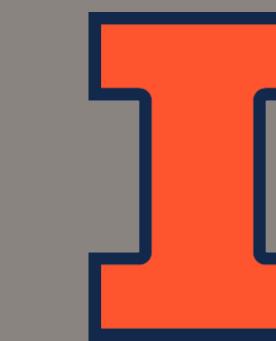


QMC-HAMM: High accuracy multiscale models using quantum Monte Carlo

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The QMC-HAMM project is focused on using high accuracy ab initio simulations to develop multiscale models with unprecedented accuracy.

Our collaboration focuses on four goals:

- 1) Improving models of materials by linking them to high accuracy first principles calculation, with error assessments
- 2) Methods to make quantum Monte Carlo simulations more useful for deriving larger length scale models
- 3) Application of the above two ideas to understanding the structure and electronic behavior of layered graphene and other 2D materials, and
- 4) Application of the above two ideas to understanding hydrogen at high pressure.

First principles calculations

Almost all of condensed matter can be described by the electronic Hamiltonian

$$-\frac{1}{2} \sum_i \nabla_i^2 + \sum_{i < j} \frac{1}{r_{ij}} - \sum_{i,\alpha} \frac{Z_\alpha}{r_{i\alpha}} + \sum_{\alpha < \beta} \frac{Z_\alpha Z_\beta}{r_{\alpha\beta}}$$

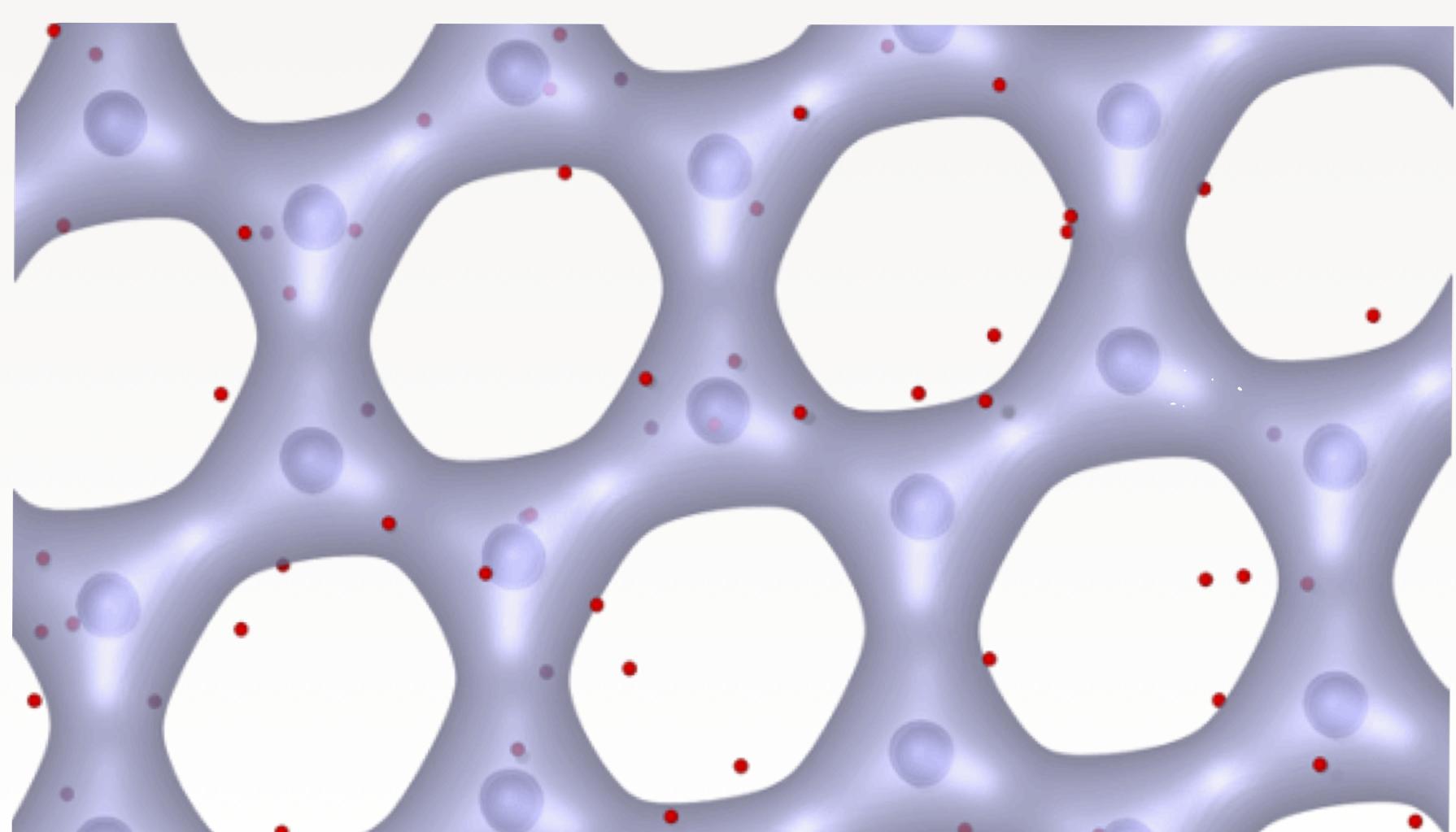
With the Schrödinger equation

$$H\Psi_i(r_1, r_2, \dots) = E_i\Psi_i(r_1, r_2, \dots)$$

Our primary technique is quantum Monte Carlo.

Electron interactions are treated explicitly via sampling.

One Monte Carlo sample of electrons in graphene:

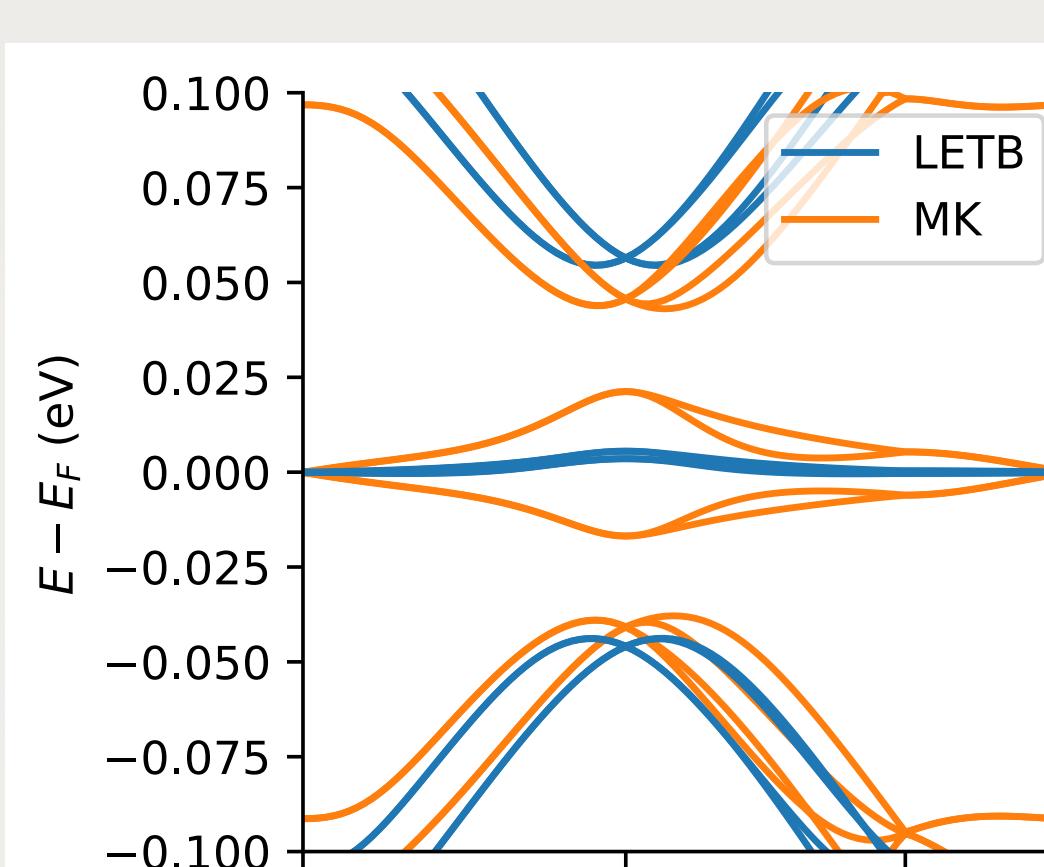


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Twisted bilayer graphene models

Twisted bilayer graphene is well-known to exhibit a number of correlated electronic phases as a function of twist. The electronic structure depends on a detailed combination of geometrical factors and electronic structure.



Local environment tight binding (Ref 10)

Using machine learning methods, we determined an accurate model based on density functional theory calculations.

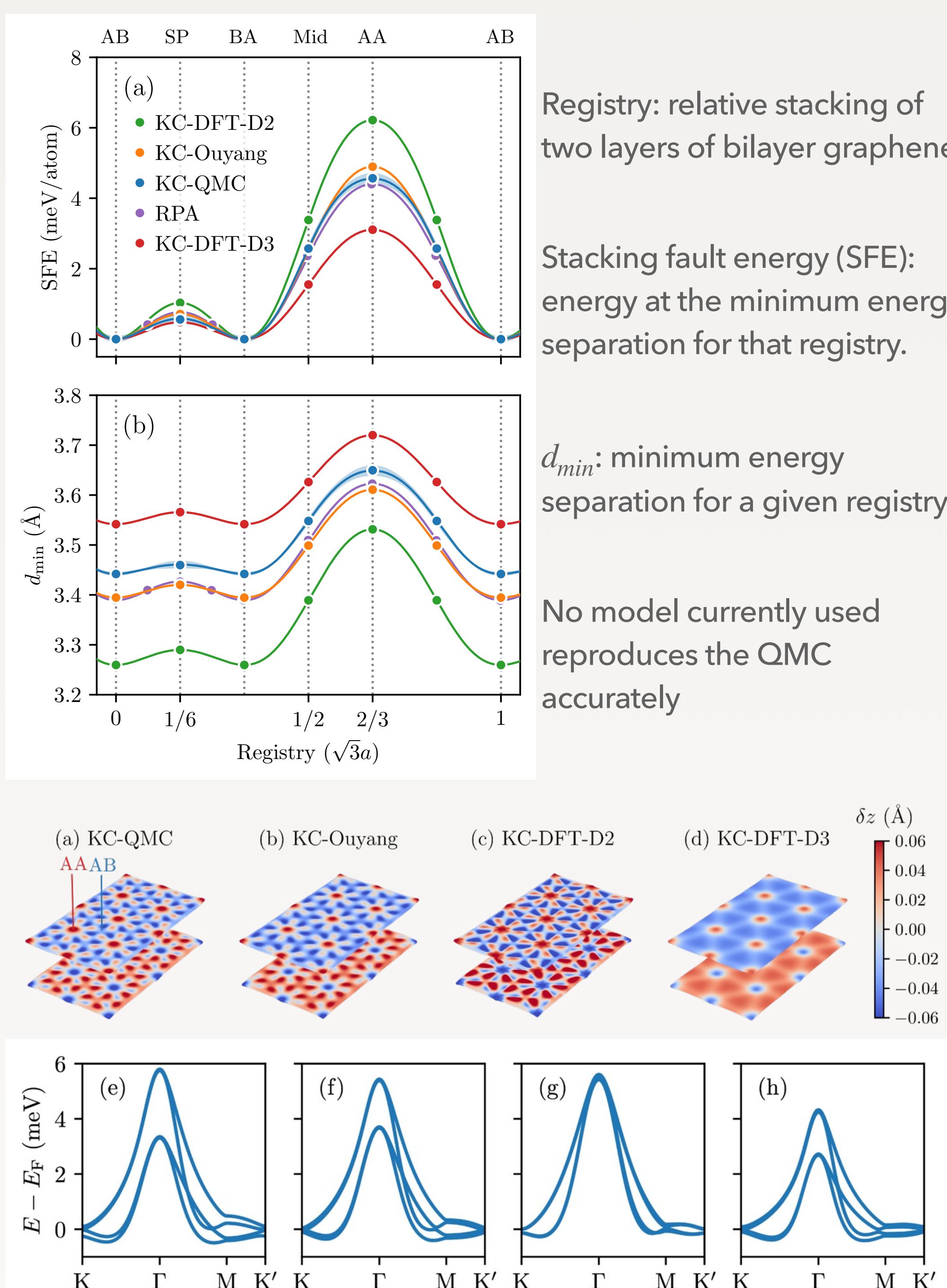
With the accurate model, we find significantly less electron-hole symmetry than in the industry standard Bistritzer-MacDonald model.

`pip install bilayer_letb`

Quantum Monte Carlo based van der Waals potentials (Ref 3)

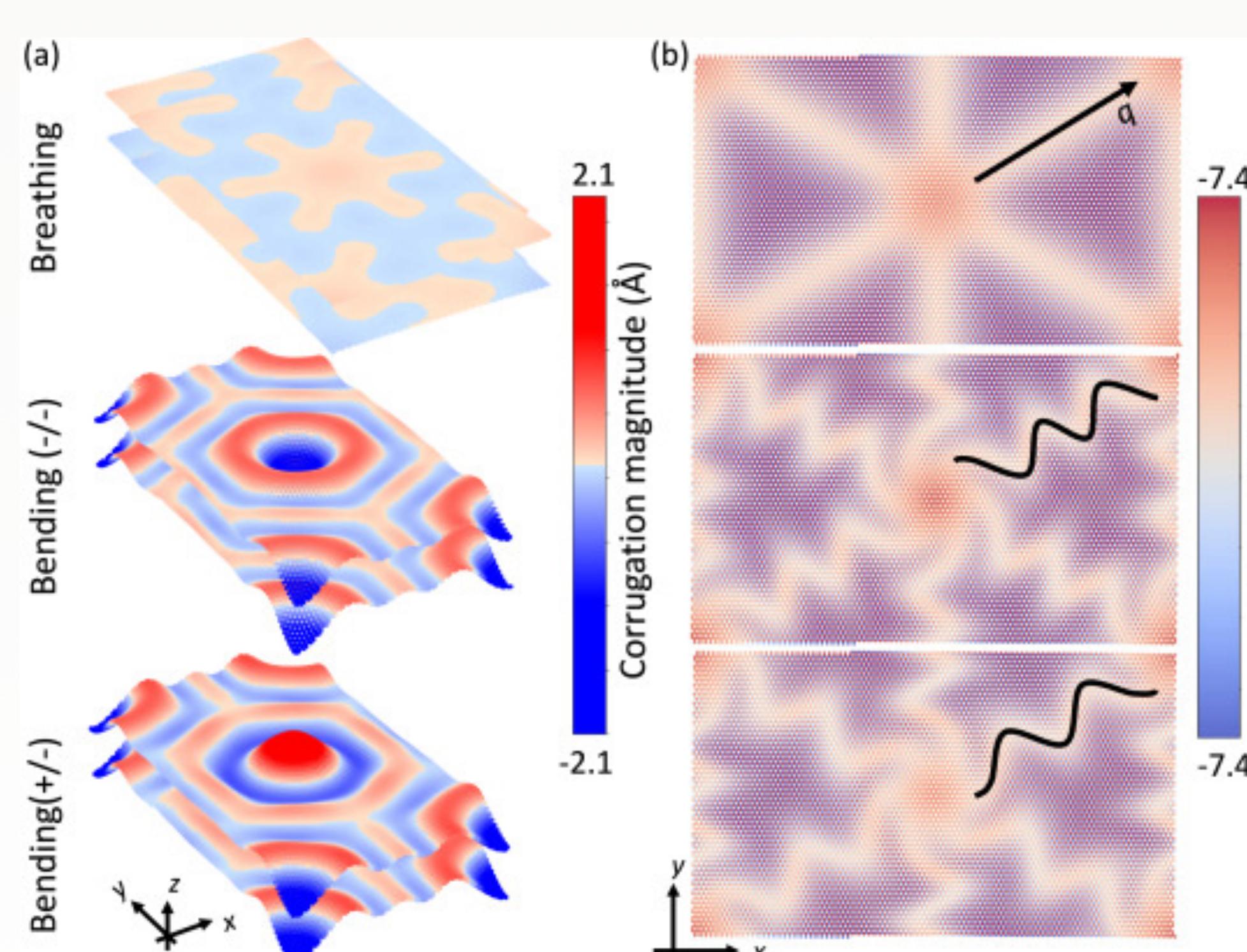
We computed the interlayer interaction between graphene bilayers using quantum Monte Carlo. The new potential leads to differences in predicted corrugation and then differences in predicted electronic structure.

Data: <https://commons.datacite.org/doi.org/10.18126/otff-eyc8>

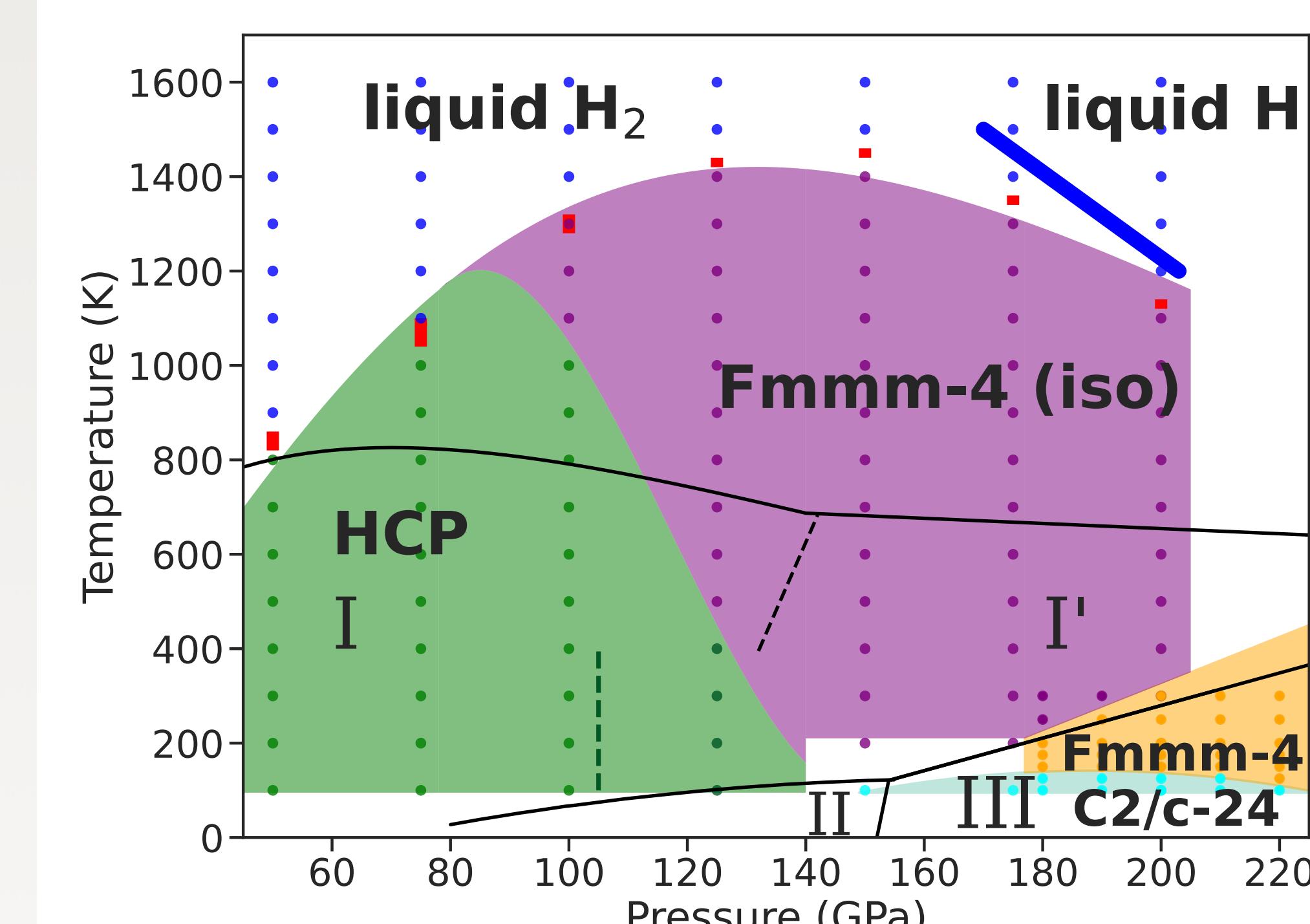


New corrugation patterns in graphene (Ref 1)

With accurate potentials, we investigated low-energy configurations in twisted bilayer graphene. Helical patterns of corrugation are competitive with the lowest energy structures found so far.



New phase diagrams of hydrogen (Ref 2)



The primary interest of this application area is the determination of the phase diagram at high pressure over a large range of temperatures. In Ref 4, we used quantum Monte Carlo simulations to derive an atomicistic potential accurate across multiple phases of hydrogen. These simulations resulted in the discovery of a new potential phase in hydrogen at high pressure and temperature, as shown in the figure above.

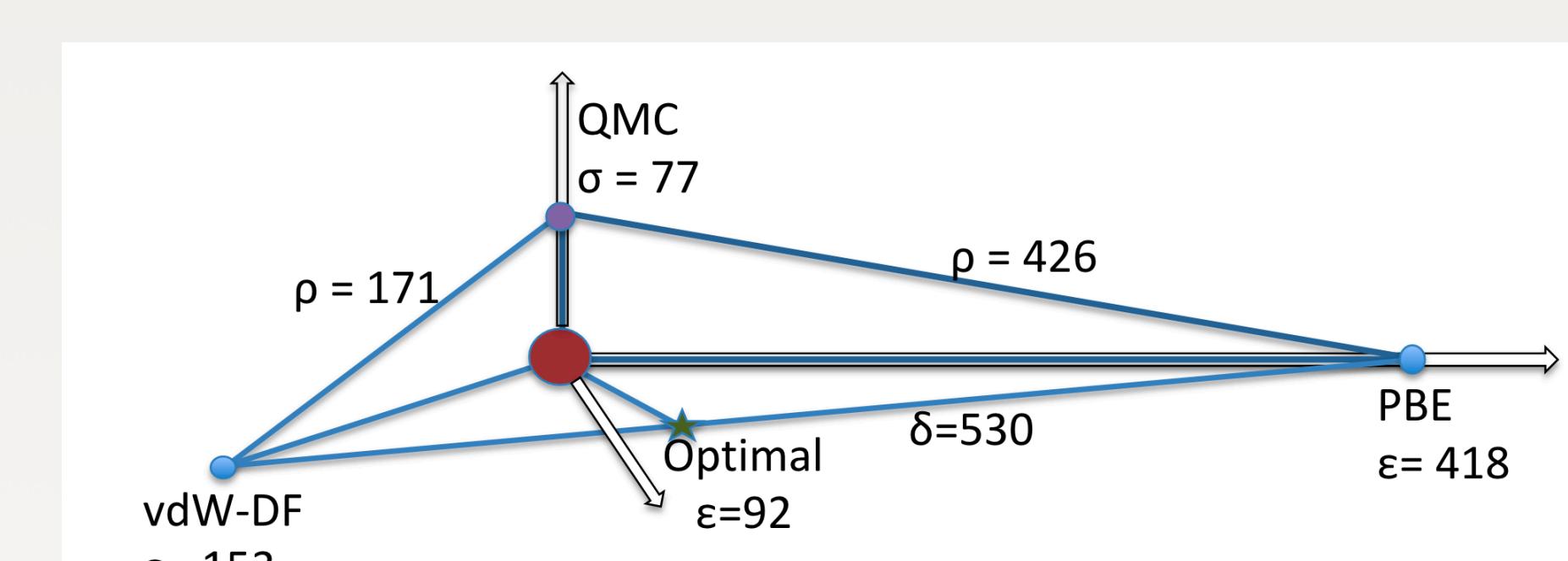
The QMC data is available on our website and in the Materials Data Facility. This data will be helpful in developing models of both electrons in hydrogen and in developing atomic-level models.

Data available at <https://qmc-hamm.hub.yt/data.html>

Why we need more accurate data for mesoscale models/ uncertainty quantification (Ref 6)

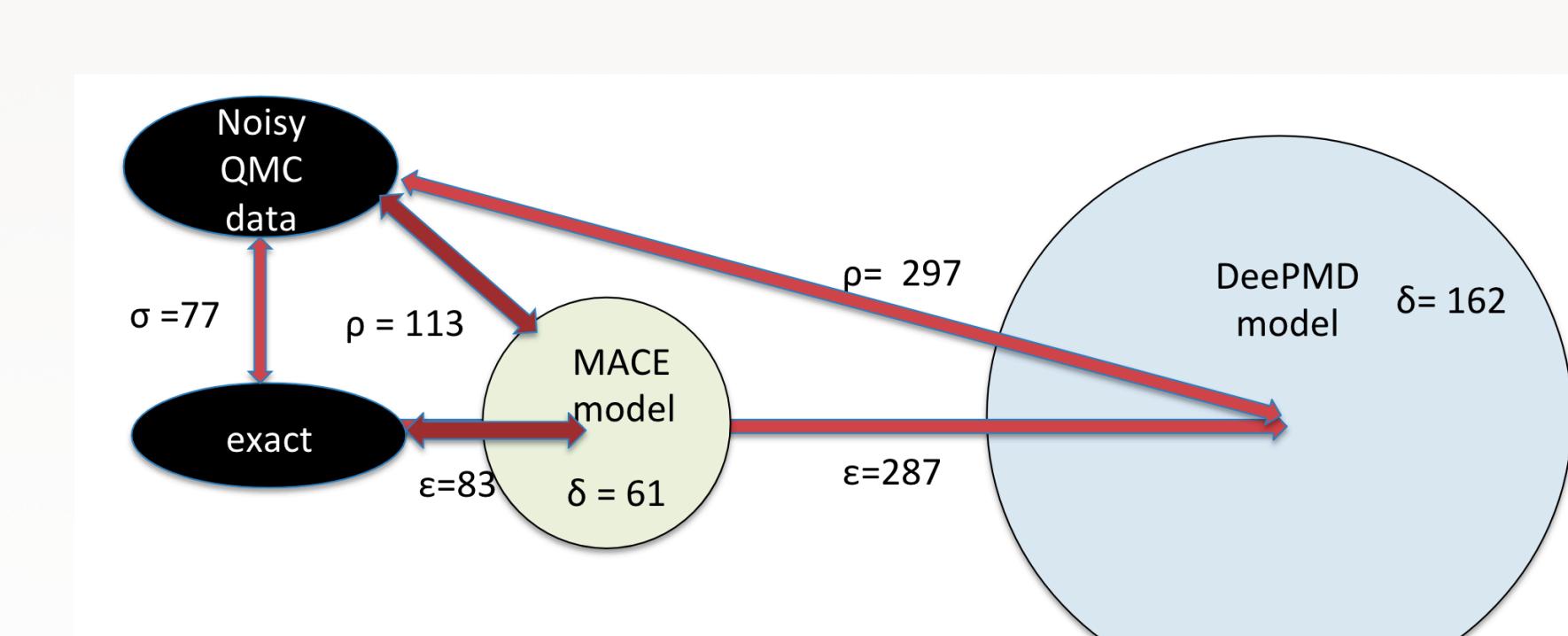
With quantum Monte Carlo, we assess the quality of density functional theory approximations, here in hydrogen forces.

Here ρ is the RMS error.



Models fit to QMC are better than DFT

The error in machine learning models MACE and DeepMD when fit to QMC data is smaller than running first principles DFT, at a computational cost orders of magnitude smaller.



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