Automatic differentiation efforts in DFTK

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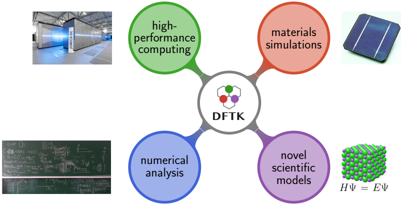
Slides: `https://michael-herbst.com/talks/2021.10.07_dftk_ad_update.pdf`



Contents

Modelling electronic structures

- Seek variational energy: $\min_P E(P)$
- But: Experiments can't measure energies!

- Changes in the energy are what is interesting
- Key question: How is the response to external perturbation?
- Examples:
  - Forces (response to atomic position shifts)
  - Dipole moment (response to electric field)
  - Elasticity (cross-response to lattice deformation)
  - ...
- Often directly measurable (or closely linked)

$\Rightarrow$ We care very much about derivatives

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Need for automatic derivatives: Practical argument

- Many (many) models
- Need derivatives to judge usefulness of method
- Deriving / implementing analytic derivatives takes time ...
- ...so does fixing the bugs

- Even standard codes don't have all relevant derivatives
- Standard fallback: Finite differences

Need for AD: New and improved methods

- Any model building needs error control ...
- Error control needs derivatives (sensitivities)

- Data-driven model construction
- Scientific machine learning
- E.g. neural-network functionals / pseudos / ...

- Challenge: Requires *unusual* derivatives:
  - Density *vs.* XC parameters
  - Atomisation energy *vs.* pseudo parameters
  - ...

Property computation

- SCF fixed-point problem in density matrix P

$$0 = f(P, \lambda) = f_{\mathsf{FD}}\Big(H^\lambda(P)\Big) - P$$

  with
  - λ: Parameter of external perturbation
  - f_{FD}: Fermi-Dirac function
  - H^λ: Non-linear Kohn-Sham Hamiltonian
- Defines implicit function $P(\lambda)$ for density matrix
- Quantities of interest:

$$\frac{dQ(P)}{d\lambda} = \frac{\partial Q}{\partial \lambda} + \frac{\partial Q}{\partial P}\frac{\partial P}{\partial \lambda}$$

  - Forces: $Q = E, \quad \lambda = R$ (atomic displacement)
  - Polarisability: $Q =$ dipole moment, $\quad \lambda = \mathcal{E}$ (electric field)

Hellmann-Feynman theorem

$$\frac{dQ(P)}{d\lambda} = \frac{\partial Q}{\partial \lambda} + \frac{\partial Q}{\partial P}\frac{\partial P}{\partial \lambda}$$

- Special case of $Q = E$
- Recall $P_* = \text{argmin } E(P) \quad \Rightarrow \quad \left.\frac{\partial E}{\partial P}\right|_{P_*} = 0$
- Hellmann-Feynman theorem

$$\left.\frac{dE}{d\lambda}\right|_* = \left.\frac{\partial E}{\partial \lambda}\right|_*$$

- First energy derivatives are (comparatively) easy!

Response theory (1)

- If $Q \neq E$ we need $\frac{\partial P}{\partial \lambda}$
- Consider at $\lambda = \lambda_*$ and corresponding P_* and H_*:

$$
\begin{aligned}
0 &= \frac{\partial}{\partial \lambda} \left[f_{\text{FD}}\Big(H^\lambda(P)\Big) - P \right]\Big|_* \\
&= f'_{\text{FD}}(H_*) \cdot \frac{\partial H^\lambda}{\partial \lambda}\Big|_* + \frac{\partial P}{\partial \lambda}\Big|_* \cdot \frac{\partial}{\partial P} \left[f_{\text{FD}}\Big(H^\lambda(P)\Big) - P \right]\Big|_* \\
&= f'_{\text{FD}}(H_*) \cdot \frac{\partial H^\lambda}{\partial \lambda}\Big|_* + \frac{\partial P}{\partial \lambda}\Big|_* \cdot \left[f'_{\text{FD}}(H_*) \cdot \boldsymbol{K}^{\lambda_*}(P_*) - I \right]
\end{aligned}
$$

where $\boldsymbol{K}^{\lambda_*} = \dfrac{\partial H^{\lambda_*}}{\partial P}$

Response theory (2): Sternheimer equation

$$0 = f'_{\mathsf{FD}}(H_*) \cdot \left.\frac{\partial H^\lambda}{\partial \lambda}\right|_* + \left.\frac{\partial P}{\partial \lambda}\right|_* \cdot \left[f'_{\mathsf{FD}}(H_*) \cdot \boldsymbol{K}^{\lambda_*}(P_*) - I\right]$$

- Rearrange:

$$\left.\frac{\partial P}{\partial \lambda}\right|_* = -\left[f'_{\mathsf{FD}}(H_*)\boldsymbol{K}^{\lambda_*}(P_*) - I\right]^{-1} f'_{\mathsf{FD}}(H_*) \left.\frac{\partial H^\lambda}{\partial \lambda}\right|_*$$

$$= -\left[\boldsymbol{K}^{\lambda_*}(P_*) + \boldsymbol{\Omega}(H_*)\right]^{-1} \left.\frac{\partial H^\lambda}{\partial \lambda}\right|_*$$

where $\boldsymbol{\Omega}(H_*) = -\left(f'_{\mathsf{FD}}(H_*)\right)^{-1}$

- Sternheimer equation (implicit differentiation)

Example: Computing polarisabilities

- Homogeneous electric field $\lambda = \mathcal{E}$ along x-direction
- Cubic cell (length L_x)

- Hamiltonian $H^{\mathcal{E}}(P) = H_{\mathsf{DFT}}(P) - \mathcal{E}(x - L_x/2)$
- Perturbation $\frac{\partial H^{\mathcal{E}}}{\partial \mathcal{E}}\Big|_* = (x - L_x/2)$
- Dipole moment:

$$\mu(P) = \int_\Omega (x - L_x/2)\rho(r)\,\mathrm{d}r, \qquad \rho = \mathrm{diag}(P)$$

- Polarisability $\dfrac{d\mu}{d\mathcal{E}} = \dfrac{\partial \mu}{\partial P}\dfrac{\partial P}{\partial \mathcal{E}}$

1. Solve SCF $P_* = H^0(P_*)$ at zero field
2. Solve Sternheimer $\frac{\partial P}{\partial \mathcal{E}} = -[\boldsymbol{K} + \boldsymbol{\Omega}]^{-1} \frac{\partial H^{\mathcal{E}}}{\partial \mathcal{E}}$ *(implicit differentiation)*
3. Compute polarisability

Role of automatic differentiation

- Universal building blocks:
  - Primal pass: Solve SCF
  - f-rule: Solve Sternheimer

$\Rightarrow$ Code up once, use AD to take care of repetitive glue

- Adjoint-mode is goal:
  - Faster for larger number of parameters (neural net)
  - Support for higher derivatives
  - Sparsification techniques
- Adjoint-mode is feasible:
  - $\boldsymbol{K} + \boldsymbol{\Omega}$ is self-adjoint

  $\Rightarrow$ SCF r-rule: Adjoint-solve Sternheimer
- Let's look at things in practice ...

Contents

AD scenarios considered

- Forward-mode AD (ForwardDiff.jl)
  - Stresses via Hellmann-Feynman
  - Polarisability via implicit differentiation of SCF
- Adjoint-mode AD (Zygote.jl)
  - Stresses via Hellmann-Feynman
  - XC-functional gradients

Forward-mode AD with Hellman-Feynman

- For stresses $Q = E$, $\lambda = L$ (unit cell vectors)

$\Rightarrow$ Hellmann-Feynman applies

- Computing stresses:

$$\text{Stress} = \frac{1}{\det(\mathbf{L})} \left. \frac{\partial E\left[P_*, (I + \mathbf{M})\,\mathbf{L}\right]}{\partial \mathbf{M}} \right|_{\mathbf{M}=0}$$

- In julia code:

```
scfres = self_consistent_field(basis)  # Run SCF, get P*
     L = basis.model.lattice
stress = 1/det(L) * ForwardDiff.gradient(M -> recompute_energy(scfres, (I + M) * L),
                                         zero(L))
```

Stresses using ForwardDiff

ForwardDiff.jl workarounds & LIVE DEMO

Status of reverse-mode AD

ChainRules.jl workarounds & LIVE DEMO

AD scenarios considered

- Forward-mode AD (ForwardDiff.jl)
  - Stresses via Hellmann-Feynman **work**
  - Polarisability via implicit differentiation of SCF **works**
- Adjoint-mode AD (Zygote.jl)
  - Stresses via Hellmann-Feynman **work/WIP**
  - XC-functional gradients **WIP**

Strategies learned

- ForwardDiff.jl
  - ensure array-allocations can hold Dual numbers
  - custom overloads for non-Julia code (FFTW, spglib, ...)
- Zygote.jl with ChainRules.jl
  - avoid mutation
  - avoid indexing into large arrays
  - generating rrules from alternative primals
  - generating rrules from frules
  - general rrules for NLsolve.jl, IterativeSolvers.jl

Contents

Conclusion

- We got the building blocks for 1st derivatives
- The challenge now is gluing it all together
- TODO:
    - Hide the details ...
    - Minimise code duplication
    - Optimise performance
    - Fix the details (symmetries, external libraries ...)
    - Higher derivatives?
- Happy for any input!

What next?

- Sensitivities:
  - Structural, alchemical, model parameters
  - Band gaps, atomisation energies, forces, geo-opt
- Data-driven design:
  - DFT models
  - Pseudopotentials
  - Tight-binding models

Acknowledgements

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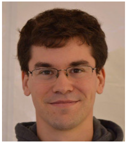

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Questions?

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