Gaussian Approximation Potentials at work: from silicon to water

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Outline

- gap between QM and interatomic potentials
- generating potentials
- Gaussian Process
- results
  - silicon
  - tungsten
  - water

From QM...

- quantum mechanics is the 'ultimate truth' but...
- … expensive to solve
- sequence of approximations
  - full CI: 1 atom
  - Coupled Cluster: few atoms
  - QMC: few molecules
  - DFT: few hundred atoms
  - tight-binding: few thousand atoms
  - interatomic potentials: millions of atoms

Massive gap!

… to interatomic potentials

- energy is the sum of atomic (pair, triplet, etc.) energies
- atomic energy depends on neighbouring atoms
- electronic problem is *not* solved

$$E_{tot} = \sum_i \varepsilon\left(\{r_{ij}\}\right) + \sum_{i,j} \hat{L}_i \hat{L}_j \frac{1}{r_{ij}}$$

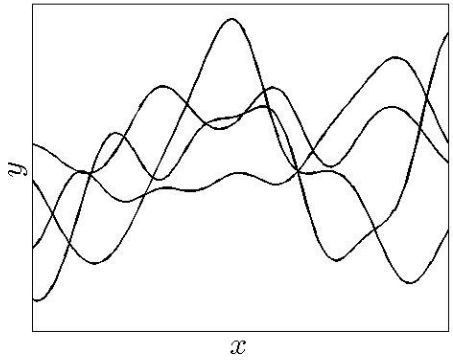
finite range

electrostatics

Generating potentials

- How is an interatomic potential generated?
  - empirical, analytic formula
  - choose target properties
  - fit free parameters to reproduce properties
  - hope that the formula remains reasonably valid everywhere in the configuration space
- the GAP way
  - no fixed formula
  - search in the space of smooth functions
  - identify target configurations
  - use arbitrary precision QM data as evidence
  - extend target set if needed

$$V_{ij} = 4\epsilon\left(\frac{\sigma^{12}}{r_{ij}^{12}} - \frac{\sigma^{6}}{r_{ij}^{6}}\right)$$

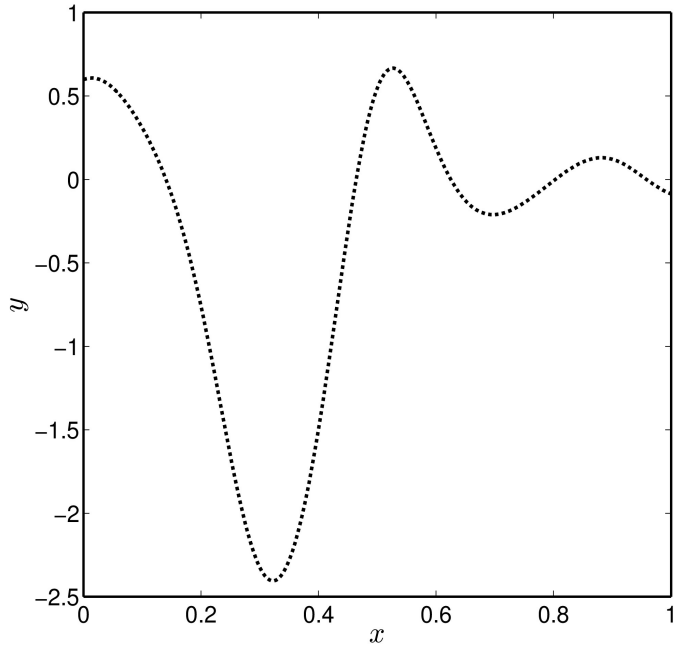


Gaussian Process

- inference method: given some data, infer function values at arbitrary points

$$G(x, x') = \exp\left(\frac{-(x - x')^2}{2\theta^2}\right)$$

$$y(x) = \sum_n \alpha_n G(x, x_n)$$



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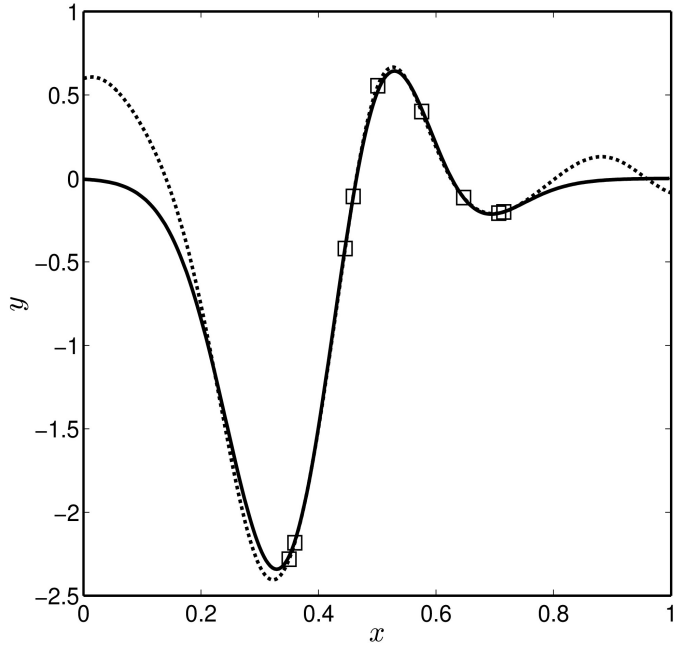

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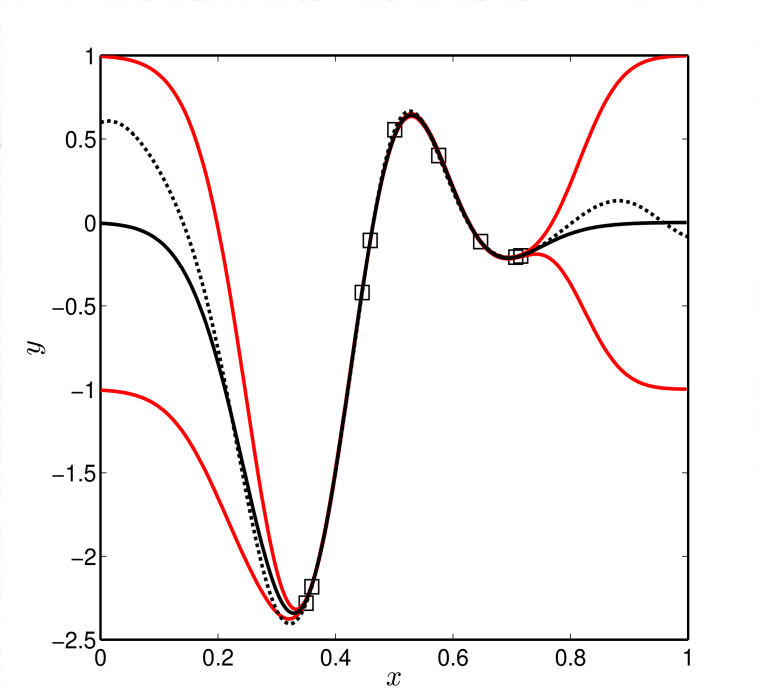

Silicon

- Data: CASTEP, DFT-PW91
  - crystalline configurations
    - diamond, β-tin, simple hexagonal
  - liquid
  - amorphous
- Results:
  - elastic constants
  - phonon spectrum
  - force correlation
  - phase diagram

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	CASTEP	GAP
C_{11} / GPa	154	154
C_{12} / GPa	56	55
C_{44} / GPa	75	73

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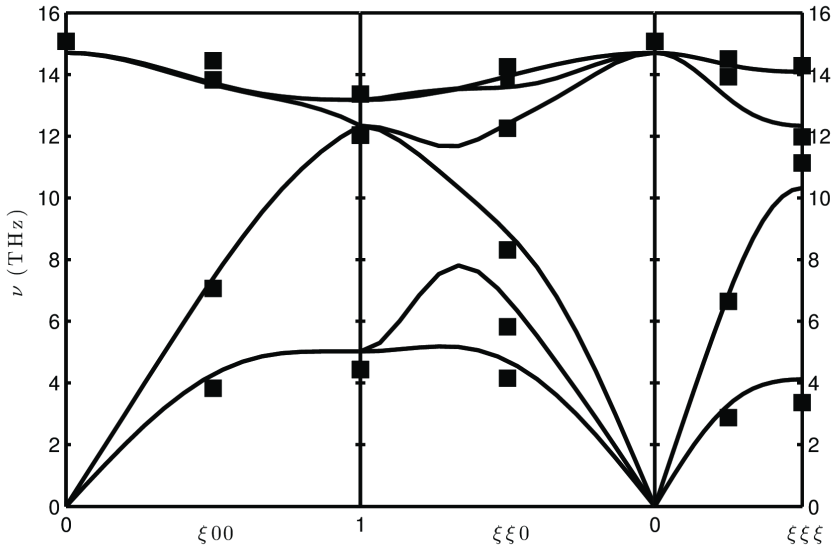

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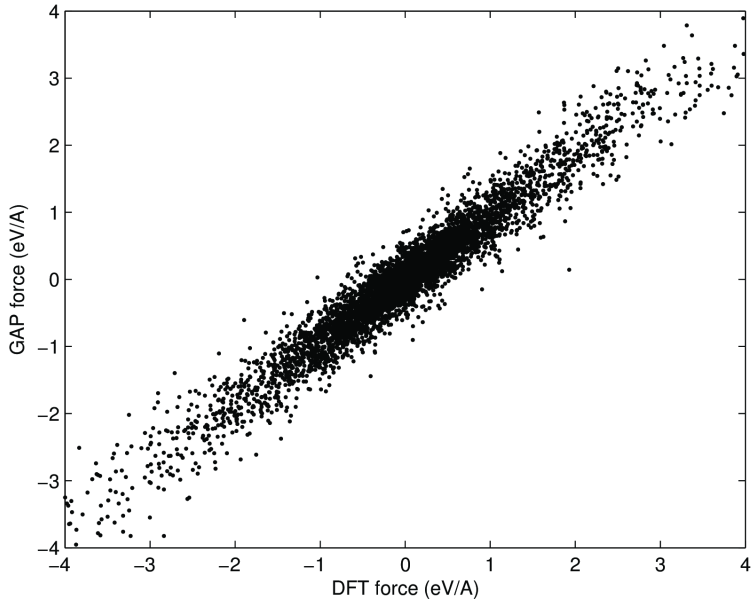

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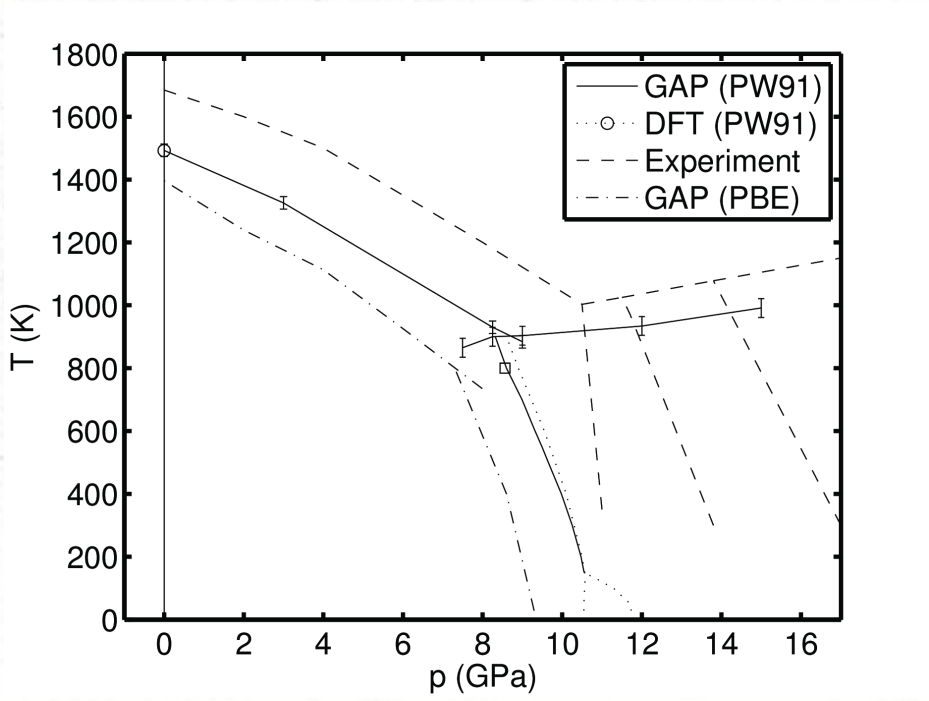
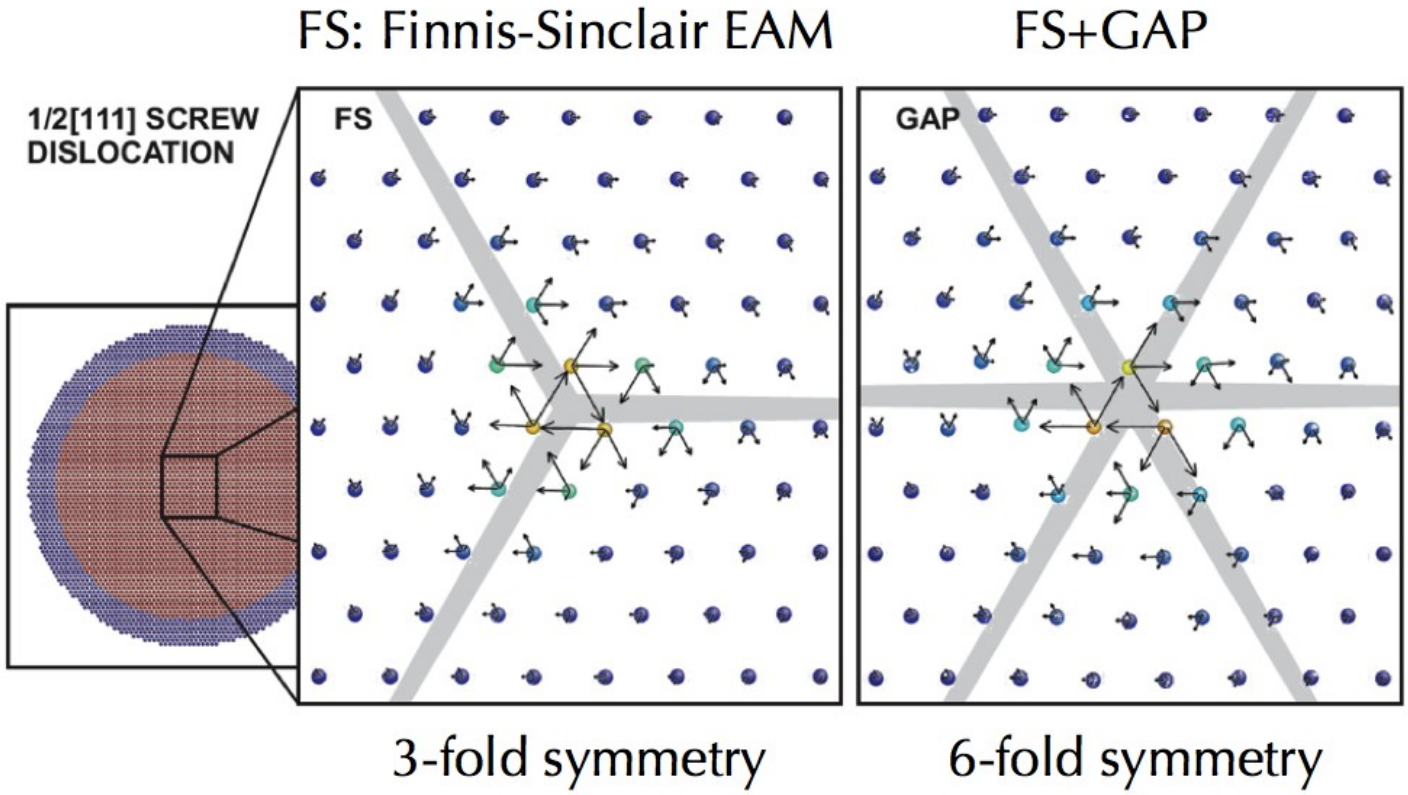


Tungsten

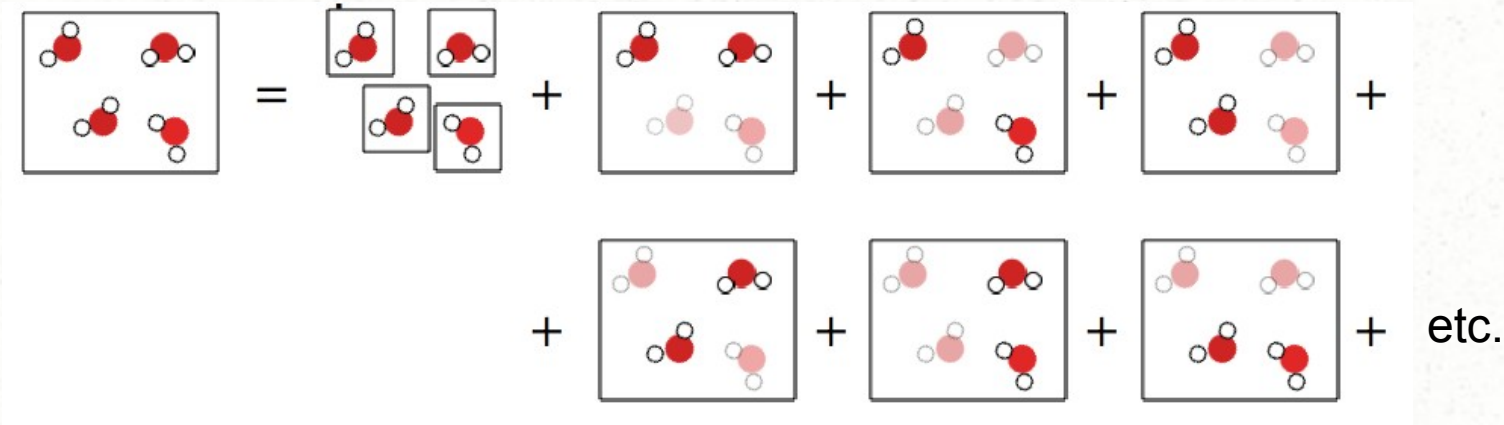

FS: Finnis-Sinclair EAM
FS+GAP
1/2[111] SCREW
DISLOCATION
FS
GAP
3-fold symmetry
6-fold symmetry

Water

- notoriously difficult to simulate
- interatomic potentials
  - fitted to reproduce some properties (rdf, vaporisation)
  - not very predictive otherwise
- DFT
  - water frozen at room temperature
- CC, QMC
  - expensive
  - systems not big enough – network structure...

Water

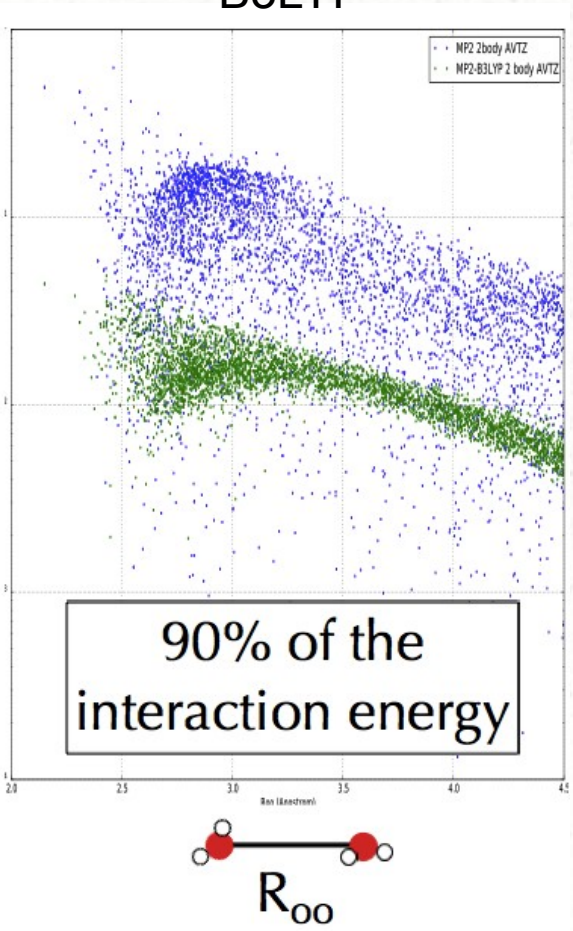
- Cluster expansion



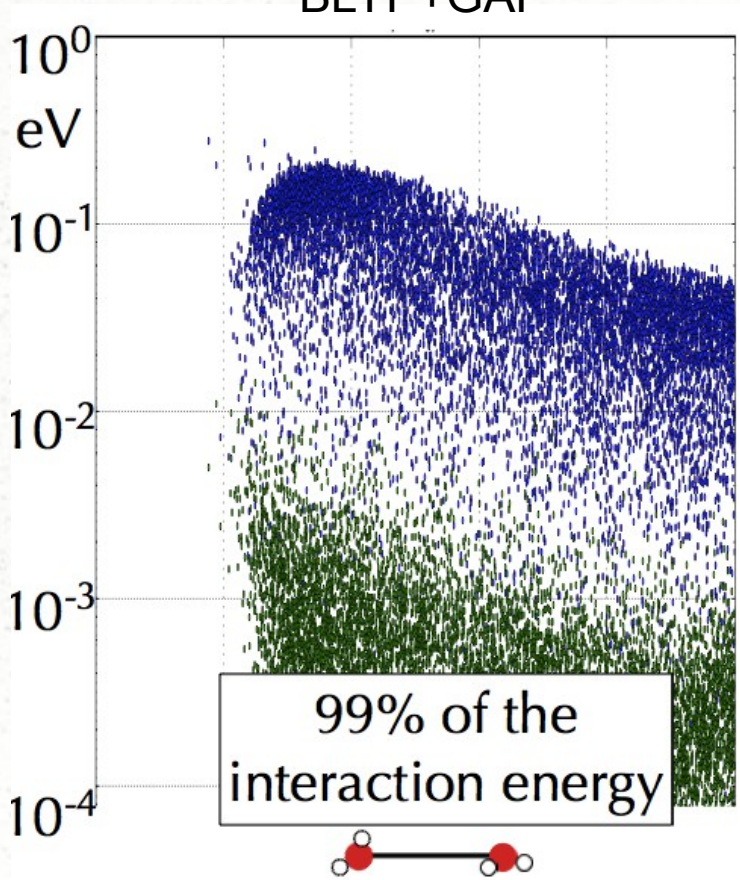
- GAP on dimers

Water

B3LYP



BLYP+GAP



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