DFTK: A Julian approach for simulating electrons in solids

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29th July 2020

MATHerials

Inria

École des Ponts ParisTech

→ `https://michael-herbst.com/talks/2020.07.29_juliacon_dftk.pdf`

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Why care about electrons?

- Electrons glue the world together
- Electrons keep the world apart

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Materials and semiconductors

CC-by-3.0 https://en.wikipedia.org/wiki/File:Silicon.jpg

Chemical and pharmaceutical industry

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Aren't experiments good enough?

- Experiments are expensive (money, people, time)
- 1 droplet water[1]: $1.7 \cdot 10^{21}$ particles
- Experiments only measure averages
- Sometimes hard to link to physical laws

$\Rightarrow$ Cooperative research of experiment and theory

$\Rightarrow$ Standard practice in industry and research

[1] Assume $0.05\,\mathrm{ml}$.

Electronic structure theory

- Goal: Electronic properties (conductivity, photoactivity, . . .)
  - Construct physically sound models
  - Validate / help to interpret experiment
- Sketching the approach:
  - Regime of quantum mechanics
  - System: Hamiltonian $\hat{\mathcal{H}}$, differential operator
  - Minimisation problem: Ground state Ψ with energy

$$E = \min_{\Psi} \int_{\mathbb{R}^{3N}} \Psi\left(\underline{\boldsymbol{r}}_1, \underline{\boldsymbol{r}}_2, \ldots, \underline{\boldsymbol{r}}_N\right) \hat{\mathcal{H}} \Psi\left(\underline{\boldsymbol{r}}_1, \underline{\boldsymbol{r}}_2, \ldots, \underline{\boldsymbol{r}}_N\right) \mathrm{d}\underline{\boldsymbol{r}}_1 \cdots \mathrm{d}\underline{\boldsymbol{r}}_N$$

  - Properties: Derivatives of the energy

Density-functional theory

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- Challenge: Size of N
- 2 Silicon atoms: $N = 28 \Rightarrow \mathbf{2}^{84} \approx 2 \cdot 10^{25}$ quadrature points

$\Rightarrow$ Finished in 1 year:

$\Rightarrow$ Density-functional theory (DFT) approximation

- Effective one-particle model ($N = 1$)
- May construct DFT model for specific context
- Discretisation basis: Build known physics into model
- But: Non-convex, non-linear minimisation

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Self-consistent field procedure

- Euler-Lagrange equations (DFT):

$$\begin{cases} \hat{\mathcal{F}}_\rho = -\frac{1}{2}\Delta + V_\rho \\ \rho(\underline{\boldsymbol{r}}) = \sum_i f_{\varepsilon_F}(\varepsilon_i)\,|\psi_i(\underline{\boldsymbol{r}})|^2 \quad \text{with } \hat{\mathcal{F}}_\rho \psi_i = \varepsilon_i \psi_i, \\ \qquad \varepsilon_F \text{ chosen such that } \int \rho \, \mathrm{d}\underline{\boldsymbol{r}} = N, \\ \qquad \text{and } f_{\varepsilon_F}(x) = \left[1 + \exp\left(\frac{x-\varepsilon_F}{T}\right)\right]^{-1} \end{cases}$$

- Self-consistent field procedure (SCF):
  (1) Guess initial density ρ
  (2) Build Kohn-Sham operator $\hat{\mathcal{F}}_\rho$
  (3) Diagonalise it to get new $\{\psi_i\}_i$
  (4) Build new ρ go to (2).

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Landscape of DFT codes

- `https://www.vasp.at`
- `https://www.abinit.org`
- `http://www.castep.org`
- `https://wiki.fysik.dtu.dk/gpaw`
- `https://www.quantum-espresso.org`
- `https://crd-legacy.lbl.gov/~chao/KSSOLV`
- ...

- Represents hundreds of man-years of coding!

$\Rightarrow$ Why bother with **DFTK**?

Interdisciplinary research field

- **Mathematicians:** Toy models and unphysical edge cases
- **Scientist:** Wants to focus on science, not numerics
- **High-performance person:** Exploit hardware specialities
- **Practitioner:** Reliable, black-box, high-level interface

- Typical obstacles:
  - Difficult problem ⇒ Often complex codes
  - Hard-coded: Workflow / algorithms / hardware optimisations
  - Huge code bases (1M lines and beyond)
  - Non-standard input syntax and API
  - Unusual systems frequently require hand-tuning
  - Two-language problem: Where to cut?

DFTK — https://dftk.org

- 14 months of development, ≈ 5000 lines
- julia code (modulo required Python and C libraries)
- Sizeable feature list:
  - Multi-level threading
  - 1D / 2D / 3D systems
  - Compose your own model
  - Integration with materials-related Python modules
  - > 500 electrons
- Performance: Within factor 2ish of established codes
- Platform for multidisciplinary collaboration
- Documentation and examples: `https://docs.dftk.org`

14 months and 5000 lines

- How did we manage . . .
- . . . well, there's the awesome julia community & ecosystem

14 months and 5000 lines

- How did we manage . . .
- . . . most we needed was there:
  - Fourier transforms (`FFTW`)
  - Linear solvers (`IterativeSolvers`)
  - Non-linear solvers (`Optim`, `NLsolve`)
  - High-level data (`PeriodicTable`, `Primes`)
  - Building blocks (`Roots`, `LineSearches`, `LinearMaps`, . . .)
  - Interfacing (`PyCall`)

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  - Interfacing (`PyCall`)
- Thank you!

Contents

Lowering the entrance barrier for researchers

- Money: Always tight
- Time: 3-ish years for a PhD, master even less
- State of the art:
  - *Some* codes require software licences $\mathcal{O}$(5k€)
  - Usage: Input format and interface
  - Development: Scarce tests, comments, documentation
  - 1M lines of hardly uniform code conventions
  - Original developers have left (PhD is over)

Attempts to lower the barriers in DFTK

- julia: Zero cost and great learning resources
- Design goal: Code follows mathematical structure of DFT
- Aim for best agreement between code and equations (Unicode)
- Comments: Hint derivation or point to original articles
- `https://docs.dftk.org` with plenty of usage example
- Example projects:
  - Publication following master project
  - 8-week student project to toy with GPUs in DFT

  $\Rightarrow$ Both cases: No familiarity with DFT or julia
  - Error estimates: 10 weeks to publication

High-throughput screening

- *In silico* design of novel materials
- Challenging classifiers: Band gap, excitation energies, ...
- Narrow down 10k candidates to $\mathcal{O}(10)$
- Requirements:
  - Tunable parameters: Accuracy *versus* speed
  - Ideally: Target accuracy drives black-box workflow
  - Reliability: Breakdown of SCF not acceptable
  - Scriptability and interfacing to data science tools
  - Exploit whatever hardware exists (GPU, accelerators) ...

Screening and state of the art

- State of the art:
  - Plenty accuracy-related parameters
  - Chosen by experience: Too tight *versus* too optimistic
  - API decided *a priori* (two-language problem)

  $\Rightarrow$ Decisions hard-coded (e.g. floating-point type)

  $\Rightarrow$ Little freedom to tweak algorithms

  $\Rightarrow$ *Basically* hard fork for every accelerator / architecture

$\Rightarrow$ One long-term driving force behind DFTK

A posteriori error analysis in DFT

- *A posteriori* error: Upper bound how far solution is off
- Mathematical answer for: Has target accuracy been reached?
- Sources of error in DFT:
  - Model error
  - Discretisation error
  - Algorithm error
  - Arithmetic error
- *A posterior* bound on error $\Rightarrow$ Automatic error balancing
- For DFT: Full picture not yet understood

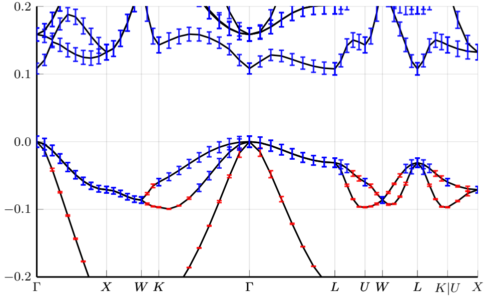
A posteriori error analysis with DFTK

- Requirements:
  - Mathematical theory can only treat reduced models
  - Step-by-step expansion
  - Ingredients not yet clear (e.g. form of integrals, derivatives)

  $\Rightarrow$ Need accessible toolbox for experimentation
  - Arithmetic error: Interval arithmetic, elevated precision
- DFTK offers:
  - Fully customisable model
  - Support for arbitrary floating-point types
  - Use julia ecosystem on DFTK datastructures:
    - Numerical quadrature, forward-mode AD, ...

    $\Rightarrow$ Rapid prototyping in numerical linear algebra

A posteriori error analysis: First results[1]



- Reduced model: Non-self-consistent Kohn-Sham
- Estimation of arithmetic error (`IntervalArithmetic.jl`)
- Used elevated floating-point type (`DoubleFloats.jl`)

- Time to submission: 10 weeks

[1]M. F. Herbst, A. Levitt and E. Cancès. Faraday Discuss. *In press.* (2020)

Reliable SCF algorithms

- Convergence of SCF depends on dielectric properties

$\Rightarrow$ Different SCF needed for metals, insulators, semiconductors

- Current schemes based on results for bulk materials

$\Rightarrow$ What to do e.g. for surfaces?

- Requirements:
  - 1D / 2D / 3D: Analyse spectral properties
  - Rapid prototyping to mix and match ideas

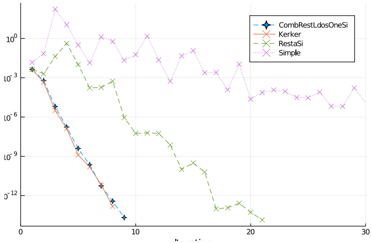
  $\Rightarrow$ High-level code inside key algorithms

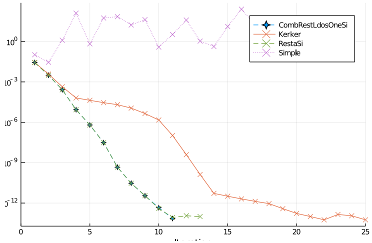
  - Testing requires realistic systems, but first version never works

  $\Rightarrow$ Scalup should not imply a rewrite of toy code

Proposing SCF algorithms with DFTK (WIP)

- Initial developments on 1D and 2D systems
- Compute spectral properties with `KrylovKit.jl`
- Tested identical implementation on systems > 500 electron
- Currently 4th refinement iteration:



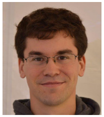


- Aluminium (10 repeats)
- Silicon (10 repeats)

Summary and outlook

- DFTK: Interdisciplinary software development
  - Closely integrated with julia ecosystem
  - Mix and match to build new algorithms
  - Rapid prototyping and toy problems
  - Scale-up for realistic testing and applications
- Near-future steps
  - SCF schemes for challenging systems (e.g. spin)
  - Error estimates and black-box workflows for DFT
  - Mixed precision and multigrid methods
  - Methods beyond DFT and SCF
  - Full GPU integration

Acknowledgements


Antoine Levitt

Eric Cancès

Other DFTK contributors:

- `@gkemlin`, `@ssirajdine`, `@louisponet`

julia & package developers

- `DoubleFloats.jl`
- `IntervalArithmetic.jl`
- `NLsolve.jl`
- `Optim.jl`
- `Plots.jl`
- `PyCall.jl`
- `Roots.jl`
- `StaticArrays.jl`
- `$THOSE_I_FORGOT`

Questions?

DFTK `https://dftk.org`

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`https://michael-herbst.com/blog`

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