Targeting Real chemical accuracy at the EXascale

User experience with TurboRVB

Webinar, 8 February 2023

Giacomo Tenti, - SISSA

Targeting Real Chemical Accuracy at the Exascale project has received funding from the European Union Horizoon 2020 research and innovation programme under Grant Agreement **No. 952165.**

My experience with TurboRVB

First contact with TurboRVB:

Study of the hydrogen chain with a Pfaffian wave function (Master's thesis with prof. Sandro Sorella)

Learning Turbo **the hard way** (complex workflow, lots of parameters to control…)

The **easy way**:

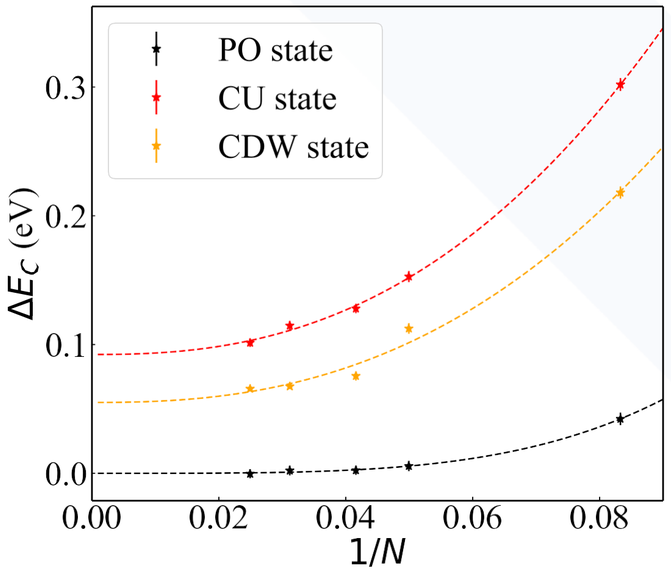
Using TurboGenius, the workflow of a typical calculation becomes much simpler!

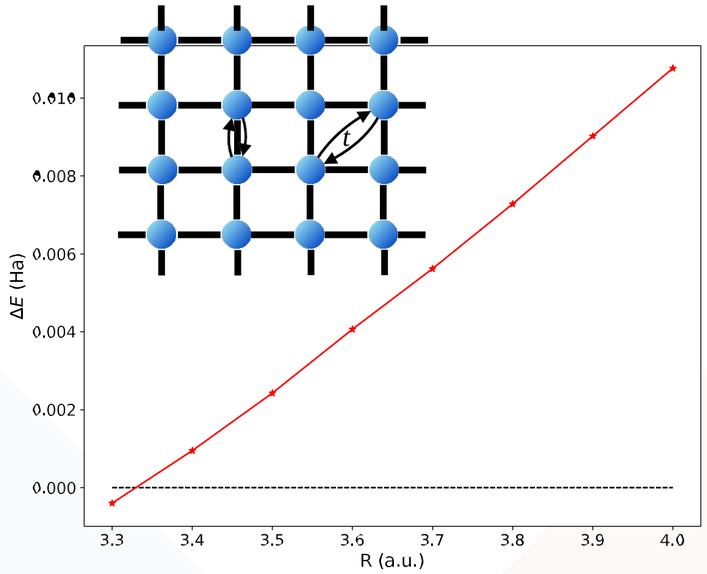
(Best way to learn how to use TurboRVB)

Moreover, advanced users still have complete control of the calculation

Projects with TurboRVB Quantum Monte Carlo Package SISSA:

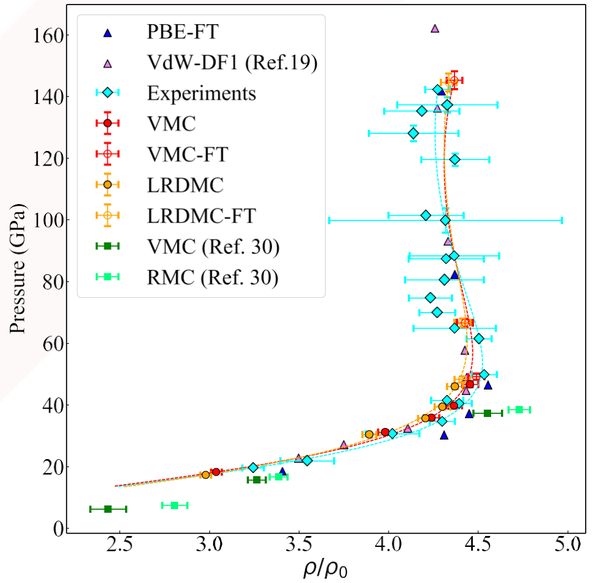
Structure optimization and CDW states





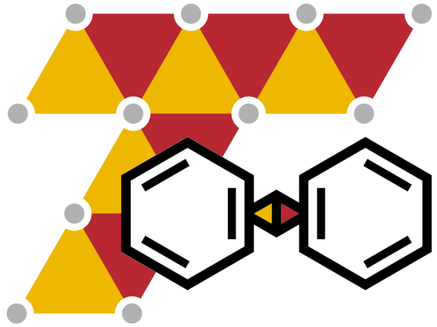
Phase diagram of hydrogen model systems

Generation of training sets for QMC - Machine Learning potentials



https://arxiv.org/abs/2301.03570

Modelling molecules and materials using TurboRVB



Andrea Zen
Università di Napoli Federico II

TREX webinar

February 8th , 2023

Implicitly multideterminat ansatz

TurboRVB: A many-body toolkit for *ab initio* electronic simulations by quantum Monte Carlo

Cite as: J. Chem. Phys. **152**, 204121 (2020); https://doi.org/10.1063/5.0005037
Submitted: 19 February 2020 . Accepted: 20 March 2020 . Published Online: 29 May 2020

Kousuke Nakano, Claudio Attaccalite, Matteo Barborini, Luca Capriotti, Michele Casula, Emanuele Coccia, Mario Dagrada, Claudio Genovese, Ye Luo, Guglielmo Mazzola, Andrea Zen, and Sandro Sorella

COLLECTIONS

Paper published as part of the special topic on Electronic Structure Software

Implicitly multideterminat ansatz

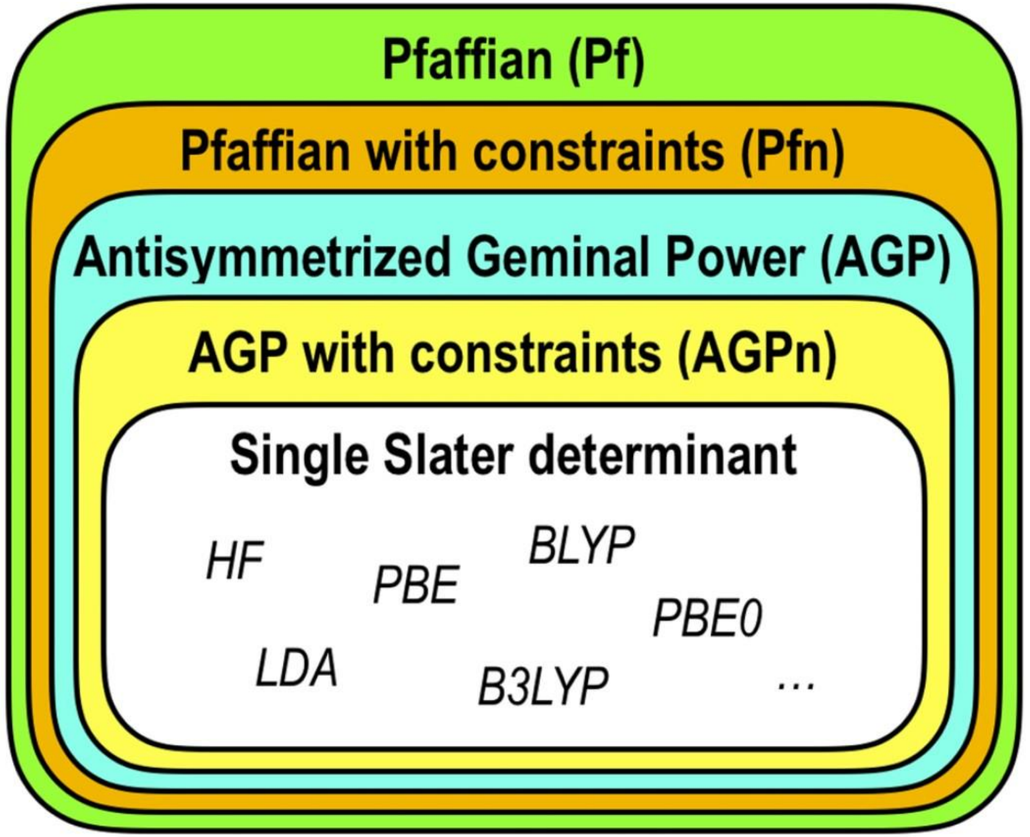


FIG. 3. *Ansatz* hierarchy. The output of Hartree–Fock (HF) or DFT simulations with different exchange-correlation functionals are special instances of the SD *Ansatz*.

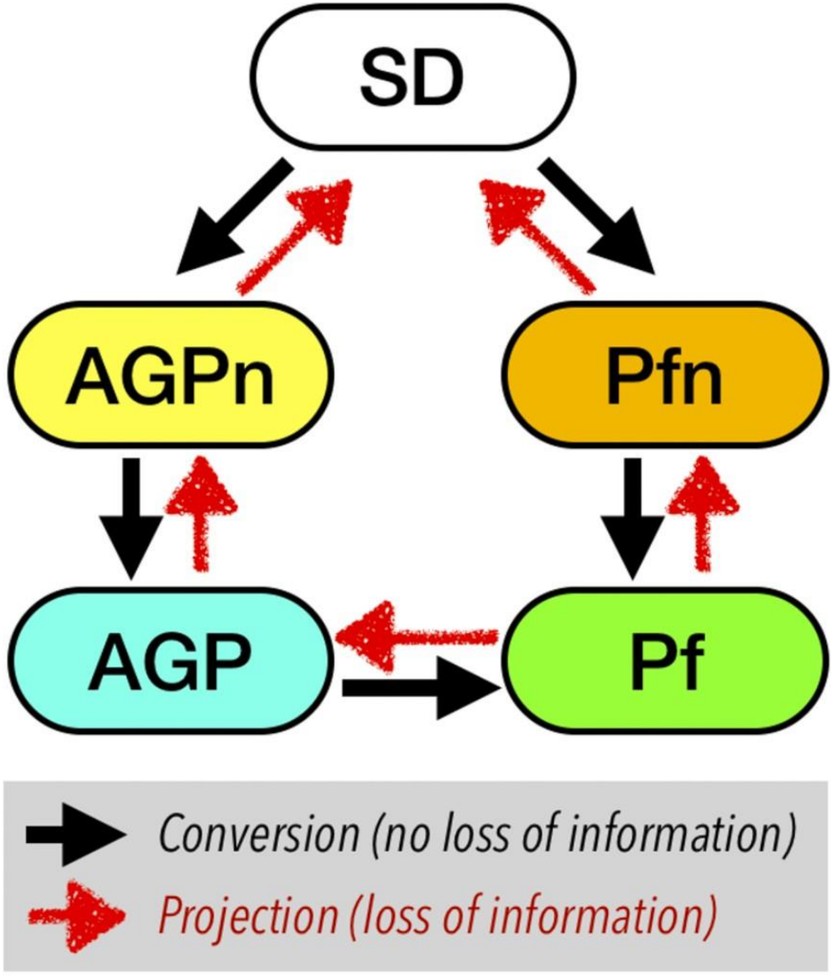
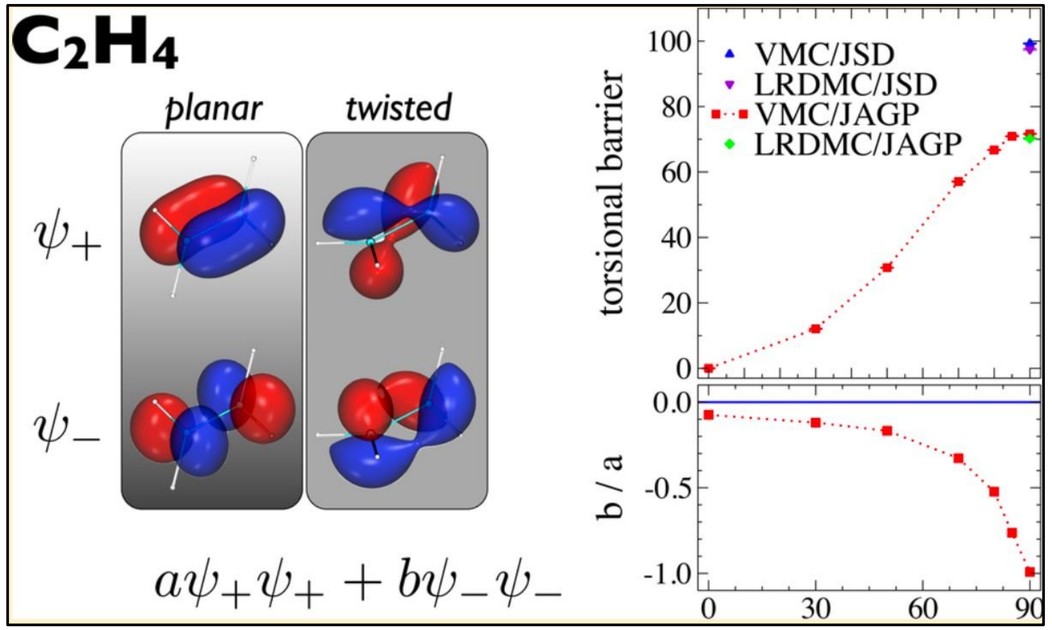


FIG. 4. *Ansatz* conversion.

J. Chem. Phys. 152, 204121 (2020)
TurboRVB: A many-body toolkit for ab initio electronic simulations by quantum Monte Carlo

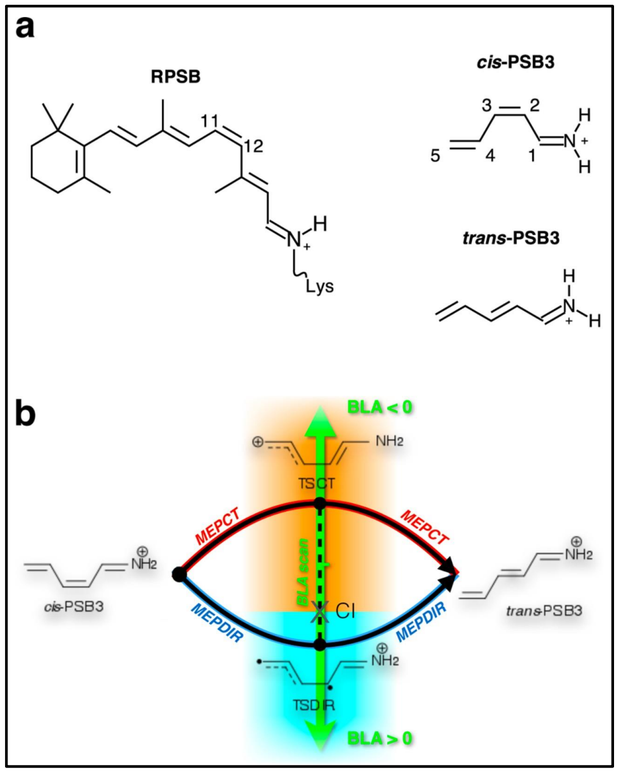
When do we need a multideterminat ansatz?

Breaking bonds



J. Chem. Theory Comput. 2014, 10, 1048−1061
Static and Dynamical Correlation in Diradical Molecules by Quantum Monte Carlo Using the Jastrow Antisymmetrized Geminal Power Ansatz

Transition states

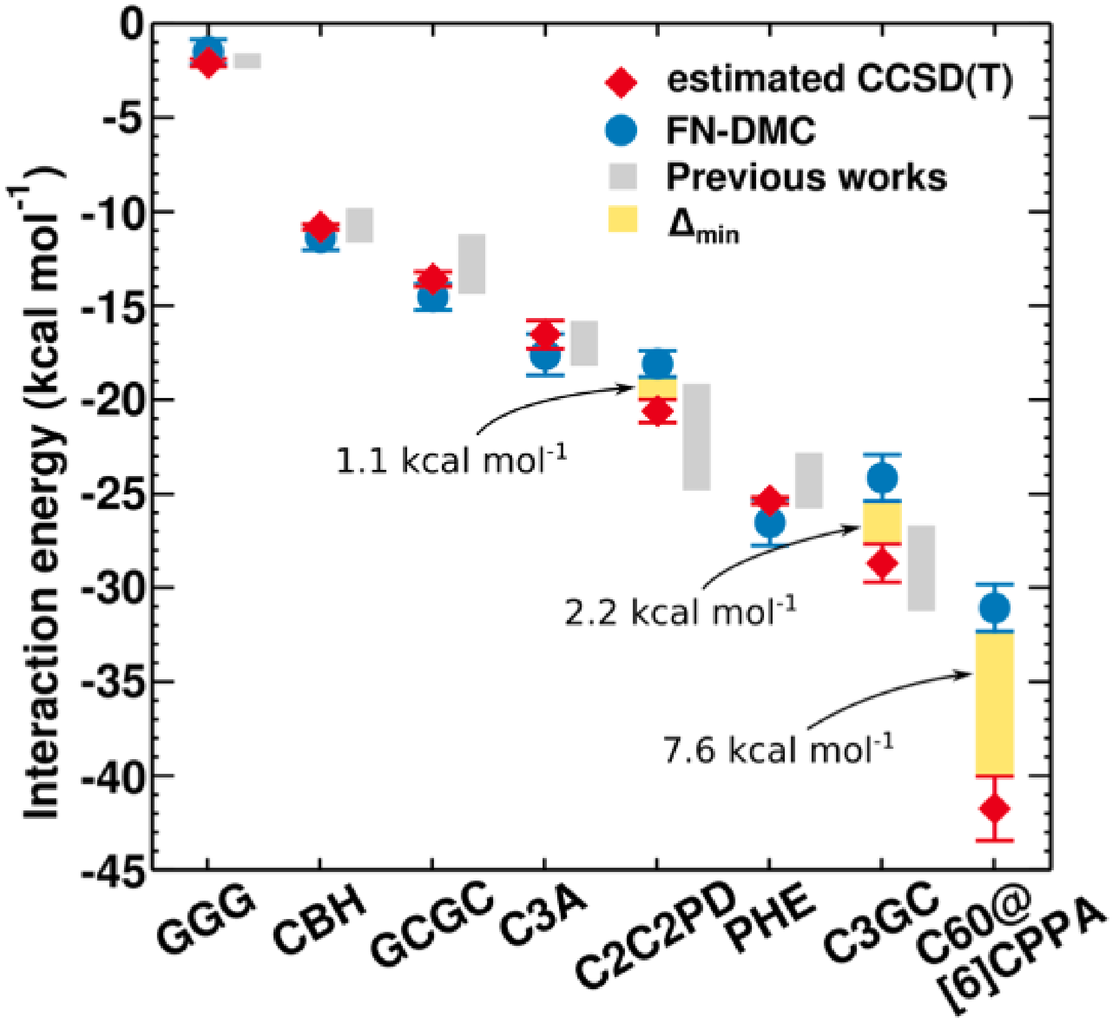


J. Chem. Theory Comput. 2015, 11, 992−1005
Quantum Monte Carlo Treatment of the Charge Transfer and Diradical Electronic Character in a Retinal Chromophore Minimal Model

Reference methods: FN-DMC & CCSD(T)

- Generally observed a **good agreement between the CCSD(T) and the FNDMC** (with a Slater-Jastrow guide function) evaluation of **non-covalent interactions**.

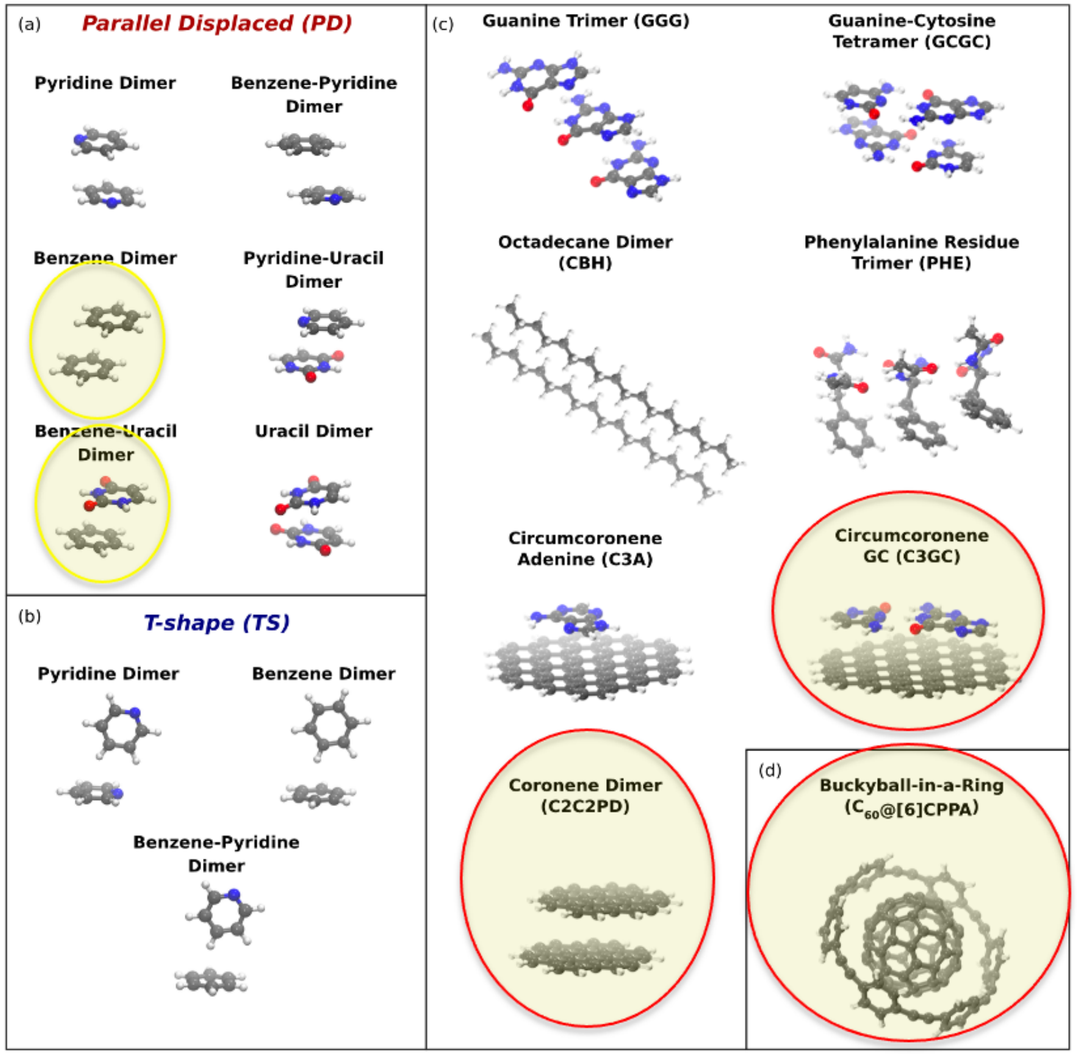
- Recently observed a **disagreement in large complexes** not coming from the known issues (small basis set, timestep bias, etc.)

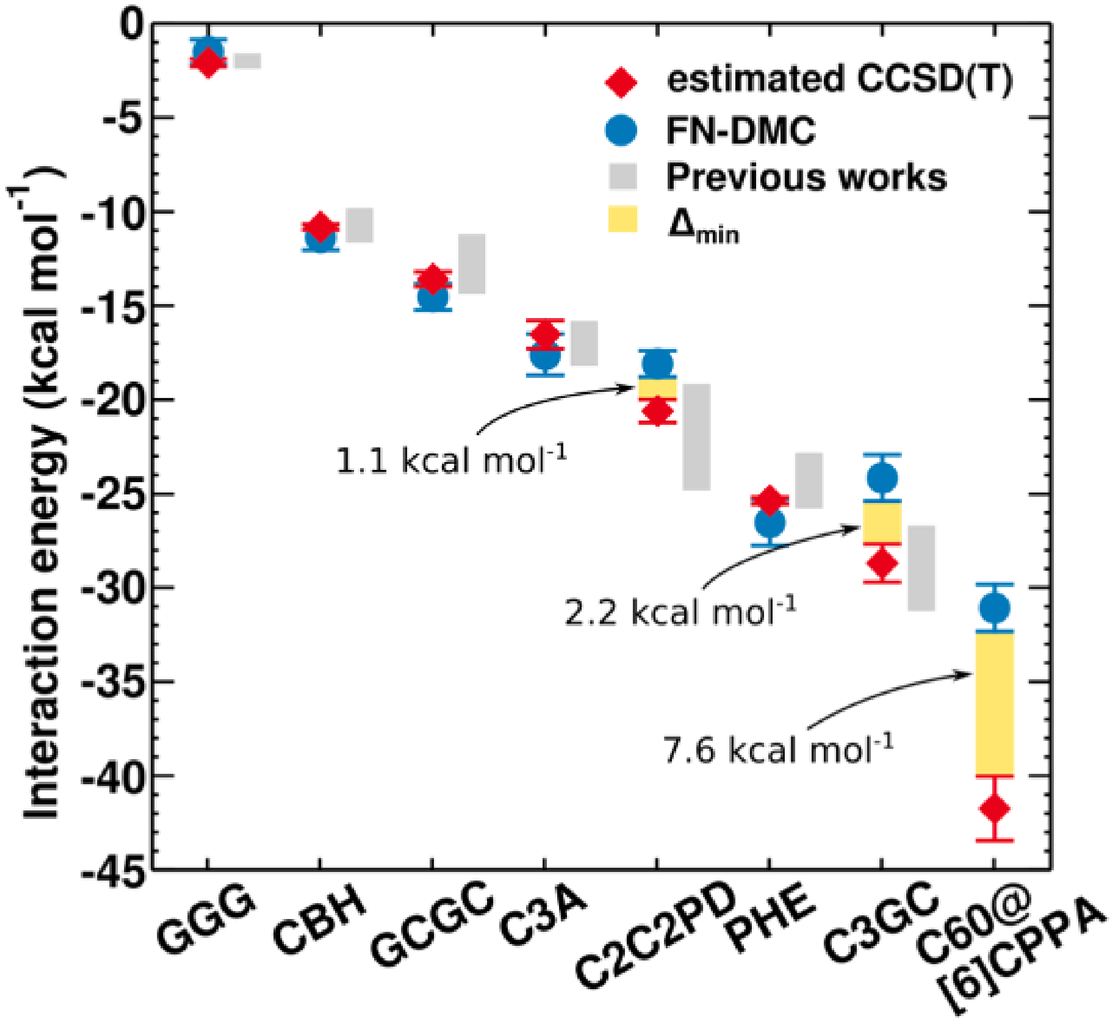


Nature Communications 12, 3927 (2021)
Interactions between Large Molecules: Puzzle for Reference Quantum-Mechanical Methods

Issue with pi-pi interactions?

Is FN-DMC or CCSD(T) having accuracy issues?

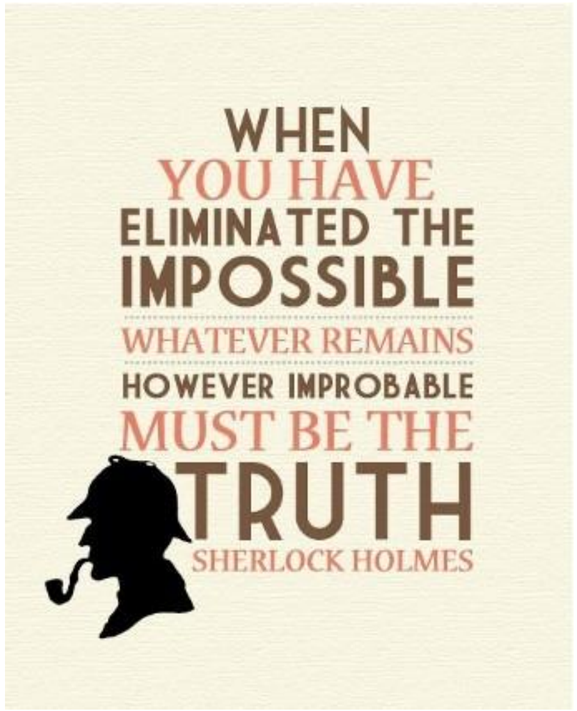




Nature Communications 12, 3927 (2021)

Interactions between Large Molecules: Puzzle for Reference Quantum-Mechanical Methods

Inspecting FN-DMC weaknesses



Weeknesses in FN-DMC:

- ~~Bugs in the code~~ *No, 2 codes agree* [1,2]
- ~~Pseudopotentials~~ *No, AE and PP agree* [1,2]
- ~~Optimization of Jastrow~~ *No: tested LA, TM & DLA* [1]
- ~~Determinant initialization~~ *No: tested LDA, PBE, PBE0* [1]
- FN beyond single Slater (**?**) *Work in progress*

1Y.S. Al-Hamdani, P.R. Nagy, A. Zen, D. Barton, M. Kállay, J.G. Brandenburg, A. Tkatchenko, *Interactions between Large Molecules: Puzzle for Reference Quantum-Mechanical Methods*, Nature Communications 12, 3927 (2021).
Uses CASINO, DMC with pseudopotentials testing LA/TM/DLA.

2] A. Benali, H. Shin and O. Heinonen, Quantum Monte Carlo benchmarking of large noncovalent complexes in the L7 benchmark set, JCP 153, 194113 (2020).
Uses QMCPACK, DMC with all-electrons.

FN beyond single Slater

Interaction energy is the energy difference between two or more systems. It’s a small fraction of the total energy of a system.
Difficulty: keeping the quality of the wave function (optimisation) consistently good in two or more systems.

pyscf

Kosuke Nakano

TREX-I/O