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Cubic vers.

$O(N)$ vers.

Bridging the gap

Facilitating processing

Outlook

Max Conference on the Materials Design Ecosystem at the Exascale: High-Performance and High-Throughput Computing

TRIESTE

Adaptive and Localized Basis Functions for Linear Scaling, Large Systems and Complex QM Simulations

Luigi Genovese, Stephan Mohr, L. Ratcliff, D. Caliste, S. Goedecker, **T. Deutsch**

INAC – CEA Grenoble

January 30, 2018

BigDFT: A DFT code based on Daubechies wavelets

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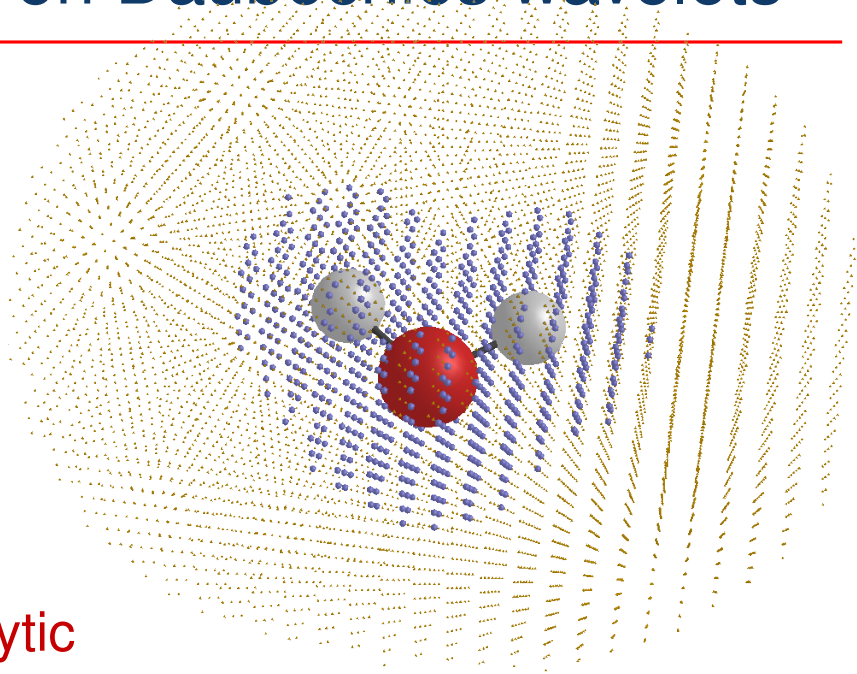
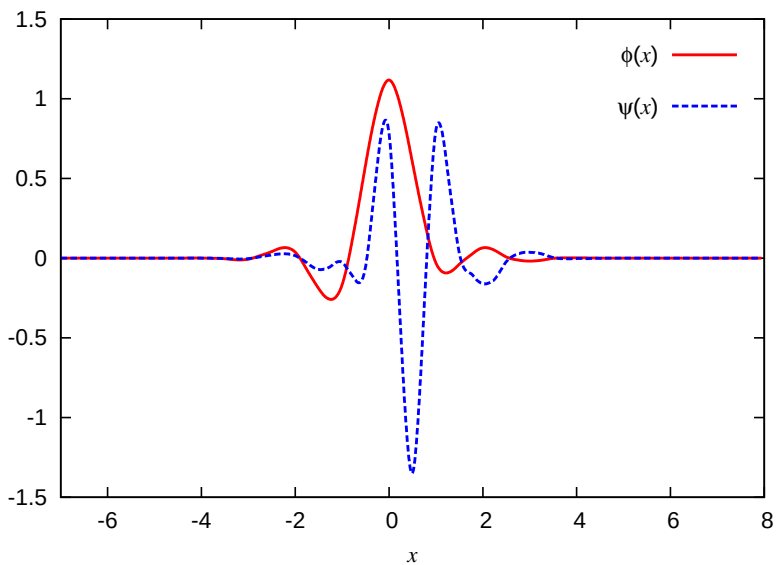
A pseudopotential Kohn-Sham code

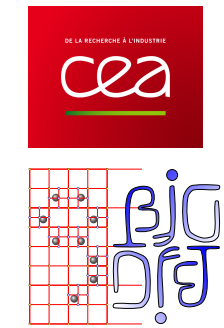


Daubechies wavelets have unique properties for DFT usage

- Systematic, Orthogonal
- Localised, Adaptive
- Kohn-Sham operators are analytic
- Efficient Poisson solver, capable of handling different boundary conditions – free, wire, surface, periodic
- Explicit treatment of charged systems



Adaptivity of the mesh



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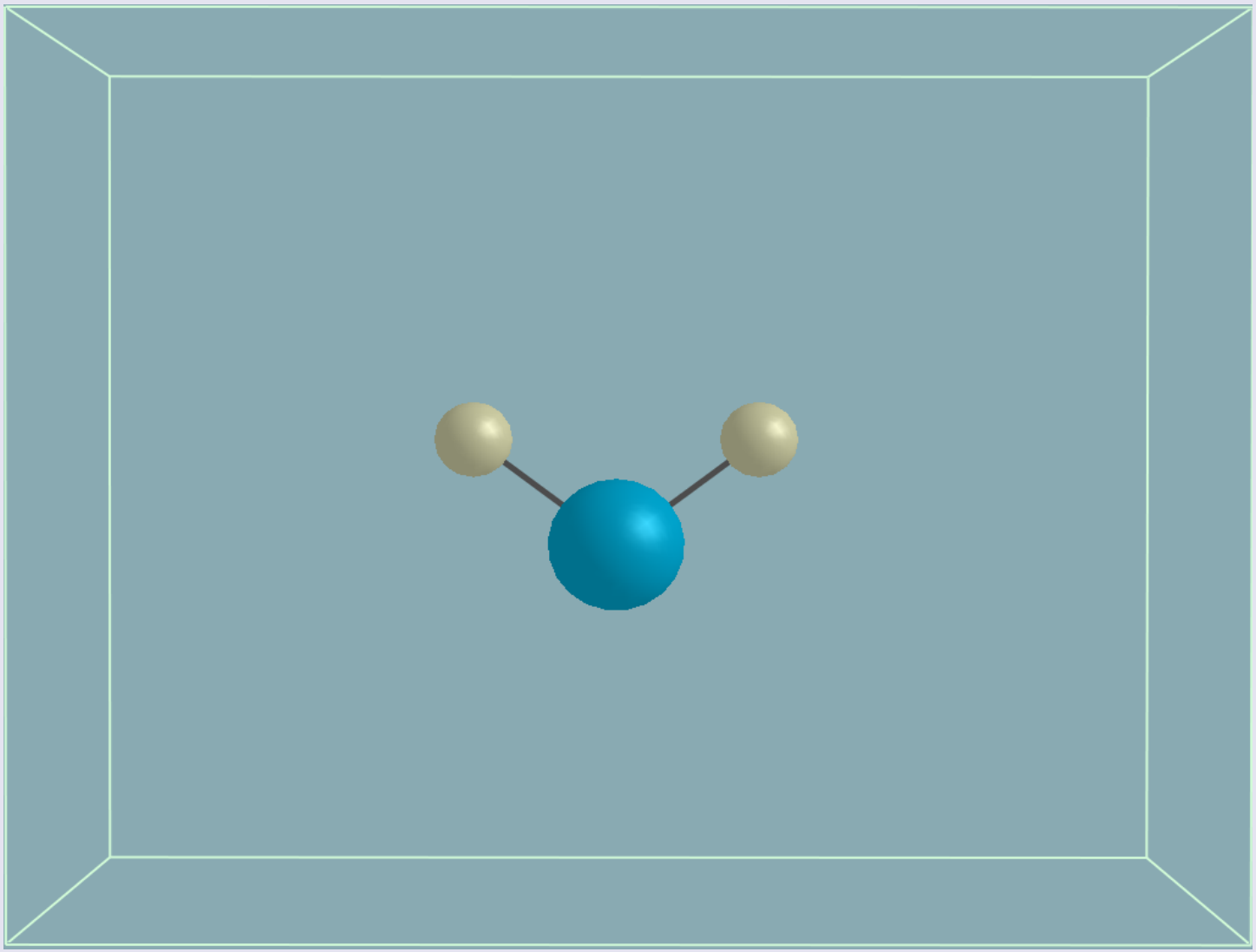
Atomic positions (H_2O)



Adaptivity of the mesh

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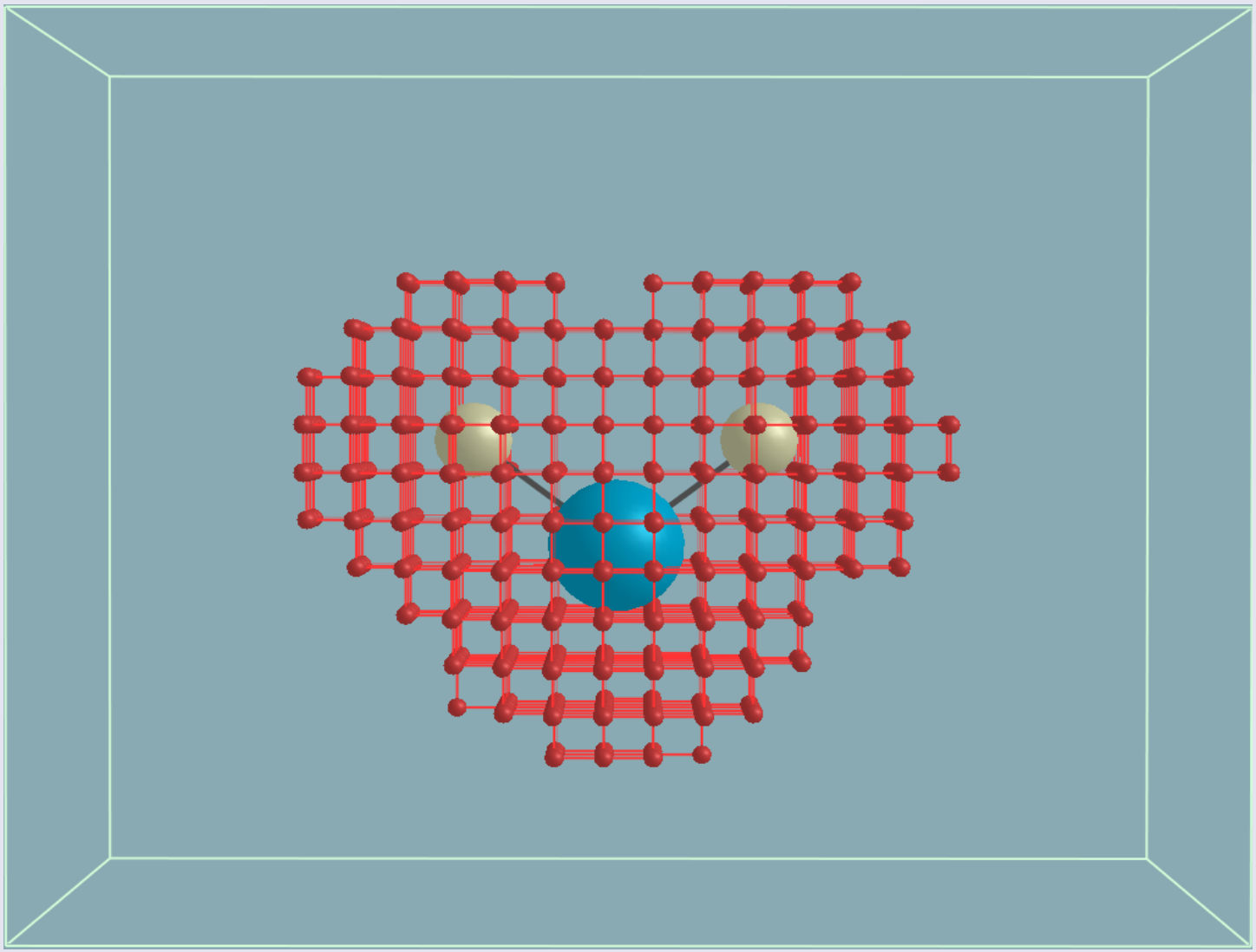
Fine grid (high resolution)



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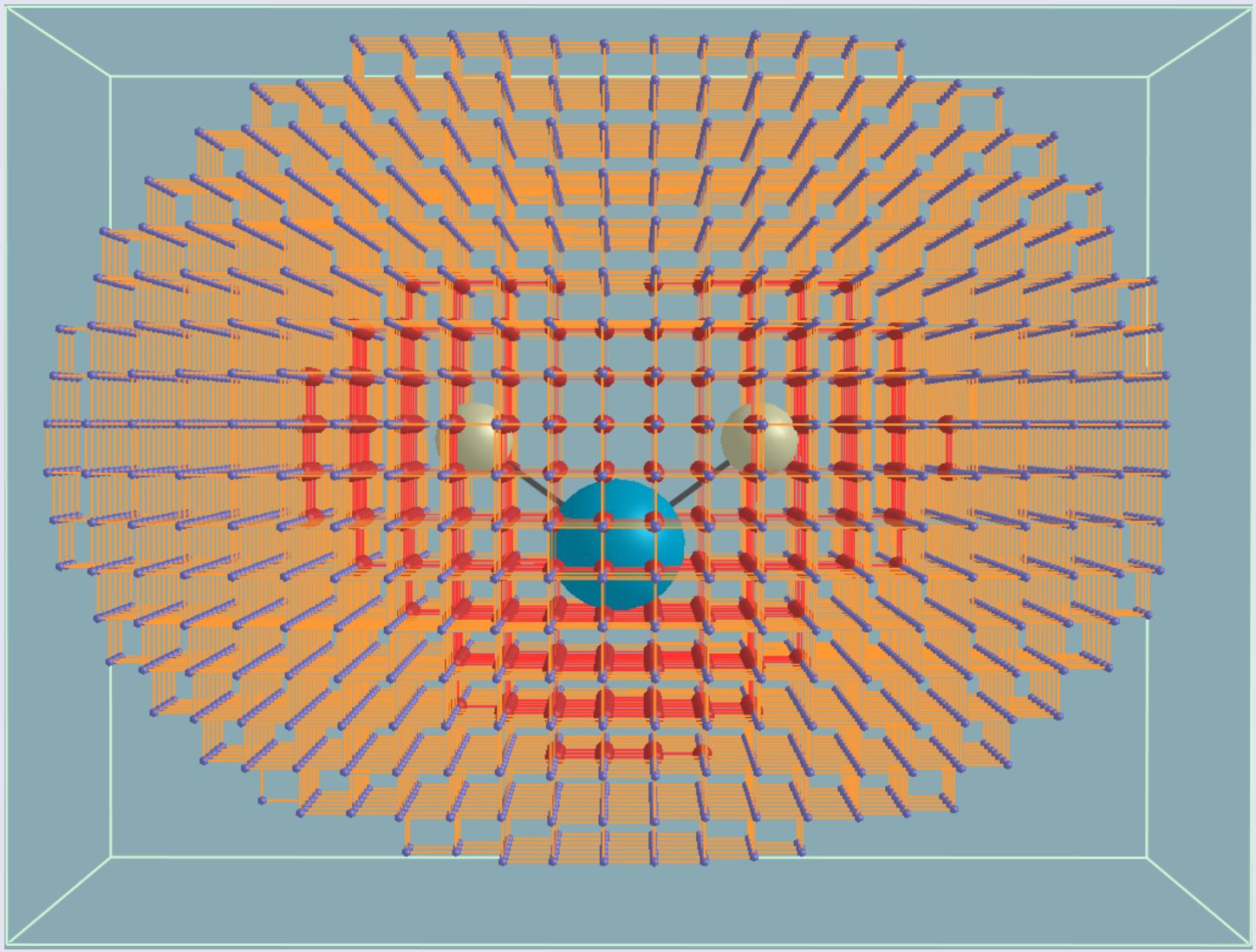
Coarse grid (low resolution)



Adaptivity of the mesh

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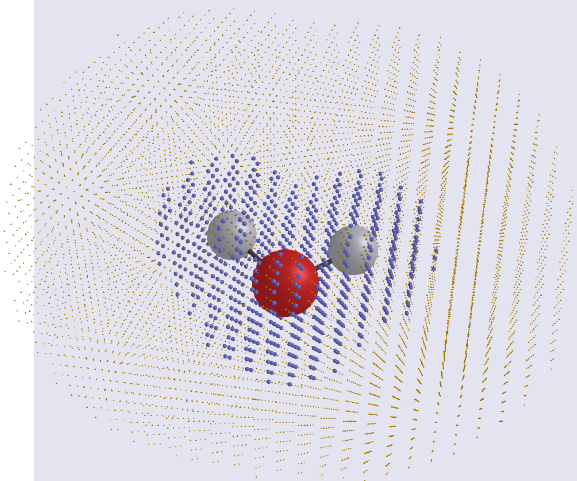
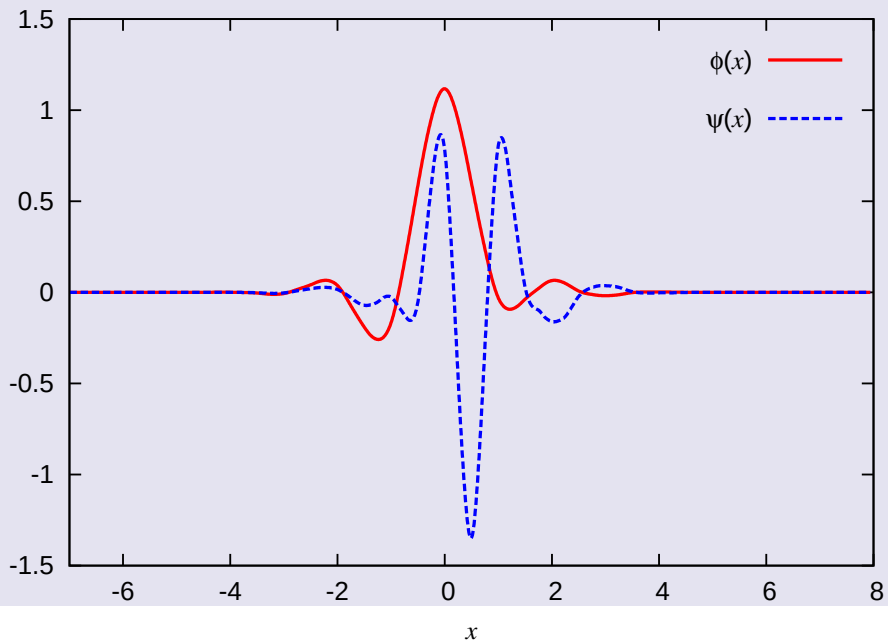
No close form: Scaling relations!

All functions have compact support, centered on grid points.

$$\phi(x) = \sum_{j=-m}^{m} h_j \phi(2x - j)$$

We only use the filters h_j: short convolutions (GPU-friendly)





DeltaTest benchmark: $\Delta = 1.0$

Science, 351, 6280 (2016)

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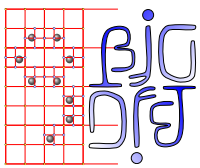
For the first three rows

Code	Version	Basis	Electron treatment	Δ-value	Authors
WIEN2k	13.1	LAPW/APW+lo	all-electron	0 meV/atom	S. Cottenier
BigDFT	1.7.6	Daubechies wavelets	HGHk-semicore and NLCC 2015 norm-conserving	0.1 meV/atom	BigDFT [11]
Elk	3.1.5	APW+lo	all-electron	0.2 meV/atom	Elk [14]
VASP	5.2.12	plane waves	PAW 2015 GW-ready (5.4)	0.2 meV/atom	K. Lejaeghere
Quantum ESPRESSO	5.1	plane waves	SSSP Accuracy (mixed NC/US/PAW potential library)	0.2 meV/atom	QuantumESPRESSO [12]
FLEUR	0.26	LAPW (+lo)	all-electron	0.2 meV/atom	FLEUR [9]
FHI-aims	081213	tier2 numerical orbitals	all-electron (relativistic atomic_zora scalar)	0.3 meV/atom	ASE [2]

Screenshot of DeltaTest webpage as of 24/02/16, elements up to Ar, new NLCC - HGH - NC - PSP (S. Saha)



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A code both for solid-state and physical chemistry

- 3D periodic, surfaces and free BC (← Poisson Solver)
- Usage of analytic HGH pseudopotentials
- Very high precision (analytic Kohn-Sham operators)
- All-electron accuracy, benchs in G2-1, (DeltaTest)

Present functionalities

Kohn-Sham DFT (metals, van der Walls, Hybrid Functionals), Systems embedded in electrostatic environments, Library of Structural Prediction, $O(N)$ calculations

Under implementation

Non orthorhombic cells, PAW, linear response TD-DFT

BigDFT breakdown process (1.8.x)

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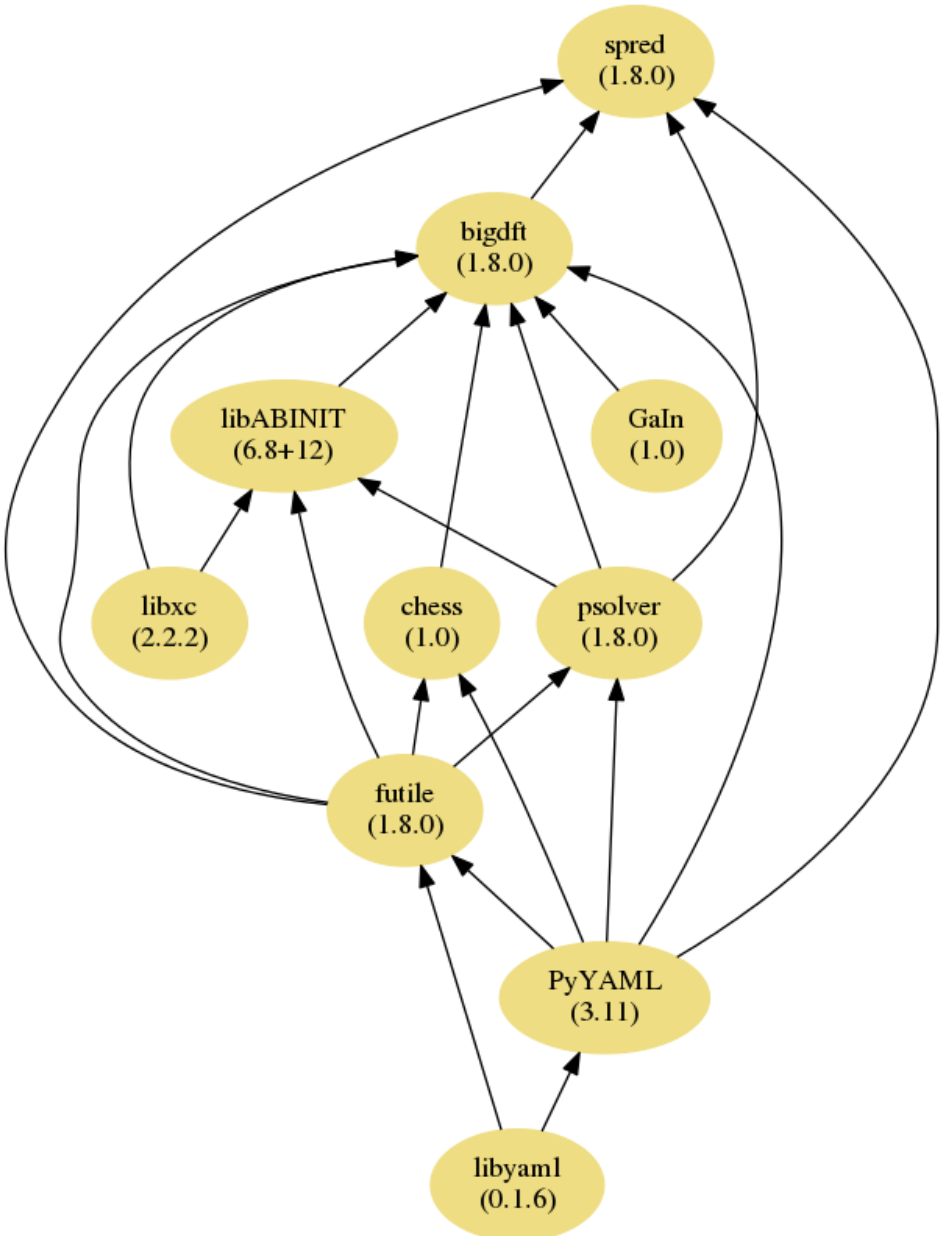

Modularity first

Each section of BigDFT is, when appropriate, defined as a module with its own build system and compilation instructions.

At present:

- FUTILE 1.0 (low level)
- GaIn 1.0
  (Gaussian Integral from Fiesta GW code)
- CheSS 1.0 (talk Stephan)
- PSolver 1.8
  Poisson solver + exchange

Hybrid Functionals for large systems γ=PBE0/PBE

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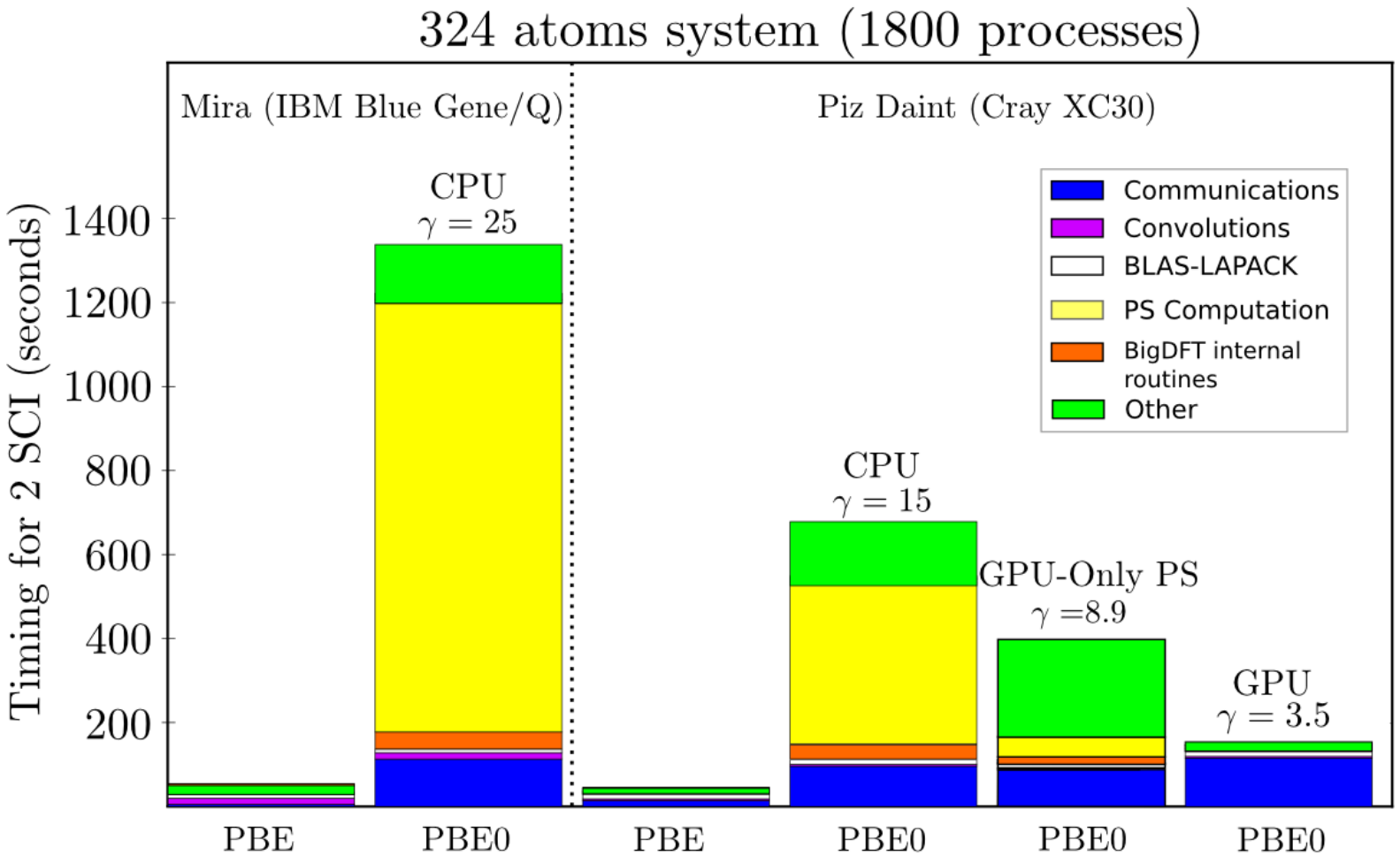
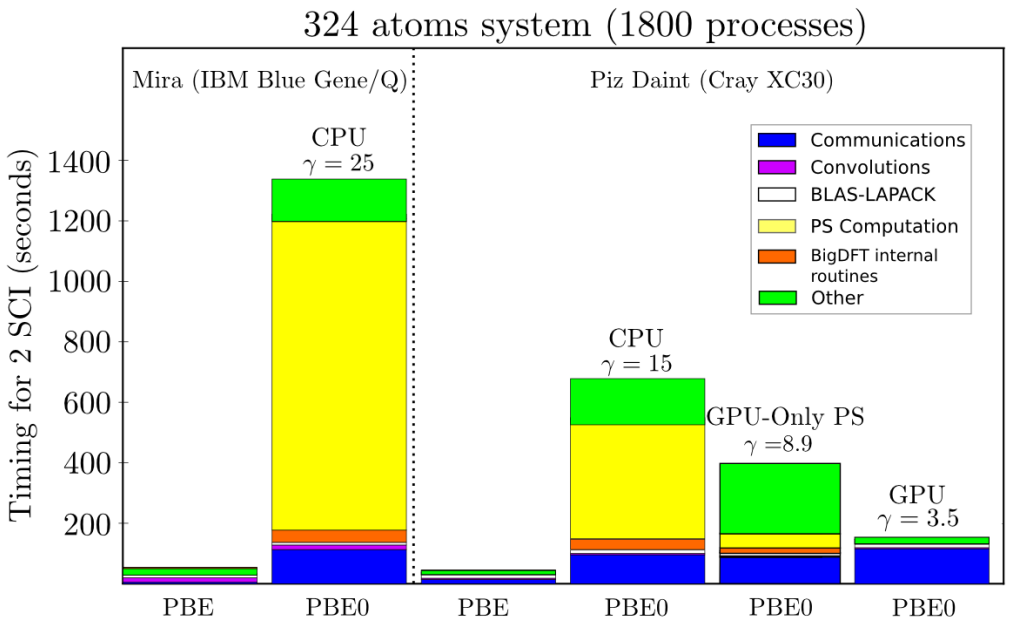
Since 2009, BigDFT ported on GPU.

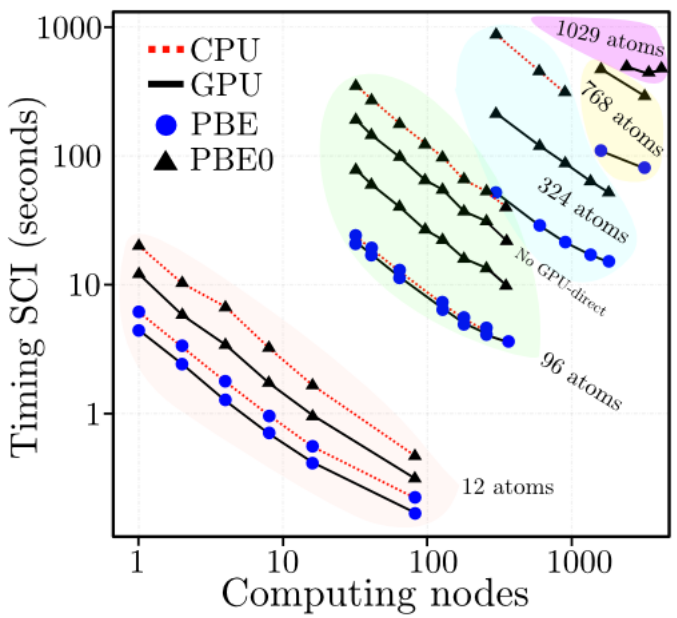


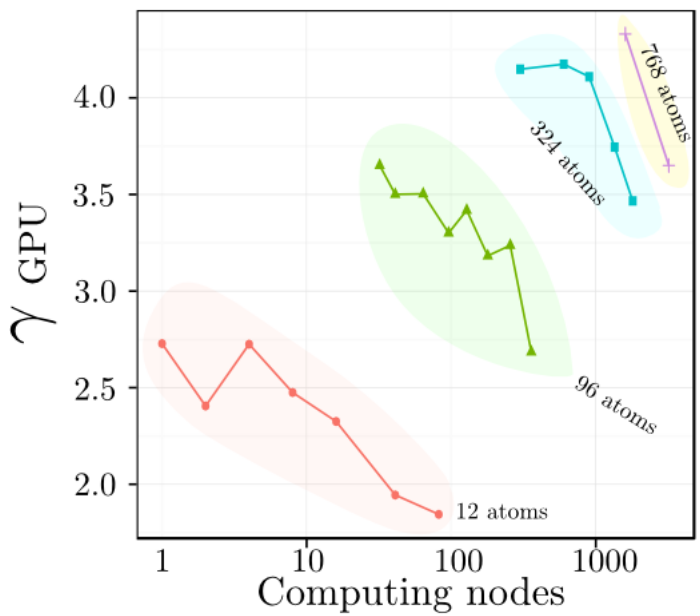
Use for the exchange part (N^2 Poisson Solver evaluations).

Hybrid Functionals for large systems γ=PBE0/PBE



UO_2 systems:	
Atoms	Orbitals
12	200
96	1432
324	5400
768	12800
1029	17150





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Scaling of "Traditional" BigDFT (linear scaling)

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"Traditional" BigDFT code

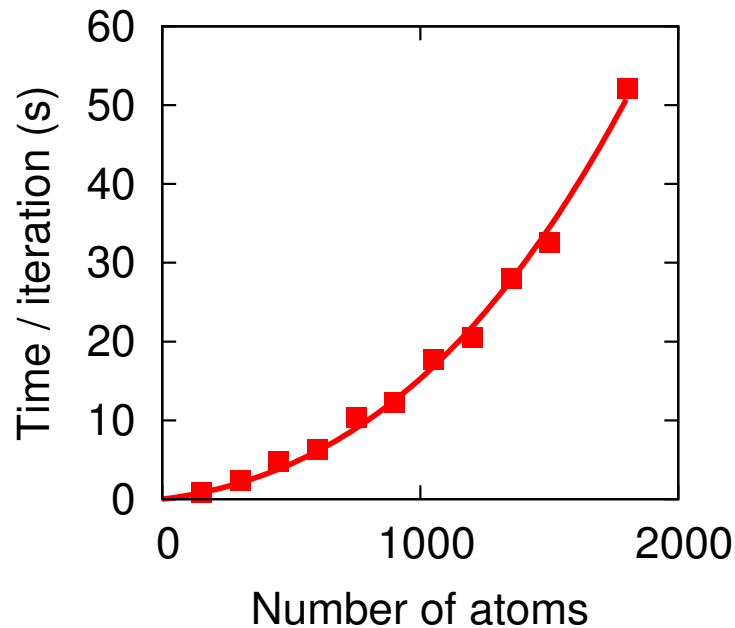
We can reach systems containing up to a few thousand electrons thanks to wavelet properties and efficient parallelization: (MPI + OpenMP + GPU)

DFT operations scale differently:

- $O(N \log N)$: Poisson solver
- $O(N^2)$: convolutions
- $O(N^3)$: linear algebra

and have different prefactors:

$c_{O(N^3)} \ll c_{O(N^2)} \ll c_{O(N \log N)}$



For bigger systems the $O(N^3)$ will dominate

☛ Motivation for a new approach

Localized optimized minimal basis set (linear scaling)

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Kohn-Sham orbitals

Linear combinations of support functions $\phi_\alpha(\mathbf{r})$:

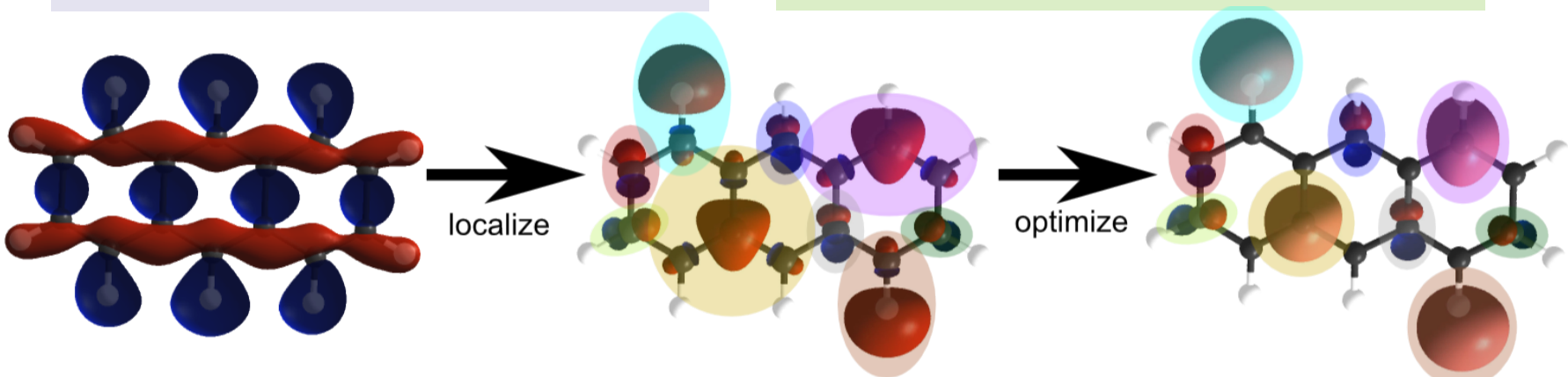
$$\Psi_i(\mathbf{r}) = \sum_\alpha c_i^\alpha \phi_\alpha(\mathbf{r})$$

- localized around atoms
- expanded in wavelets
- optimized in-situ

Density Matrix

Defined via the kernel $K^{\alpha\beta}$ in the $\phi_\alpha(\mathbf{r})$ basis:

$$\rho(\mathbf{r},\mathbf{r}') = \sum_i f_i \Psi_i(\mathbf{r})\Psi_i(\mathbf{r}') = \sum_{\alpha,\beta} \phi_\alpha(\mathbf{r}) K^{\alpha\beta} \phi_\beta(\mathbf{r}')$$



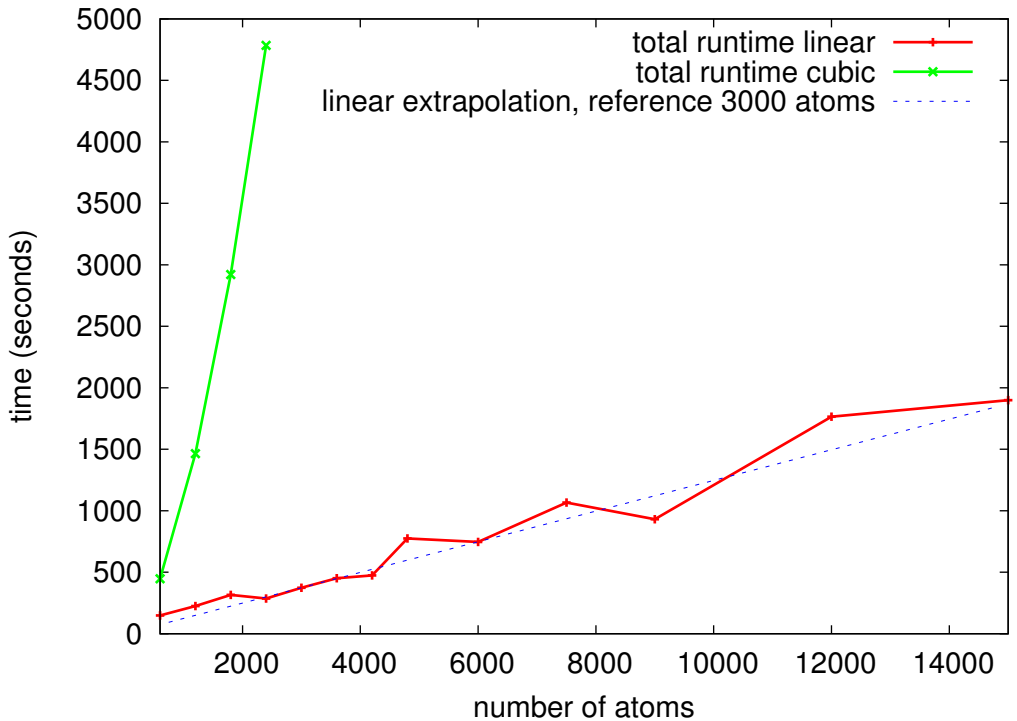
extended Kohn-Sham orbitals
cubic scaling, high accuracy

localized support functions (LCAO)
linear scaling, low accuracy

localized adaptive support function
linear scaling, high accuracy

Localization → Sparse matrices ($H^{\alpha\beta}$, $K^{\alpha\beta}$) → $O(N)$

Comparison with the cubic version (linear scaling)



- 20 min for 18 000 atoms
- CPU Time and memory $\propto$ number of atoms

High flexibility, like the cubic code

- Different levels of precision via the cutoff radii: Without fine-tuning converges to absolute energy differences of the order of 10 meV/atom, and almost exact forces.
- System sizes: 100 - 200K atoms $\rightsquigarrow$ 1M basis functions

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$O(N)$ vers.

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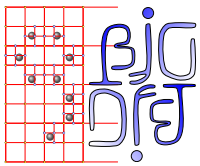
Outlook

Features of the localized optimized minimal basis set



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Ideal properties to work at the many thousand atoms scale

- Accurate results with good localization
- Low No. of degrees of freedom

☛ Low condition number (quasi-orthogonal)

☛ Small Spectral Width (thanks to pseudo-potential)

		S		H	
system	(#atoms)	sparsity	κ	sparsity	SW (eV)
DNA	(15613)	99.57%	2.29	98.46%	49.25
bulk pentacene	(6876)	98.96%	2.26	97.11%	42.30
perovskite	(768)	90.34%	2.15	76.47%	47.25
Si nanowire	(706)	93.24%	2.16	81.61%	41.54
H_2O droplet	(1800)	96.71%	1.57	90.06%	38.26

Our record: 250,000 atoms (Stephan Mohr)

Algorithm is robust and reliable on a variety of systems

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Outlook

Accurate and efficient linear scaling DFT calculations with universal applicability

S. Mohr, L. E. Ratcliff, L. Genovese, D. Caliste, P. Boulanger, S. Goedecker and T. Deutsch

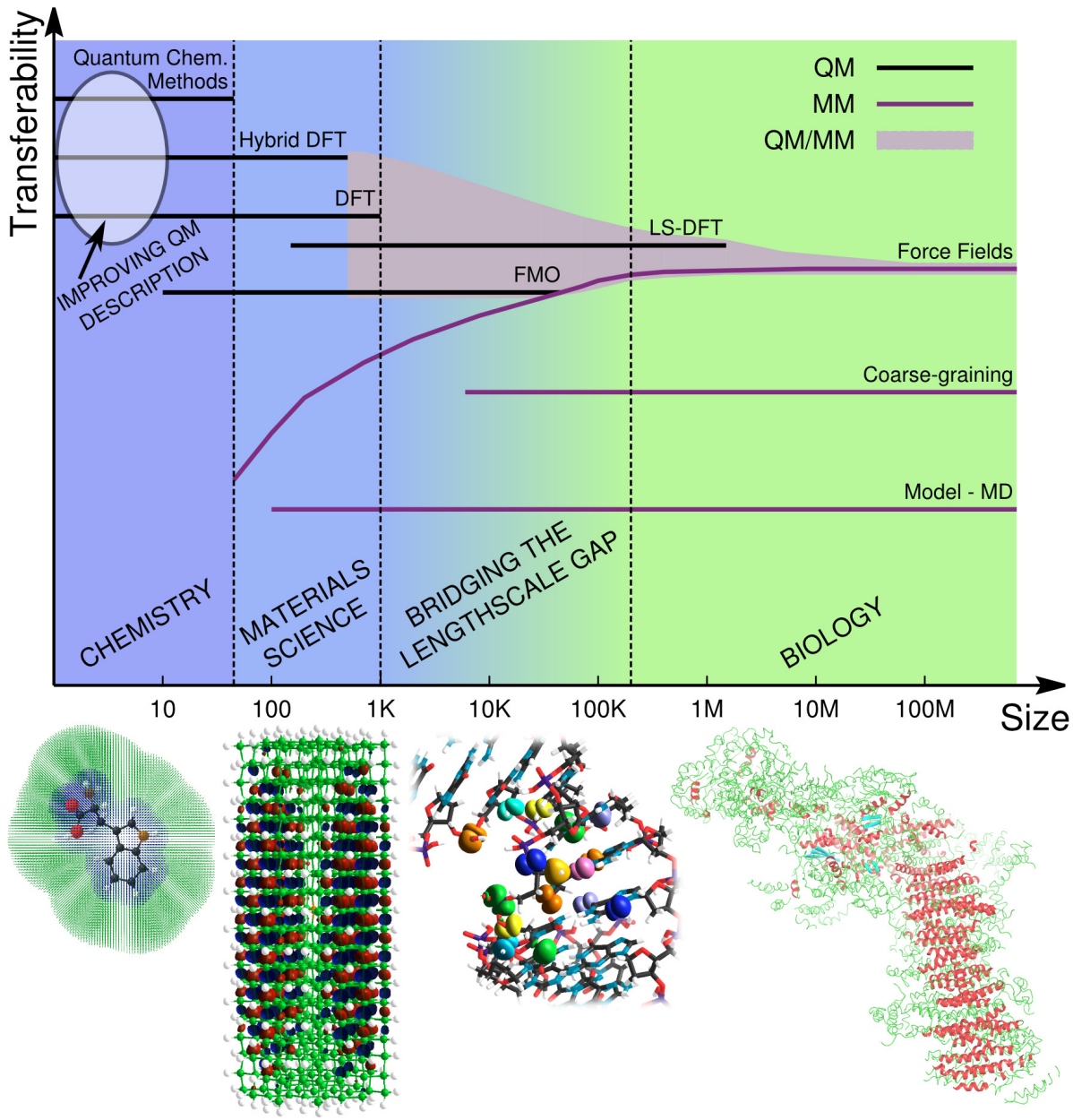
Phys. Chem. Chem. Phys., 2015, **17**, 47, 31360-31370.

DOI: 10.1039/c5cp00437c

Included in the **Real-space numerical grid methods in quantum chemistry** themed issue of *PCCP*

Guest-edited by Luca Frediani (The Arctic University of Norway) and Dage Sundholm (University of Helsinki)

Why do we need so Large scale DFT?



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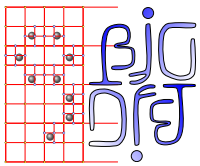
Review of $O(N)$ DFT calculations



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Advanced Review

Challenges in large scale quantum mechanical calculations

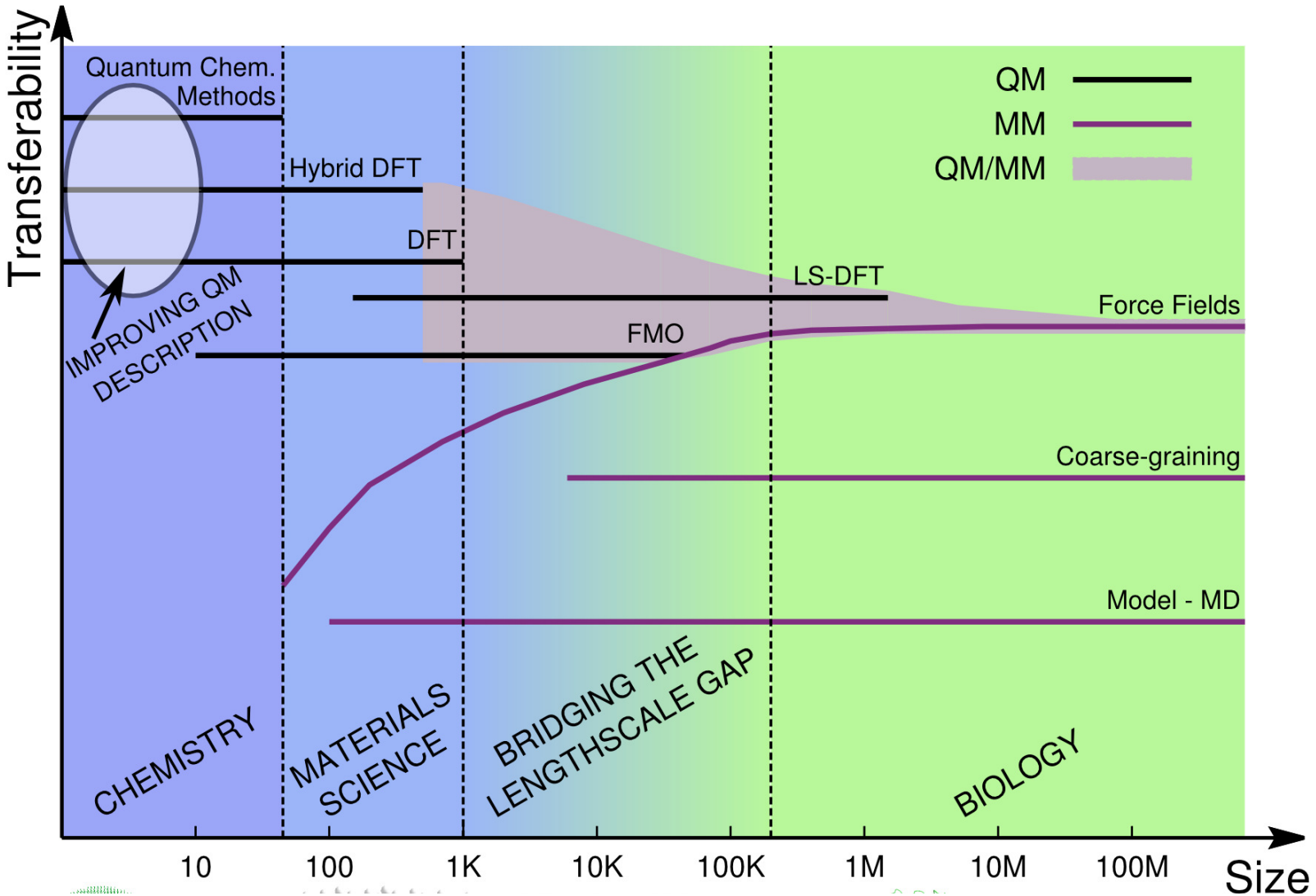
Laura E. Ratcliff,[1] Stephan Mohr,[2] Georg Huhs,[2] Thierry Deutsch,[3,4] Michel Masella[5] and Luigi Genovese[3,4]*

During the past decades, quantum mechanical methods have undergone an amazing transition from pioneering investigations of experts into a wide range of practical applications, made by a vast community of researchers. First principles calculations of systems containing up to a few hundred atoms have become a standard in many branches of science. The sizes of the systems which can be simulated have increased even further during recent years, and quantum-mechanical calculations of systems up to many thousands of atoms are nowadays possible. This opens up new appealing possibilities, in particular for interdisciplinary work, bridging together communities of different needs and sensibilities. In this review we will present the current status of this topic, and will also give an outlook on the vast multitude of applications, challenges, and opportunities stimulated by electronic structure calculations, making this field an important working tool and bringing together researchers of many different domains.

How to cite this article:
WIREs Comput Mol Sci 2016. doi: 10.1002/wcms.1290

New calculation paradigms are emerging!

Bridging the gap between different methods!



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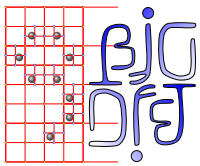
Outlook

Testing different approaches and models



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Outlook

- Fragments : Constrained DFT, Charge transfer, Excitations
- Atomic charge analysis
- Statistics of atomic configurations (snapshots from MD with force fields)
- Impact of the (electrostatic) environment
- Comparison between Full QM, QM/QM, and MM calculations

Dividing a system into fragments

JCP **142**, 234105 (2015)

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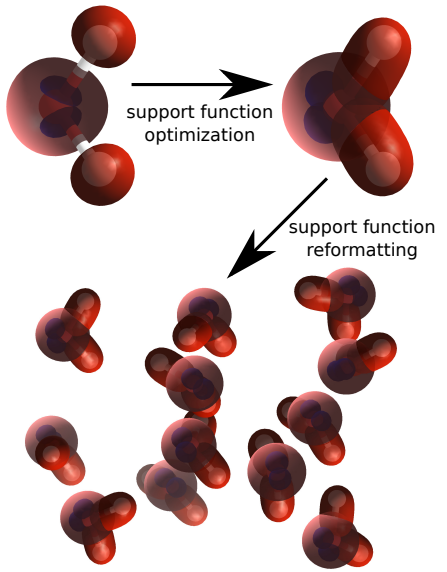
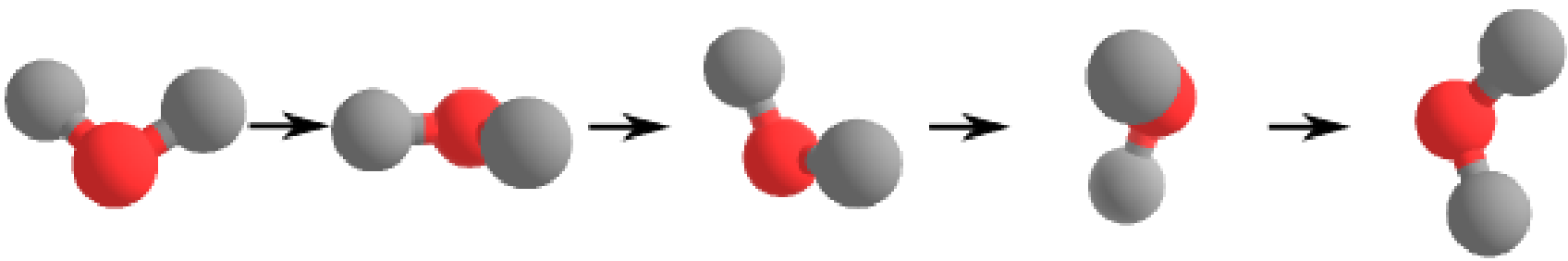
Outlook

We can duplicate the localized adapted orbitals for similar portions of large systems $\rightarrow$ considerably reduces the cost



☛ Enables manipulation of optimized basis sets (need an efficient reformatting)

✔ Efficient and precise roto-translation of localized orbitals



Reformatting the minimal basis set in the same grid

Fragment: Charge transport in OLEDs (statistics)

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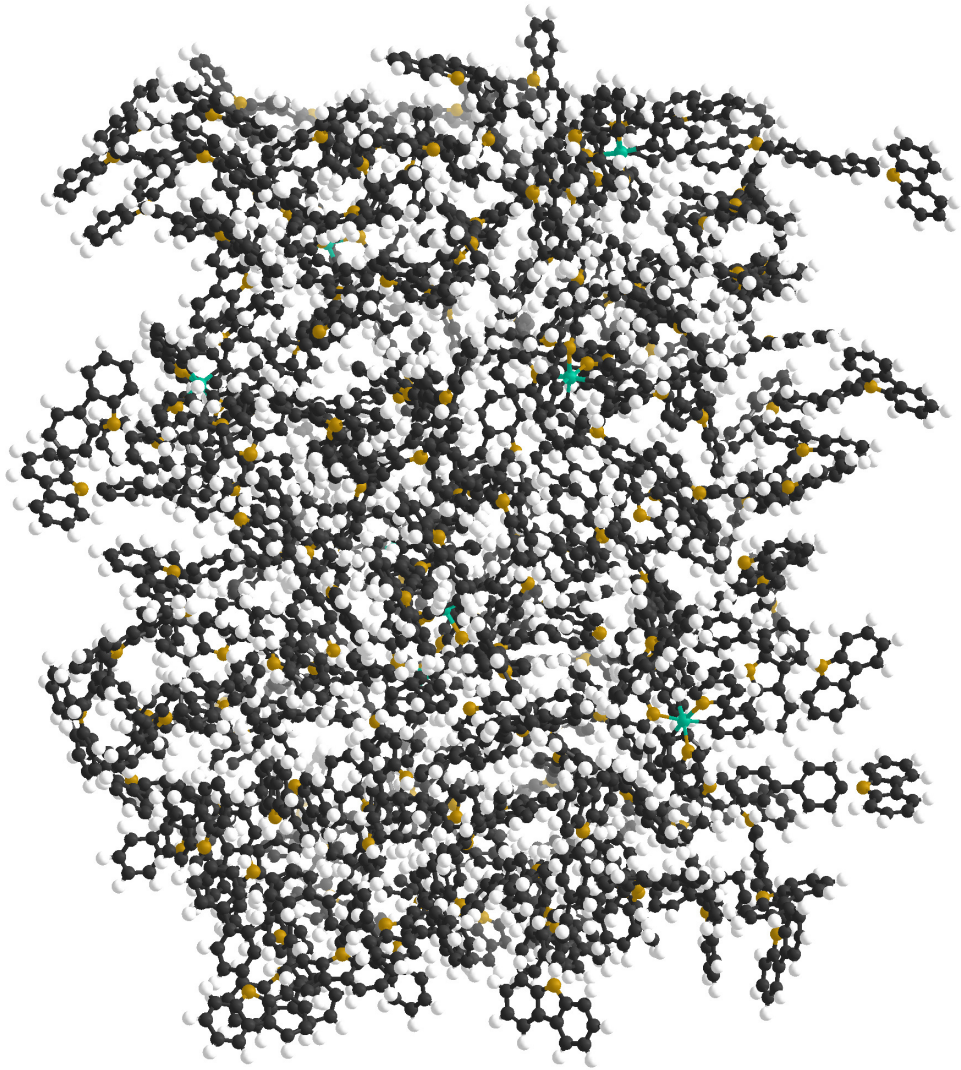
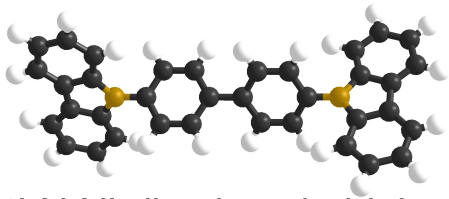
Impact of environment in a realistic 'host-guest' morphology:
6192 at., 100 molecules — L. Ratcliff et al. JCP 142, 234105 (2015)

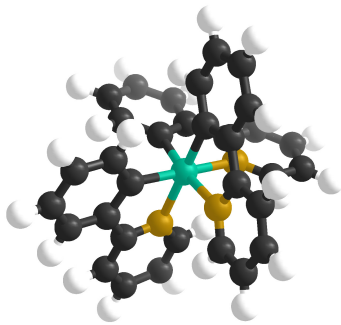


Host molecule



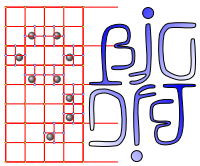
4,4'-N,N'-dicarbazole-biphenyl

Guest molecule



tris(2-phenylpyridine)iridium

Complex Input files: What we need?



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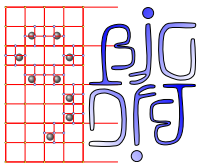
Outlook

Need to build complex files and process output files

- Use of a Human readable markup language (YAML)
- An output file can be an input file to rerun a simulation
- Easy to parse and process
  - as Python dictionaries
  - Develop the Fortran FUTILE library
- Towards workflows (Jupyter notebooks)



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Block structured YAML

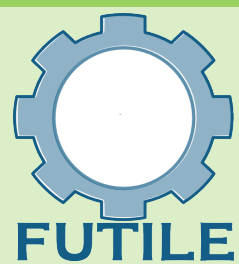
- A clean and very human readable format.
- Very good choice for configuration files that are human readable and editable while at the same time interpretable and modifiable by a program.

☛ YAML parsers are available for scripting languages (Python, Ruby)

"FUTILE" approach to input variables and options

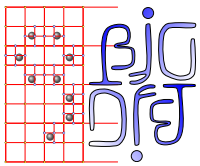
- Options defined like an input file

  (`key` → `value`)



- Logfiles come structured similarly
- Usage of structured I/O at FORTRAN level
- Allows for legacy code and enhanced modularity

Example of the input dictionary (YAML format)



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Cubic vers.

$O(N)$ vers.

Bridging the gap

Facilitating processing

Outlook

Use for Input and Output files!

```
setup:
  taskgroup_size :  0 # Size of the taskgroups
  accel :  none # Acceleration
  global_data :  No
  verbose :  Yes #Verbosity switch
  output :  none #Quantities to be plotted
kernel:
  screening :  0 #Mu screening parameter
  isf_order :  16 #Order of the ISF family
  stress_tensor :  Yes #Extract stress tensor
environment:
  cavity :  none #Type of the cavity
  cavitation :  No # Extra cavitation terms
  input_guess :  Yes #Input guess procedure
  fd_order :  16 #Order of FD derivatives
  itermax :  50 #Max.  No.of GPS iterations
  minres :  1.e-6 #Convergence threshold
  pb_method :  none #Poisson Boltzmann Equation
```

How can we benefit from this?

- Understanding code behaviour on a new architecture
- Identify optimization strategies *for the end-user* (modification of the input file)
- Small demo, notebooks based on jupyter

Perspectives

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Outlook

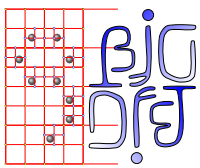
Linear-Scaling DFT: Opens up new possibilities

- Robust convergence, high accuracy and flexibility (BC)
- Reduction in degrees of freedom $\rightarrow$ large systems via moderate sized machines ($\sim$ TFlop/s) Lab-scale
- Different level of descriptions (controlling the precision) QM $\supset$ Fragments $\supset$ Atomic charges

Challenges and future directions

- Explore interplay environment $\leftrightarrow$ electronic excitations (CDFT, QM/MM, statistics, . . .)
- Provide high quality back end for extraction of atomic multipoles from QM calculations
- Explore Linear Response Time-Dependent DFT, . . .

Acknowledgments



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Outlook

- Main maintainer — Luigi Genovese
- Group of Stefan Goedecker — Basel University
  B. Schaefer, A. Ghazemi, S. Saha, G. Fisicaros
- Order N methods (fragments, constrained DFT)
  Stephan Mohr (BSC), L. Ratcliff (Imperial College), Paul Boulanger (U. Montreal)
- Link with ABINIT and python bindings
  Damien Caliste (CEA)
- Hybrid Functionals, GPU — A. Degomme (CEA)
- Optimized convolutions — B. Videau, J.-F. Méhaut (LIG, computer scientists, Grenoble)