Ab initio photodynamics calculations on the Grid: approaches and applications

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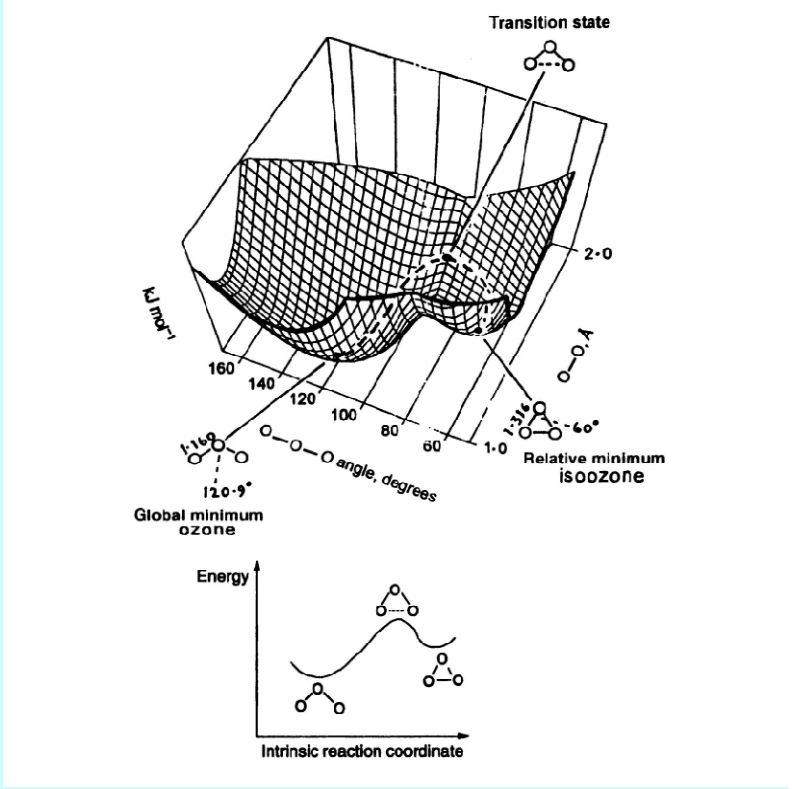
Quantum Chemistry and Molecular Simulations

- Computation of molecular properties: structures and spectroscopical data
- Chemical dynamics
- Energy surface plays a fundamental role
- Quantum Chemistry has made dramatic progress in accurate calculations

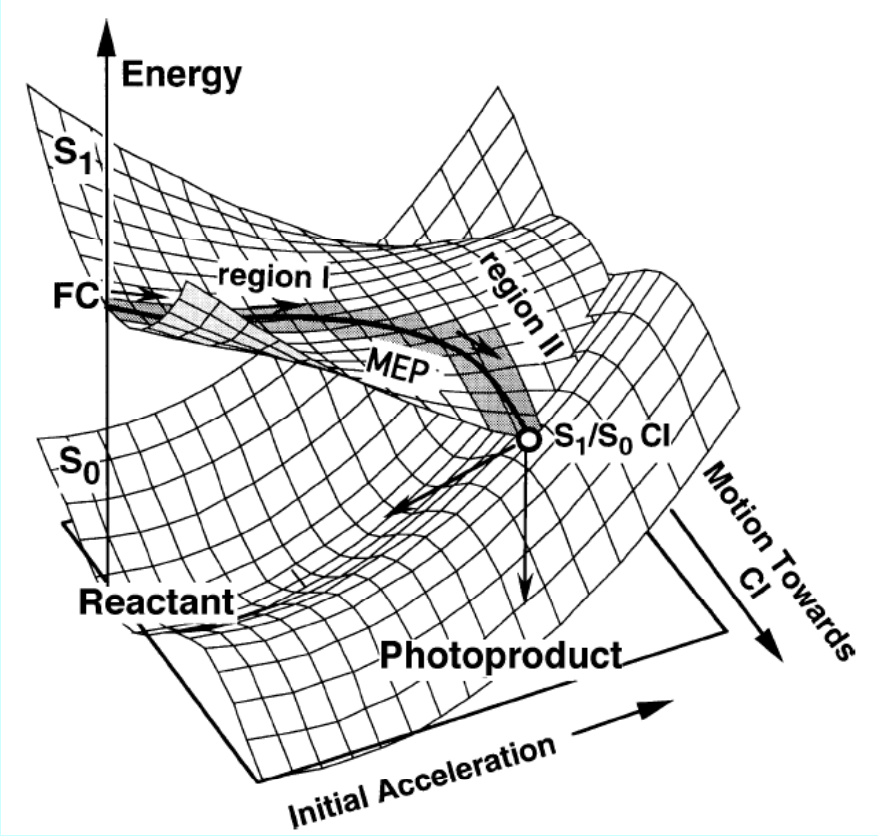


Advances in Quantum Chemical Methodology and Applications

- Isolated molecules
  - Fixed geometry
  - Geometry relaxation and vibrational frequencies
  - Reaction dynamics
  - Electronic ground state and excited states
- Solid state
  - Bulk phase and adsorption processes on surfaces
  - Geometry relaxation and vibrational frequencies
  - Dynamics
- Solution
  - Most difficult case (no periodicity), many methods still in development

Photochemistry

- Several electronic states at the same time
- Theoretical framework very difficult



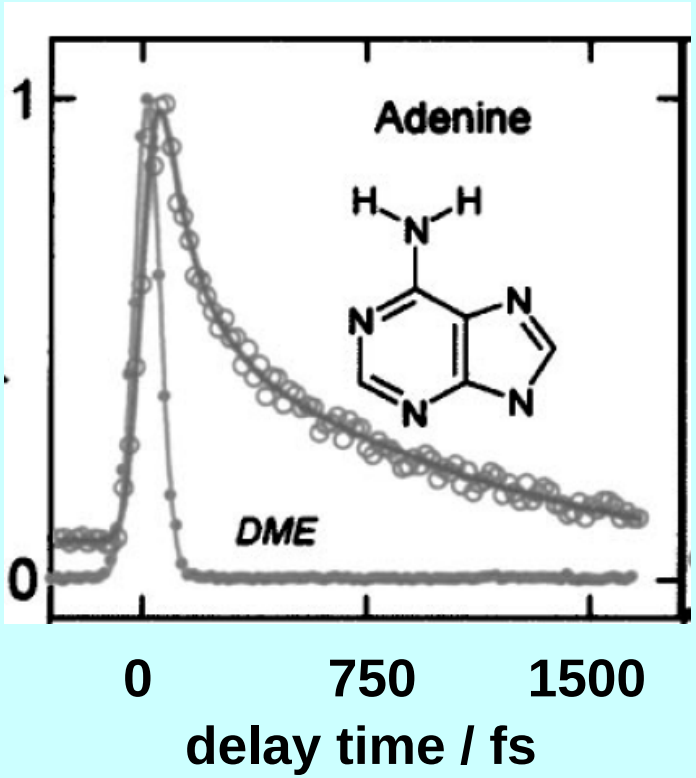
Cembran et al. JACS **126**, 16018 (2004)

Photostability of DNA Bases

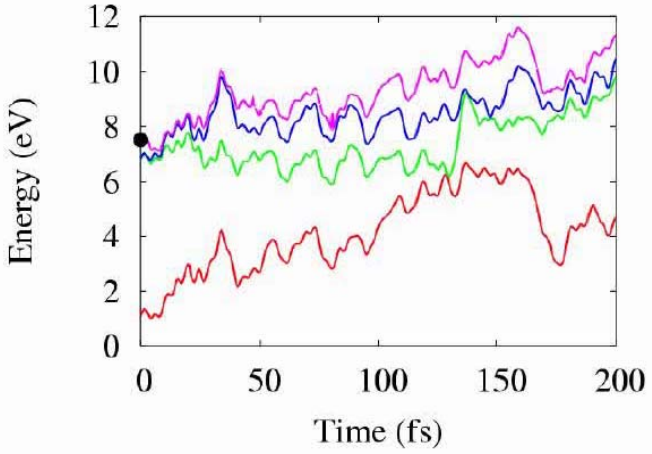
- What is the mechanism?
- Is this property important for the evolution processes in pre-biotic stages of the earth?

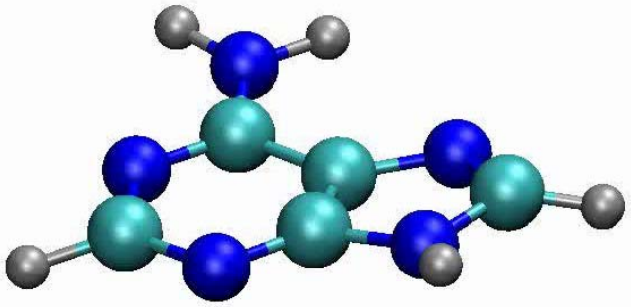
Adenine as example:

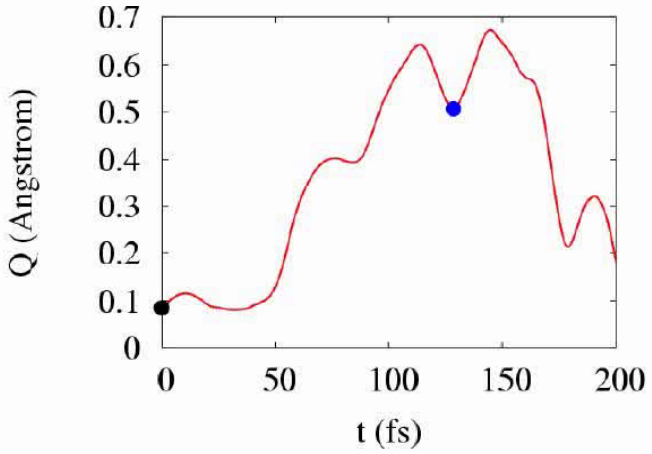
Ultrafast (femtosecond) laser experiments

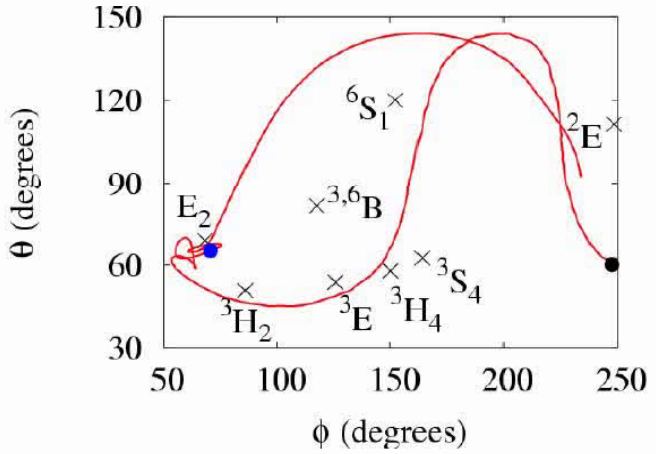


Adenine Dynamics









Methods and Computer Programs

COLUMBUS
- MRCI, CASSCF
- Analytical gradients
- non-adiabatic couplings

www.univie.ac.at/columbus

NEWTON-X
- Classical and mixed quantum-classical dynamics (surface hopping)
- Interfaces to COLUMBUS, TURBOMOLE, DFTB

www.univie.ac.at/newtonx

Public domain programs

COLUMBUS has more than 500 registered users

Columbus: Technical Details

- Quantumchemical Ab-Initio Software for high-level Calculations
- 64 executables
- Choice of BLAS and LAPACK influences performance strongly
  - Execution time reduced by a factor of two when using GOTO-blas instead of GNU-libblas
- Compiler Optimization gives ~10% speedup

Columbus

- Huge (but mostly sparse) symmetric matrices to be handled
  - Dimension up to 1 billion
- Execution time ranging from minutes to weeks
  - Mostly 1h - 24h
- Memory requirement ≥ 2GB per node
- Temporary diskspace ~20 MB to several GB

Columbus

- Installation platform dependent
- Unsupervised compilation unfavorable
- Validation procedure not fully automatized (yet)
- Static linked executables for some, but not all platforms
- Diskspace-requirement ~100MB for executables

Columbus

System	linked executables available
Intel PIV IA-32 Linux	Yes, static
Intel PIV EM64T Linux	Yes, static
Intel Itanium-2 Linux	Yes, static
Intel Core Duo Linux	Yes, static using cores single

System	linked executables available
Intel Core2Duo/Quad Linux	Yes, dynamic using cores single
Athlon64 Linux	Yes, only GNU-blas
Opteron64 Linux	Yes, static
Opteron64 Solaris	Yes, dynamic

Newton-X: Technical Details

- Ab-Initio Molecular dynamics package
- Excited-state- and nonadibatic dynamics
- Requires third-party ab-initio program for energy and gradient
  - Columbus
  - Turbomole
  - ...

Newton-X

- Calculation of many **completely independent trajectories** (Tully's surface-hopping on-the-fly)
- In each time step of a trajectory call of the quantum chemical program (COLUMBUS, TURBOMOLE,...) – sequential ordering!
- Hundred thousands of calls times hundreds of trajectories $\Rightarrow$ **extended computational campaigns** composed of QC jobs with **high computational demands**

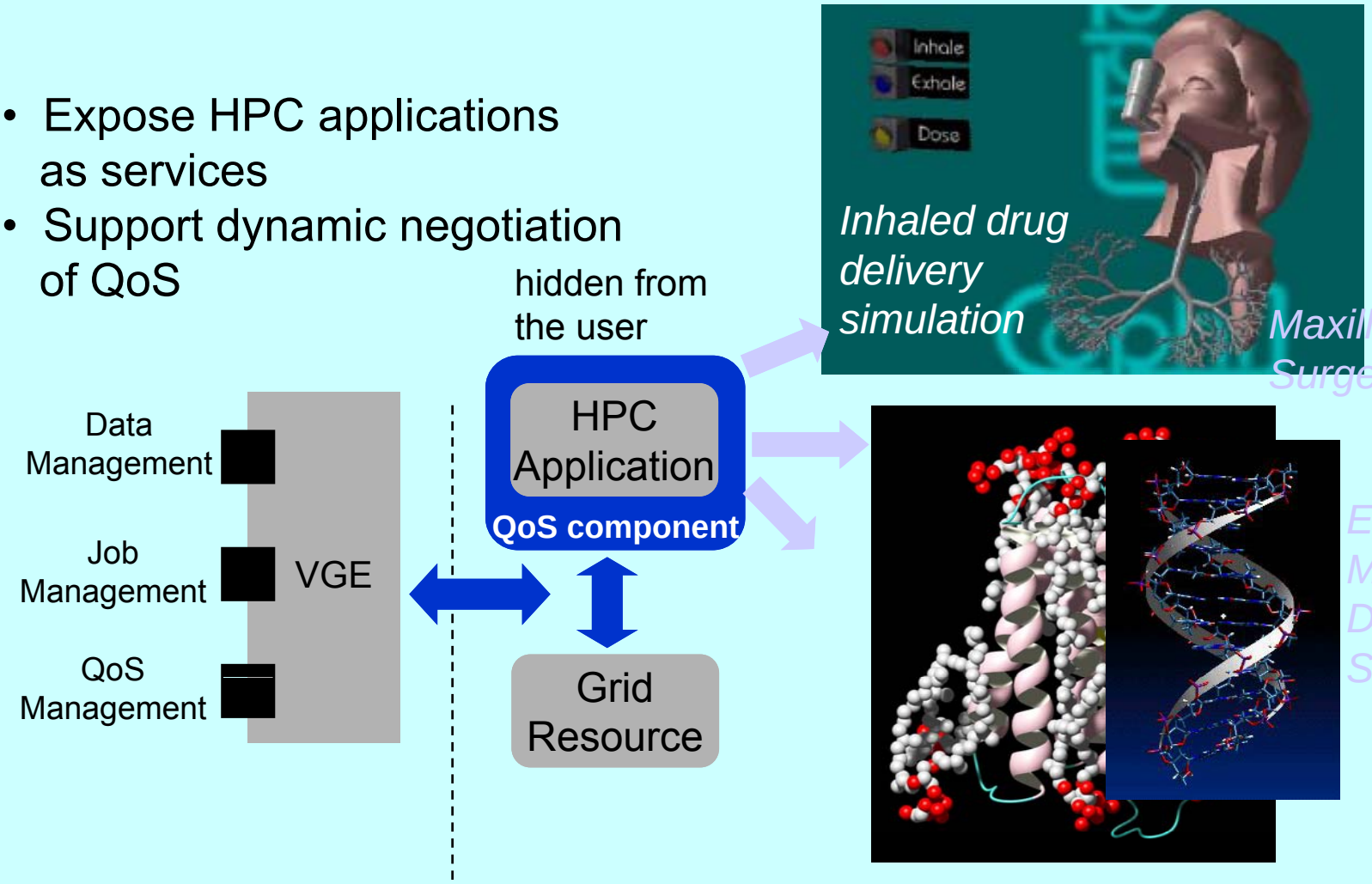
Strategy

- Each QC (COLUMBUS/TURBOMOLE) job step requires high-speed interprocess connection $\Rightarrow$ sequential job on a single node or parallelization (no screen-saver application)

  Parallel COLUMBUS implementation e.g. on JUMP and JUBL (Blue Gene L) at the John von Neumann-Institute for Computing in Jülich
- Grid management for computational campaigns

Vienna Grid Environment

- Expose HPC applications as services
- Support dynamic negotiation of QoS



EGEE

- **E**nabling **G**rids for **E**-Scienc**E**
- Globus/Condor based Grid (→gLite)
- Over 30.000 CPUs
- Over 5 PByte of Storage
- Virtual Organisations
  - CompChemVO (University of Perugia)
- Columbus now available

EGEE: First Experiences

- Automatic Grid installation difficult, finally successful (more information from Osvaldo Gervasi), more experience has to be gained
- COLUMBUS calculations in Perugia successful
- Extension to other Grid segments planned for the next future
- Experience for implementation of other QC program packages $\Rightarrow$ COST Chemistry D37 GRIDCHEM

Summary

- Columbus/Newton-X deployed on EGEE Grid and as VGE-Service (LUNA-cluster, QCCD-Cluster)
- Execution of extensive computational campaigns seem to be possible
- More has to be done on workflows when more program packages are included

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