On the Nature of Self-Consistency in Density Functional Theory

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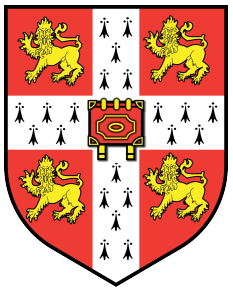

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The Talk

1) Background: Self-consistency in density functional theory.

2) Methods development: Good practice and analysis techniques.

3) New Methods: Brief comment on novel methodology.

Self-Consistency in Density Functional Theory

Atomic species and positions $\longrightarrow$ CASTEP $\longrightarrow$ Observable Properties...

Kohn-Sham density functional theory

$$H^{\mathrm{KS}}[\rho]\phi_i = \epsilon_i \phi_i$$
$$\rho = \sum_i f_i |\phi_i|^2$$

A *non-linear eigenvalue problem*: how to solve?

Self-Consistency in Density Functional Theory

Atomic species and positions $\longrightarrow$ CASTEP $\longrightarrow$ Observable Properties...

Ongoing effort to make this as efficient and robust as possible.

Self-Consistency in Density Functional Theory

Construct Kohn-Sham Hamiltonian $H^{\mathrm{KS}}[\rho^{\mathrm{in}}]$ for some input particle density ρ^{in}

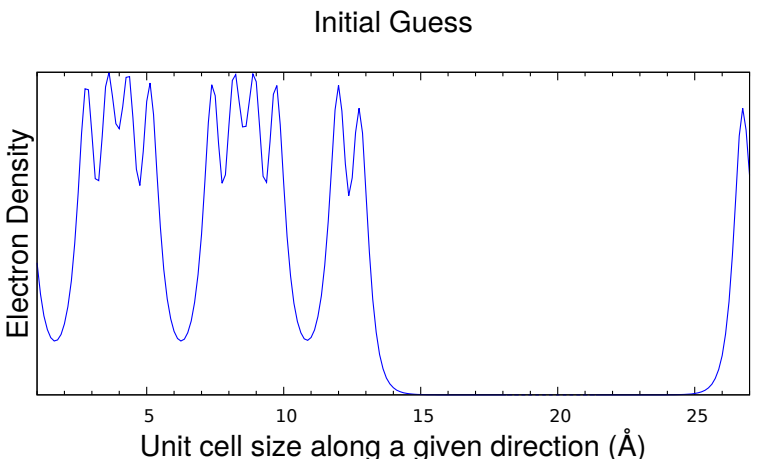
Find the eigenfunctions $\{\phi_i\}$ and use to construct an output particle density $\rho^{\mathrm{out}} = \sum_i f_i |\phi_i|^2$.

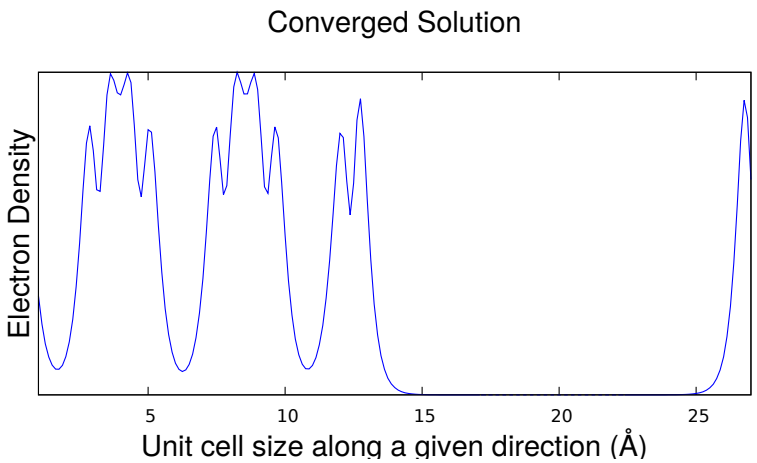
Is $\rho^{\mathrm{in}} = \rho^{\mathrm{out}}$? If so, ρ is *self-consistent*, and we have a solution to Kohn-Sham DFT.

Self-Consistency in Density Functional Theory

But what if $\rho^{\text{in}} \neq \rho^{\text{out}}$?

Define an iterative algorithm that gets us to $\rho^{\text{in}} = \rho^{\text{out}}$ starting from an initial guess ρ^{guess}





Good Practices in Methods Development

'Ease of convergence' to a self-consistent solution depends on heavily on the input system

- Bulk AI – easy
- Transition metal oxide surface – not so easy

Good Practices in Methods Development

Goals:

1) Identify and analyse all sources of divergence in Kohn-Sham DFT

2) Construct a representative ‘test suite’ for methodological development

3) Formalise/propose analysis tools that allow one to discern whether method X is ‘better’ than method Y

Good Practices in Methods Development

So, we have examined all sources of divergence, and constructed a representative test suite, what now?

Run a sample of existing methods (contemporary, historically relevant, etc.) through the test suite and analyse the output.

Good Practices in Methods Development

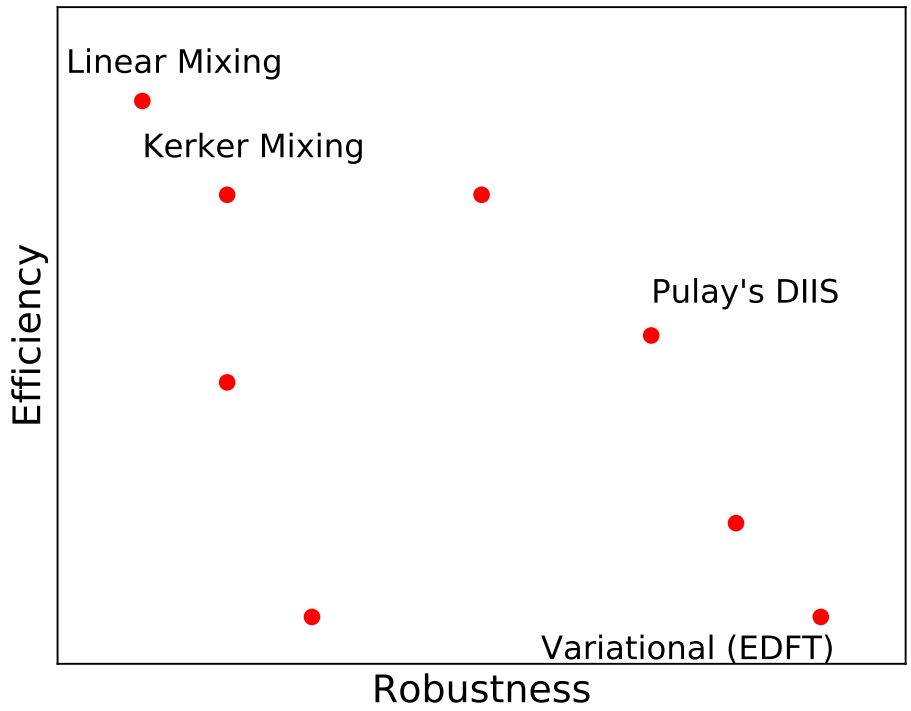
Analysis: what makes these methods 'good'?

Def: Robust. Percentage of test suite converged.

Def: Efficiency. Quantify the speed of convergence for systems that *do* converge.

Good Practices in Methods Development

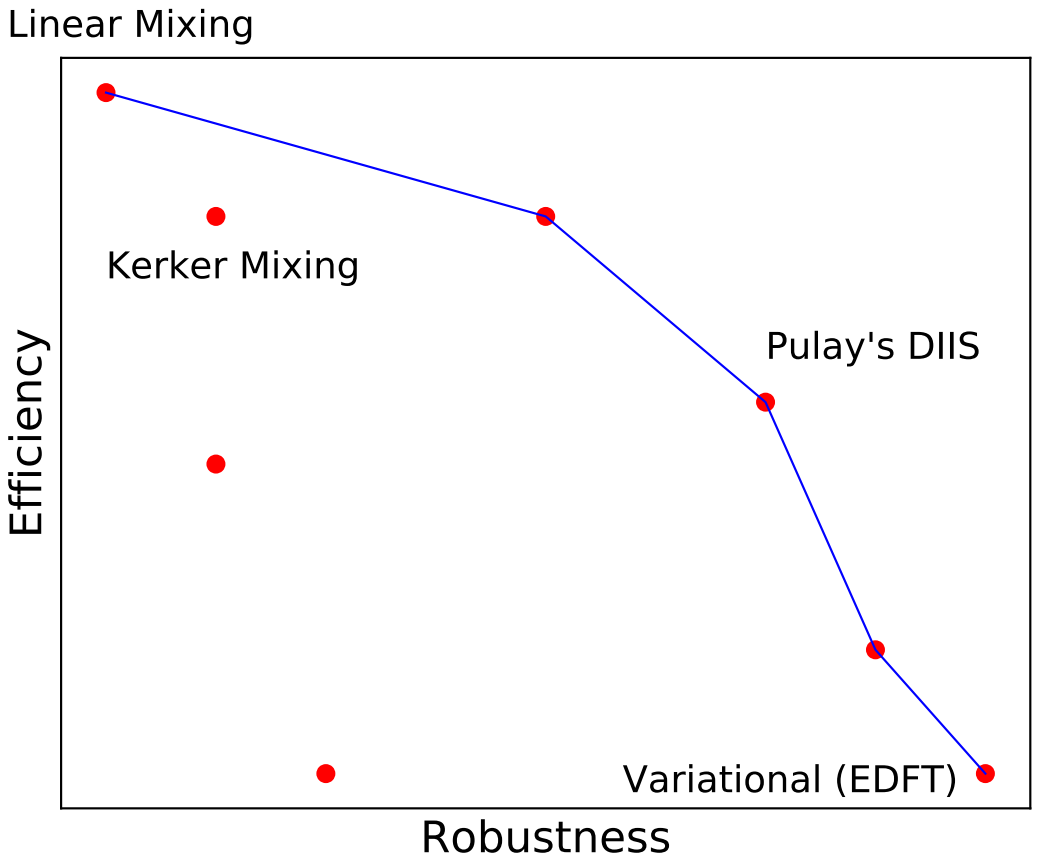
Robustness vs. efficiency: *Pareto analysis*



Choose a 'Pareto optimal' method

Good Practices in Methods Development

A DFT developer should choose a method on the Pareto front.

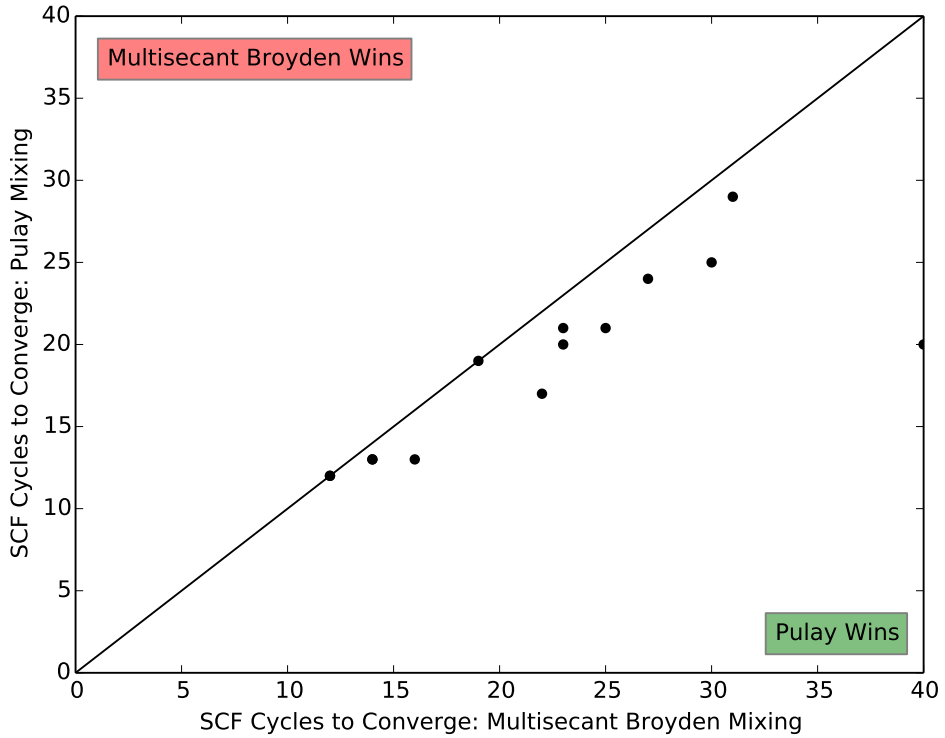


Good Practices in Methods Development

Q: If I create a method, how will I know if it lies on the Pareto front without rerunning all analysis?

Good Practices in Methods Development

A: Take a 'similar' method that lies on the Pareto front, and compare



Can identify particular strengths and weaknesses, etc.

Good Practices in Methods Development

In summary:

- Creates a well-motivated, standard benchmark
- Provides tools to systematically compare methods
- Increases transparency and creates a better practice for both DFT developers, and method developers

See arXiv:1803.01763

Thanks for listening