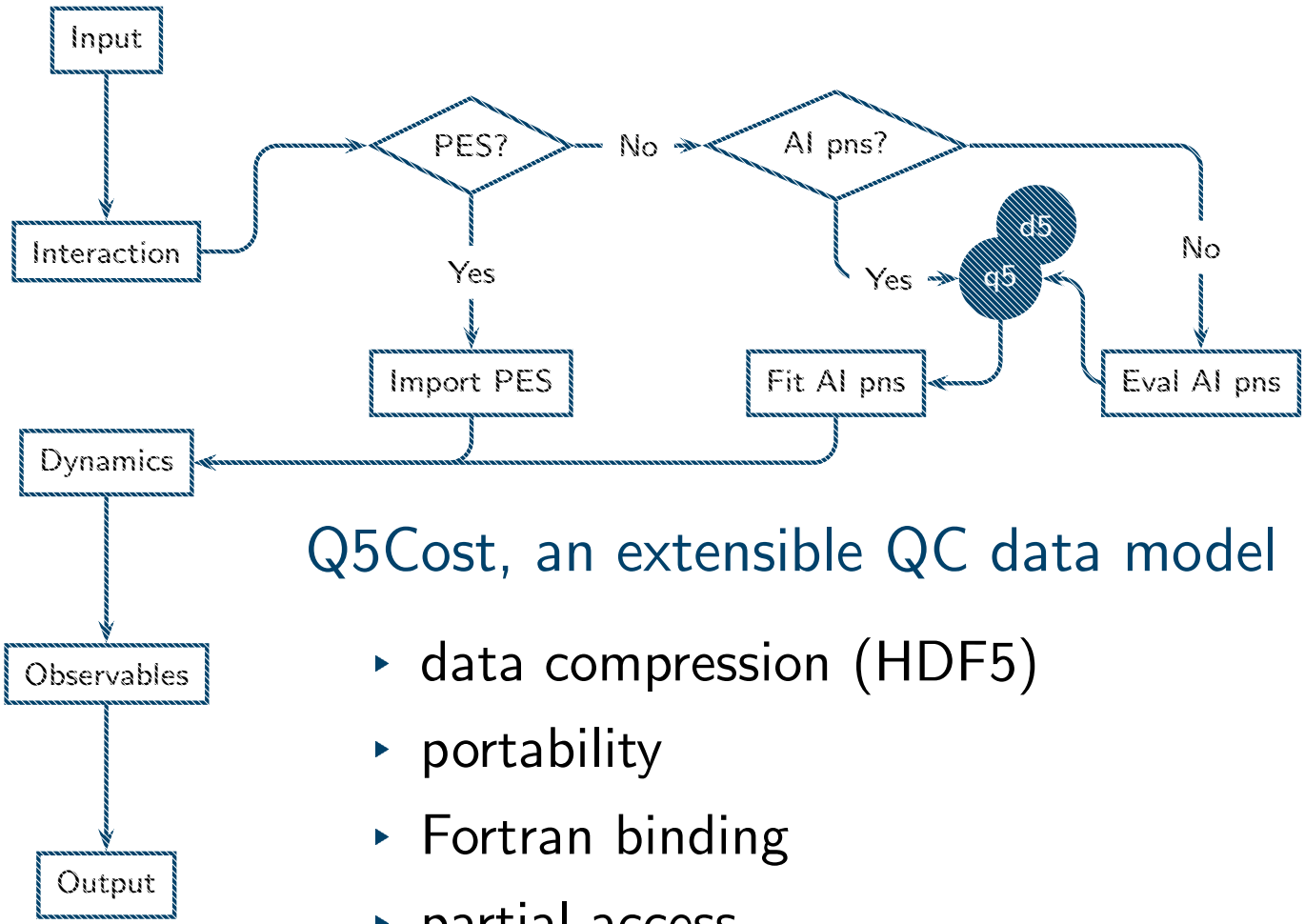
Observables

- average and get observable properties

qGEMS, Interaction



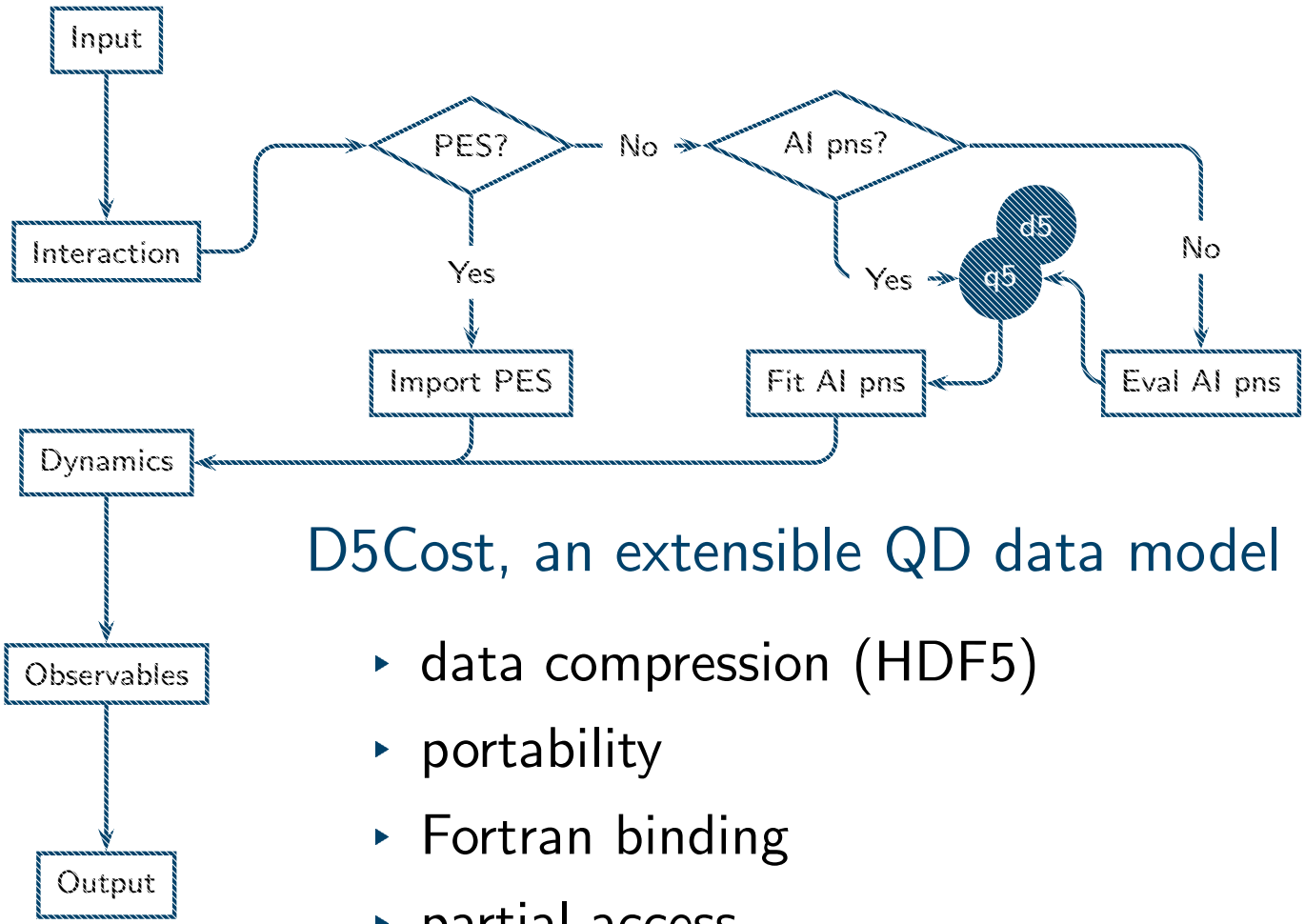
interlude, Q5Cost



Q5Cost, an extensible QC data model

- data compression (HDF5)
- portability
- Fortran binding
- partial access
- all QC info, at 1 **w**

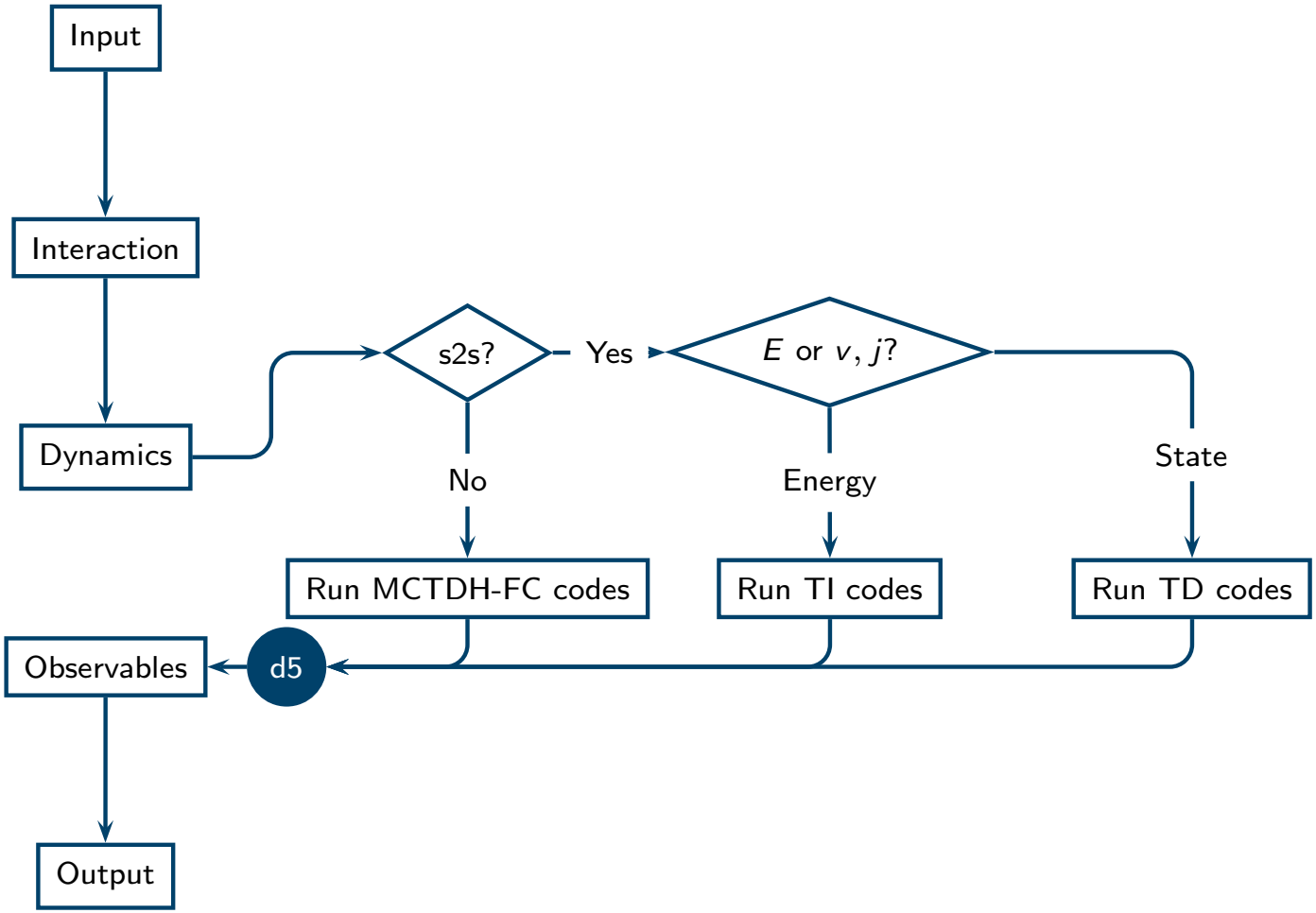
interlude, D5Cost



D5Cost, an extensible QD data model

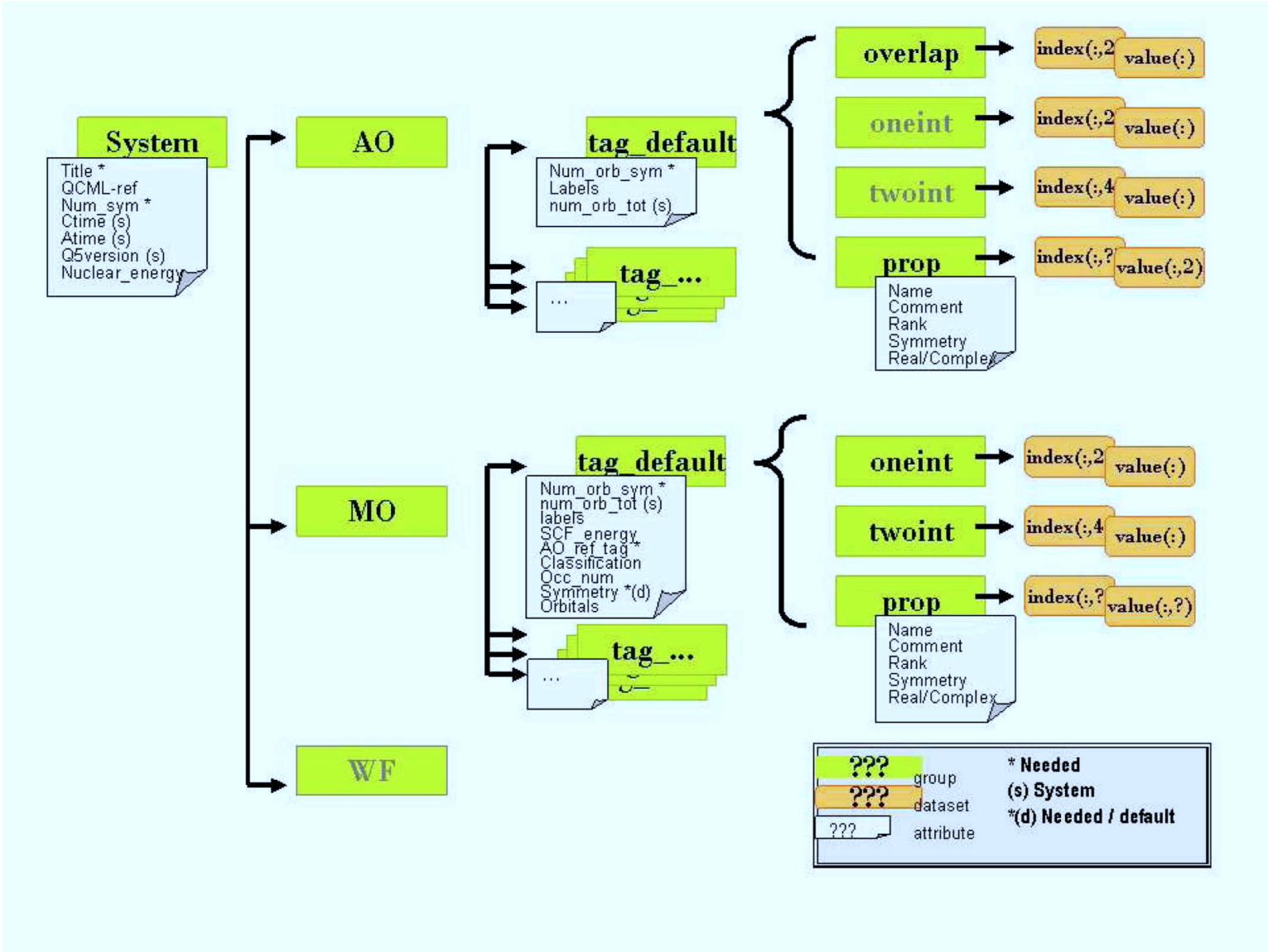
- data compression (HDF5)
- portability
- Fortran binding
- partial access
- V, ∇, $\mathbf{H}$, at many $\mathbf{w}$'s

qGEMS, Dynamics



Outline

Q5Cost, data model



Q5Cost, library

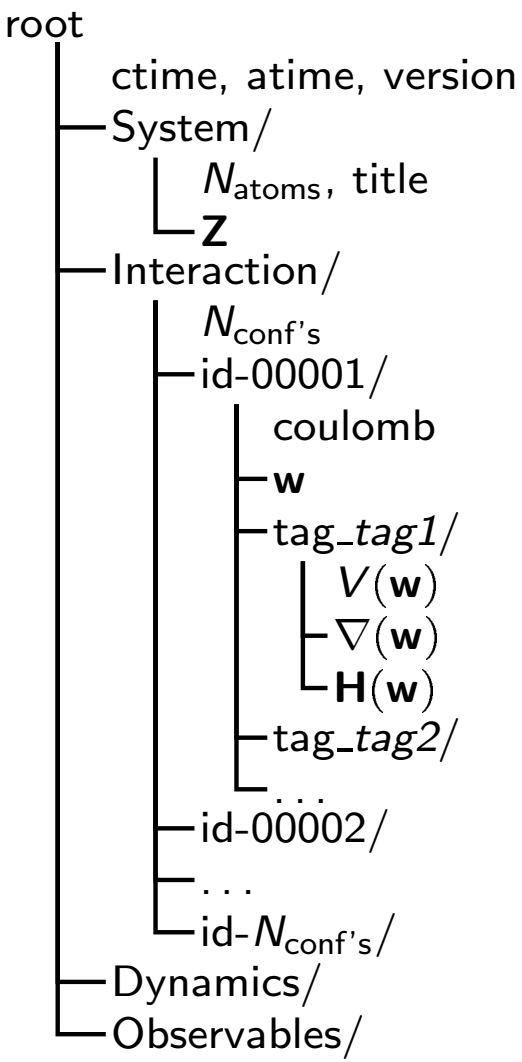
Overview

- conceived as a high level tool for quantum chemists
- based on HDF5 technology
- provides r/w access to .q5 files

Structure

- Q5Cost: API "speaking" chemistry
- Q5Core: wrapping facilities for HDF5 routines
- Q5Error: error handling

Towards D5Cost



Data model

- only System and Interaction defined at present
- abstracts from 3-body systems

Library

- D5Cost built on top of Q5Core and Q5Error
- provide r/w access to .d5 files

Usage: sample program

```
PROGRAM d5test
   USE D5cost
   IMPLICIT NONE
   ...
   CALL D5cost_init(error)
   d5file_name = "test.d5"
   CALL D5Cost_file_open(d5file_fname,d5file_id,error)
   CALL D5Cost_System_get_num_atom(d5file_id,num_atom,error)
   ...

   CALL D5Cost_<Domain>_<Action>_<Target>(...)

   ...
   CALL D5Cost_file_close(d5_id,error)
   CALL D5cost_deinit(error)
   STOP
END PROGRAM d5test
```

Outline

$H + H_2$, computing details

DALTON

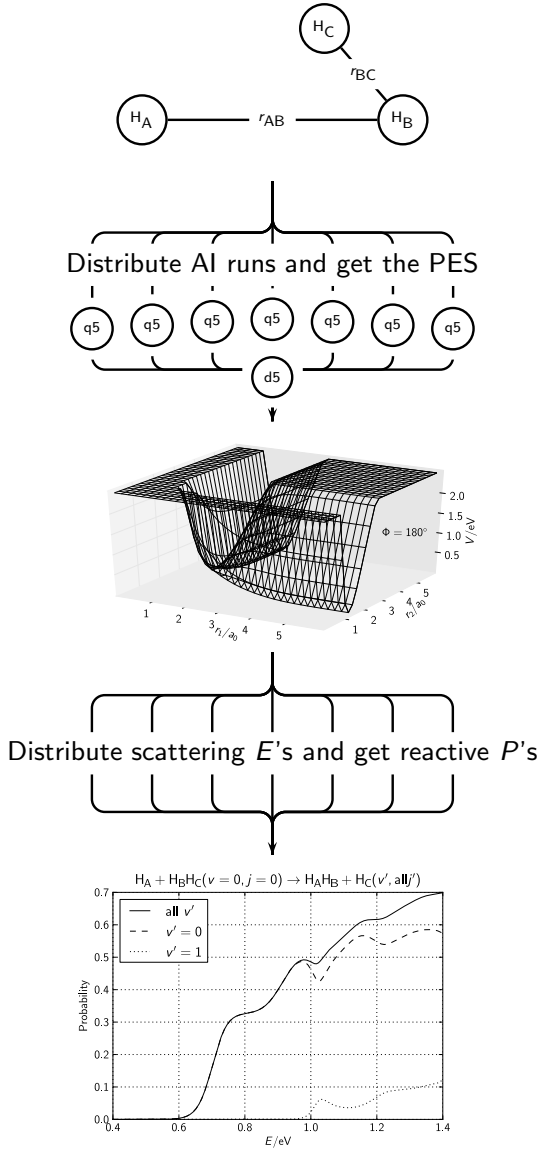
- 23 H_2 and 270 H_3 **w**'s
- NEVPT2/cc-pV5Z

GFIT3C

- 3C polynomial fit, degree 10
- rms, 0.19 kcal/mol

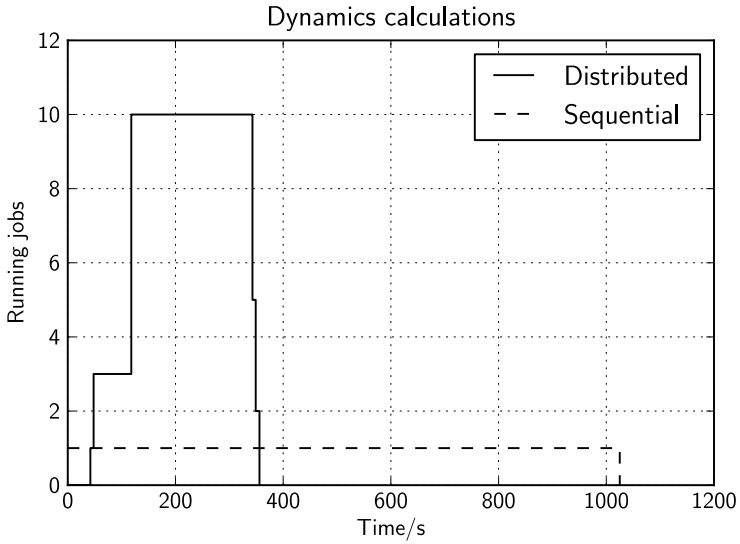
ABC

- 1000 null-J fixed-E runs
- 100 E's per 10 CE's



$H + H_2$, performances

The dynamics: execution time

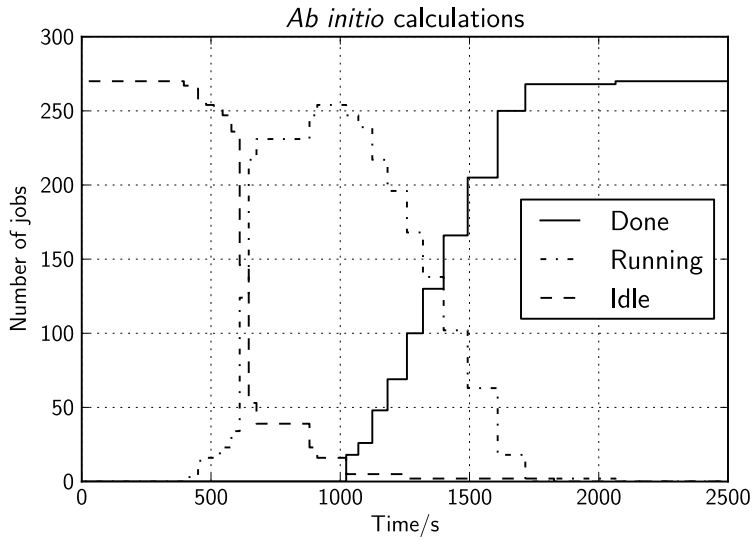


Heavier systems

- the solid box shall grow high
- the dashed box shall lenghten rightward

$H + H_2$, performances

AI runs: a little better...



However

- computation grain still too fine
- sequential execution time over 30000 s

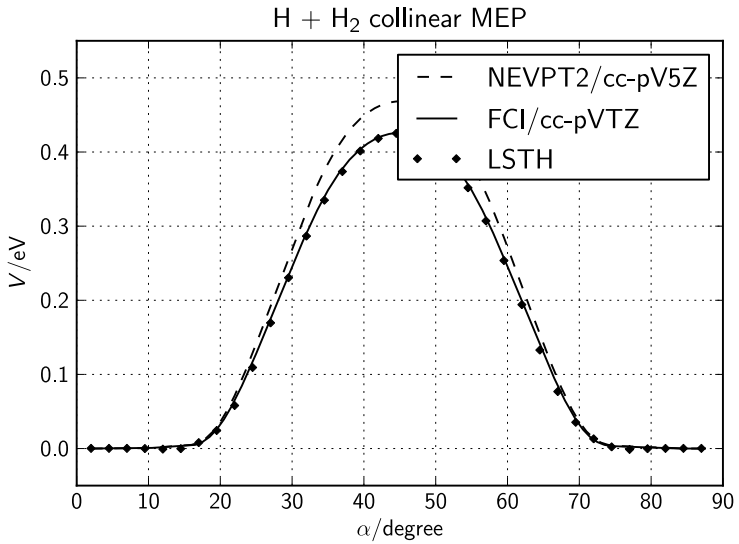
$H + H_2$, accuracy

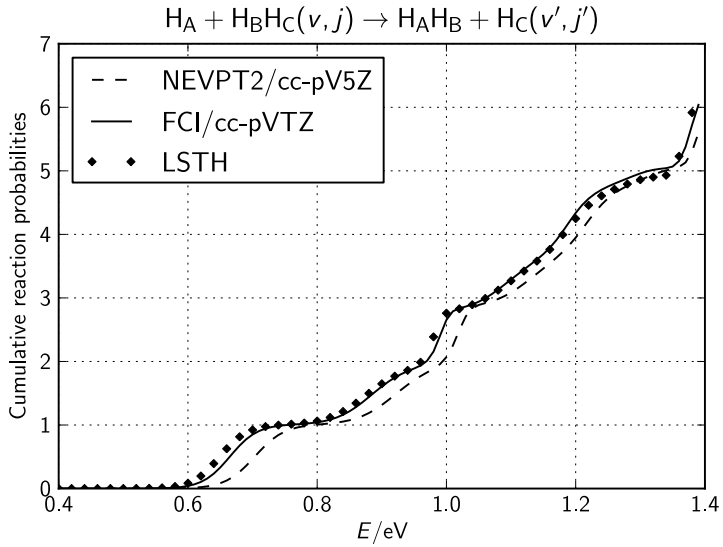
vs popular LSTH

- same MEP shape
- but shallow VdW's

Global reactivity

- same CRP's shape
- — matches ◆'s





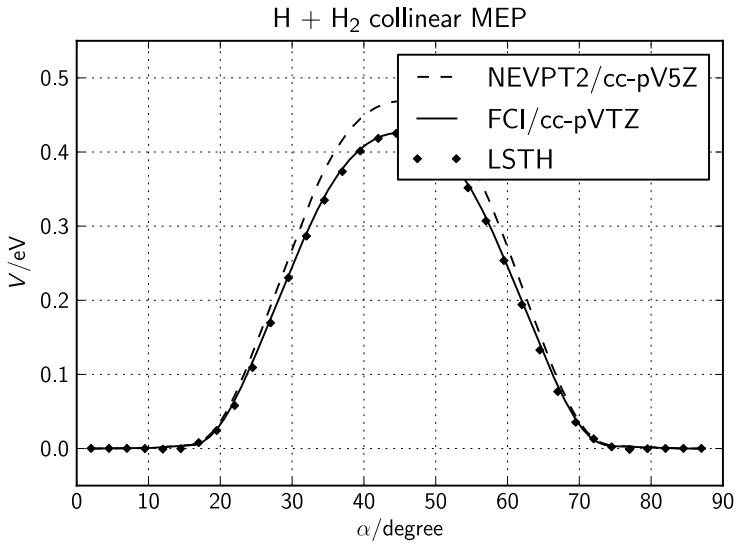
$H + H_2$, accuracy

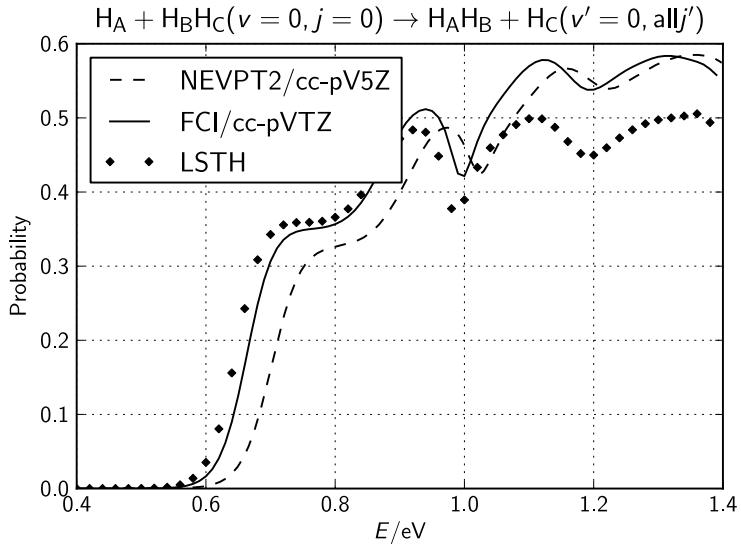
vs popular LSTH

- same MEP shape
- but shallow VdW's

Detailed reactivity

- non-ME paths weigh
- or VdWs?





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