Novel Approach for Parametrisation of Classical Potentials

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Outline

- how classical potentials work
- machine learning
- potential based on neural network
- representation of atomic environments
- bispectrum
- testing
- results

'Classic' classical potentials

$$V(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = V^{(0)} + \sum_i V^{(1)}(\mathbf{r}_i) +$$
$$\sum_{i<j} V^{(2)}(\mathbf{r}_i, \mathbf{r}_j) + \sum_{i<j<k} V^{(3)}(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) + \ldots$$

energy is sum of atomic energies

atomic energy depends on neighbouring atoms

no electronic problem is solved

Lennard-Jones potential

$$V_{LJ}(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^{6} \right]$$

Stillinger-Weber potential

$$V^{(3)} = \lambda \exp\left(\frac{\gamma}{r_{ij} - a} + \frac{\gamma}{r_{ik} - a} \right) \left(\cos\theta_{jik} + \frac{1}{3} \right)^2$$

our aim: high dimensional interpolation

Potentials based on Gaussian Process: Teaching

Atomic coordinates and local energies or forces

↓

Invariant representation

↓

Gaussian Process training

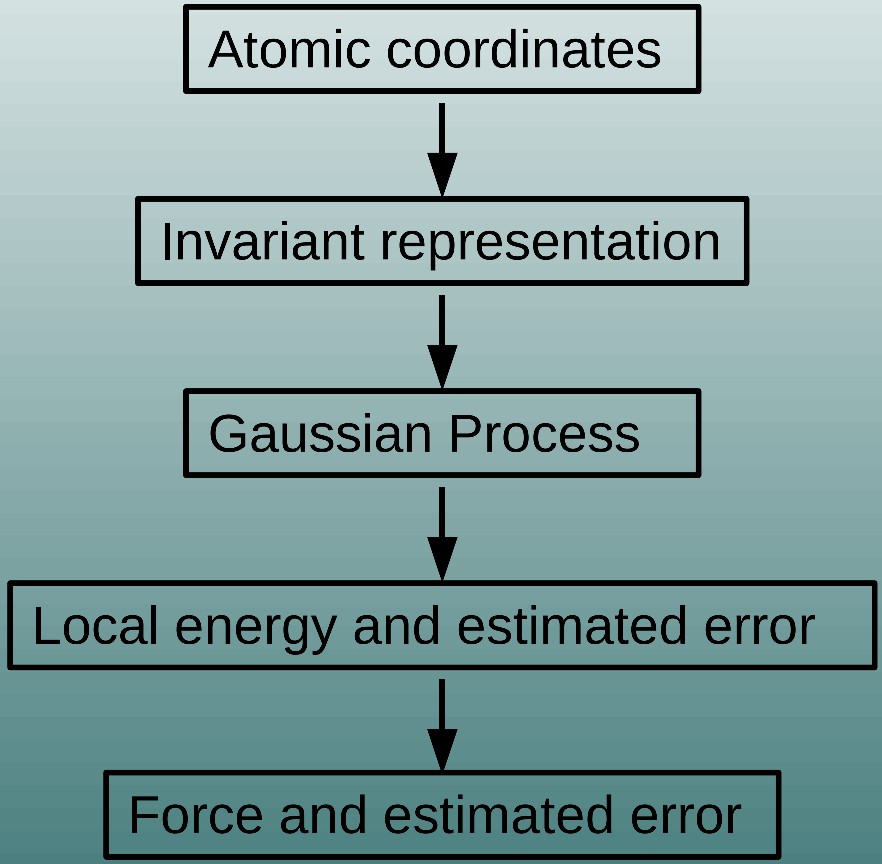
↓

Optimization of hyperparameters

Target can come from:

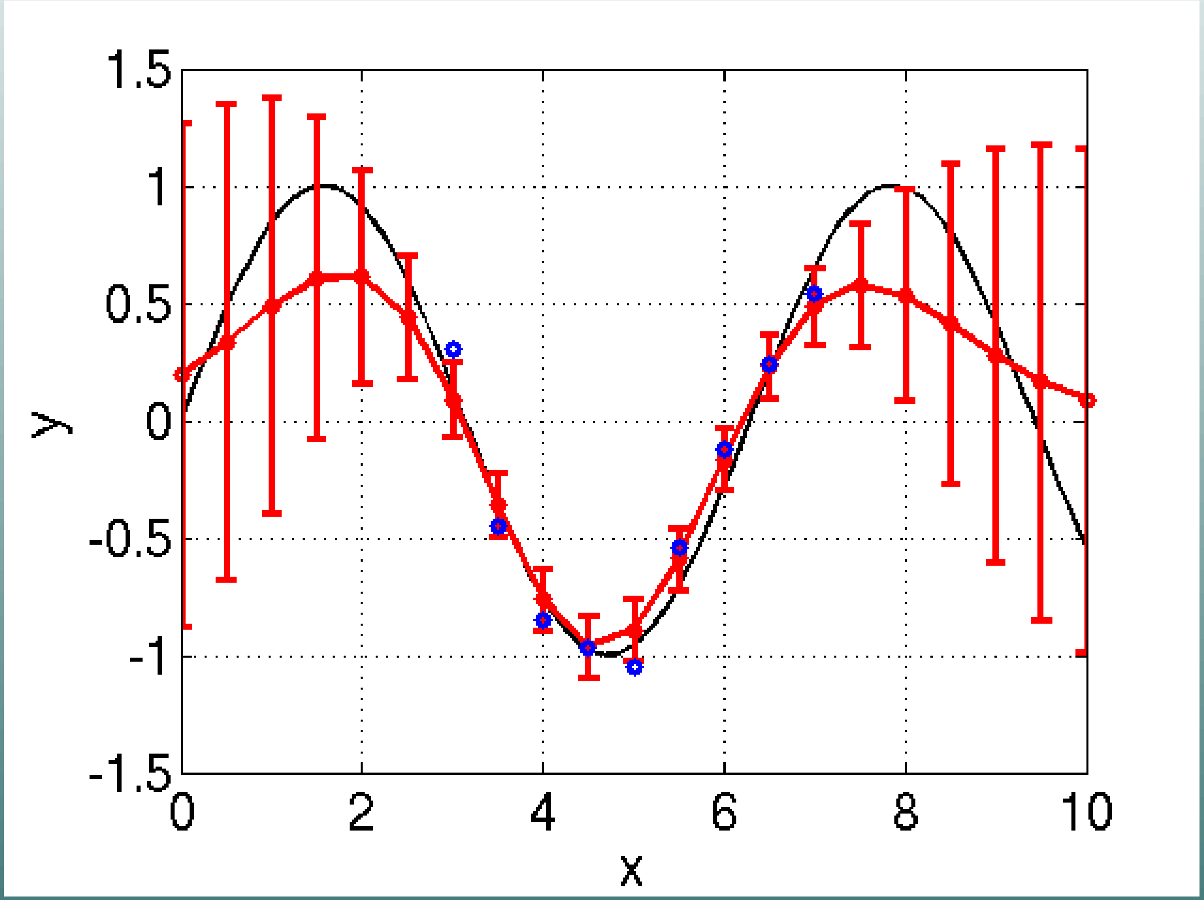
- DFT
- tight binding
- quantum chemistry

Potentials based on Gaussian Process: Usage



aim: accurate local energies and forces, cheaply: $\propto N\,M^2$
N: number of atoms
M: number of teaching points

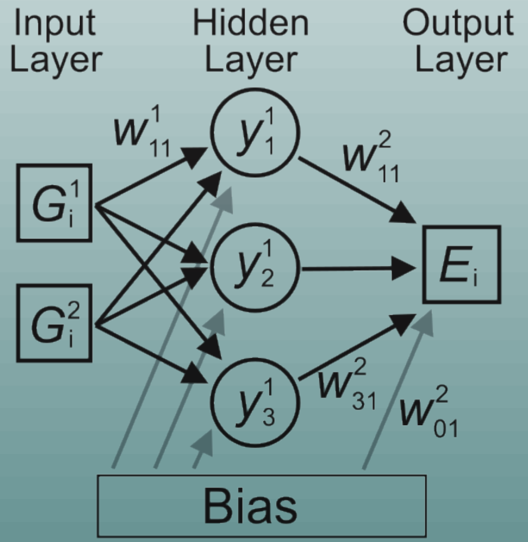
Simple Gaussian Process

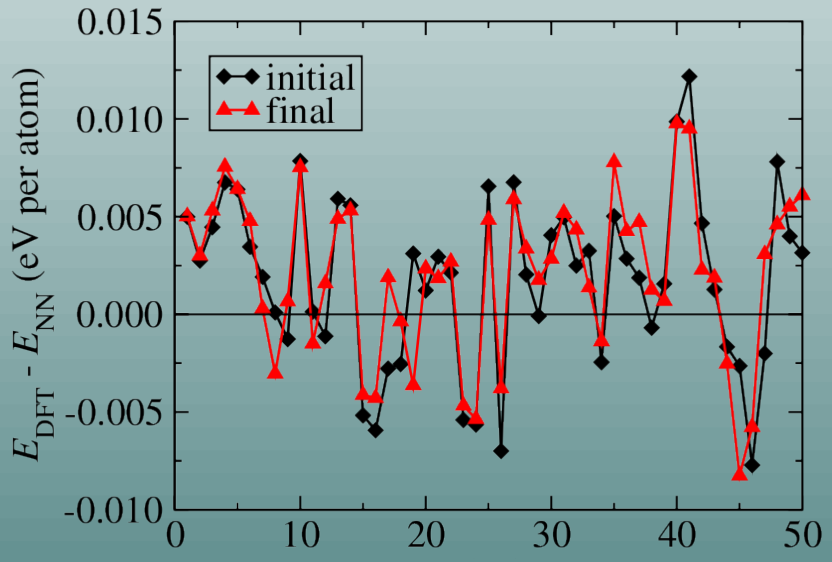


target function: sin *x*
teaching points between 3 and 7
random noise on teaching points
expectation value and variance predicted

Previous work

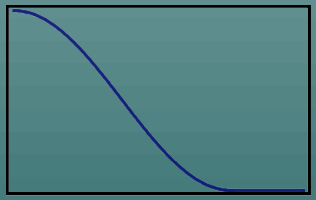
silicon potential based on quantum calculations
neural network was fitted





$$G_i^{(1)} = \sum_{j \neq i} \exp\left[-\eta(r_{ij} - r_s)^2\right] f_c(r_ij)$$

f_c



$$G_i^{(2)} = 2^{1-\zeta} \sum_{j,k \neq i} (1 + \lambda \cos\theta_{ijk})^{\zeta} \exp\left[-\eta(r_{ij}^2 + r_{ik}^2 + r_{jk}^2)\right] f_c(r_{ij}) f_c(r_{ik}) f_c(r_{jk})$$

Previous work

high dimensional interpolation with neural network
problems:

- it is not explicit interpolation
- no variance
- no accuracy
- difficult to extend

Invariants

we need to consider symmetries:

- translational
- rotational
- permutational

$x_1, y_1, z_1, \ldots, x_N, y_N, z_N$ do not represent these symmetries

$R_{ij} = \mathbf{r}_i \cdot \mathbf{r}_j$ matrix elements ordered not suitable

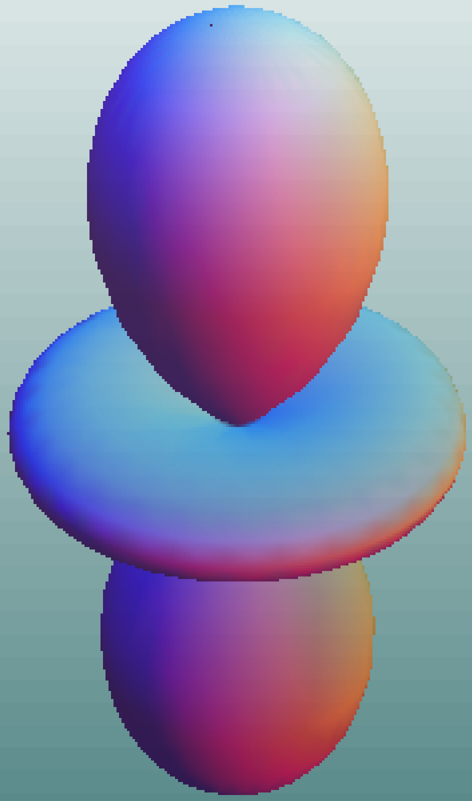
all interatomic distances ordered: no guarantee that they are complete

Invariants

$$\rho(\mathbf{r}) = \sum_{i=1}^{N} \delta(\mathbf{r} - \mathbf{r}_i)$$

$$(x, y, z) \rightarrow (r, \theta, \phi)$$

$$\rho(r, \theta, \phi) = \sum_{n=1}^{\infty} \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \alpha_{n,l}^{m} \, g_n(r) \, Y_l^m(\theta, \phi)$$



translational symmetry

permutational symmetry

Power spectrum

1D function: *f *(ω) f(ω)*

$$\hat{R}_{\mathbf{a}}(\alpha) Y_l^m = \sum_{m'=-l}^{l} D_l^{mm'}(\mathbf{a}, \alpha) Y_l^{m'}$$

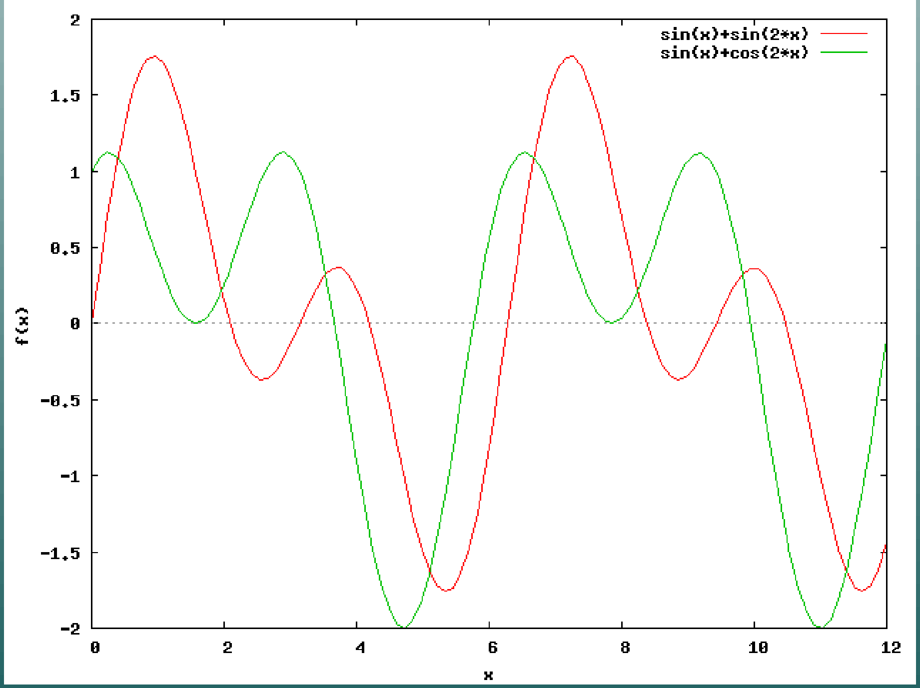
$$\mathbf{D}^{-1} = \mathbf{D}^{\dagger}$$

$$\hat{R}\,\rho(\theta, \phi) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} c_l^m \sum_{m'=-l}^{l} D_l^{mm'} Y_l^{m'}$$

$$\mathbf{c}_l \rightarrow \mathbf{D}_l \mathbf{c}_l$$

$$p_l = \mathbf{c}_l^{\dagger} \mathbf{c}_l$$

$$p_l \rightarrow \mathbf{c}_l^{\dagger} \mathbf{D}_l^{\dagger} \mathbf{D}_l \mathbf{c}_l = p_l$$



poor representation
because incomplete

Bispectrum

$$\mathbf{c}_{l_1} \otimes \mathbf{c}_{l_2} \rightarrow \left(\mathbf{D}_{l_1} \otimes \mathbf{D}_{l_2}\right) \mathbf{c}_{l_1} \otimes \mathbf{c}_{l_2}$$

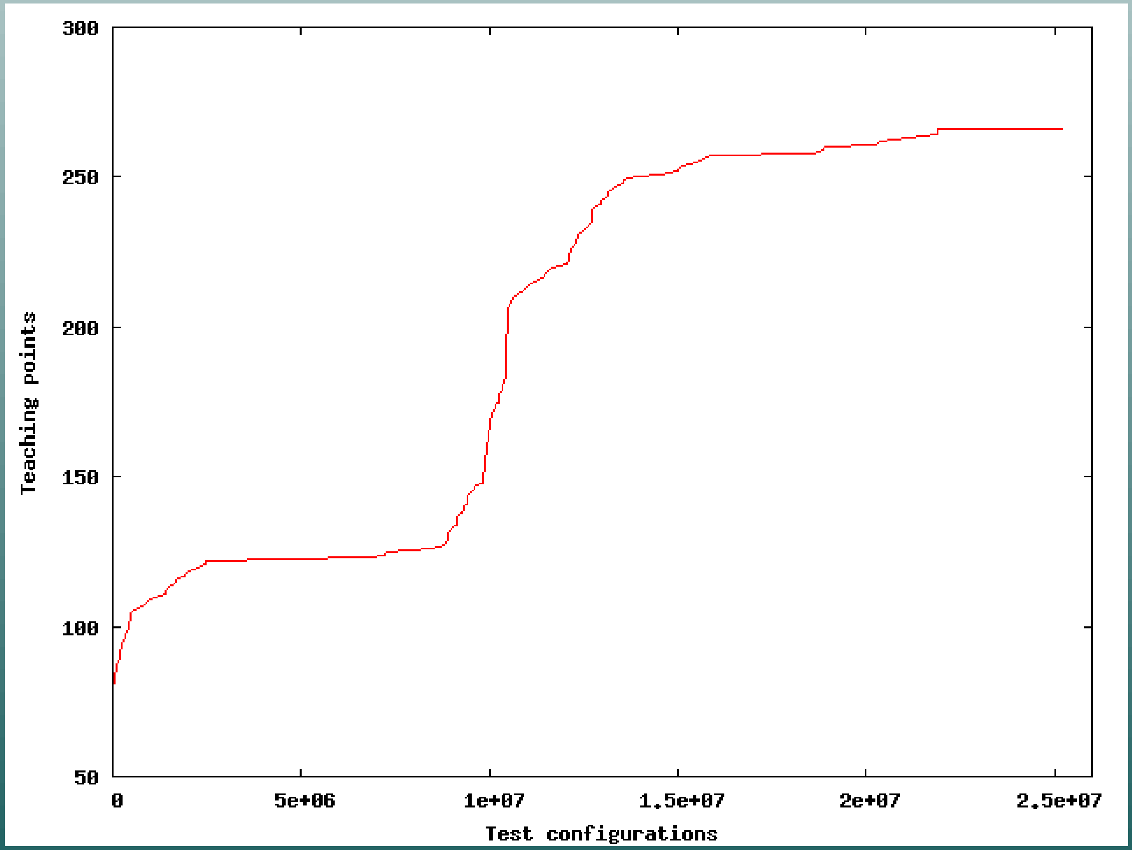
$$\mathbf{D}_{l_1} \otimes \mathbf{D}_{l_2} = \mathbf{C}^{\dagger}_{l_1,l_2} \left[\bigoplus_{l=|l_1-l_2|}^{l_1+l_2} \mathbf{D}_l \right] \mathbf{C}_{l_1,l_2}$$

$$\mathbf{C}_{l_1,l_2}\mathbf{c}_{l_1} \otimes \mathbf{c}_{l_2} \rightarrow \left[\bigoplus_{l=|l_1-l_2|}^{l_1+l_2} \mathbf{D}_l \right] \mathbf{C}_{l_1,l_2}\mathbf{c}_{l_1} \otimes \mathbf{c}_{l_2} = \bigoplus_{l=|l_1-l_2|}^{l_1+l_2} \mathbf{g}_{l_1,l_2,l}$$

$$p_{l_1,l_2,l} = \mathbf{c}_l^{\dagger} \mathbf{g}_{l_1,l_2,l}$$

First test: teaching SW potential

first neighbour shell considered
only 'interesting' part of phase space is taught
error limit: 0.5 eV
average error: 0.09 eV



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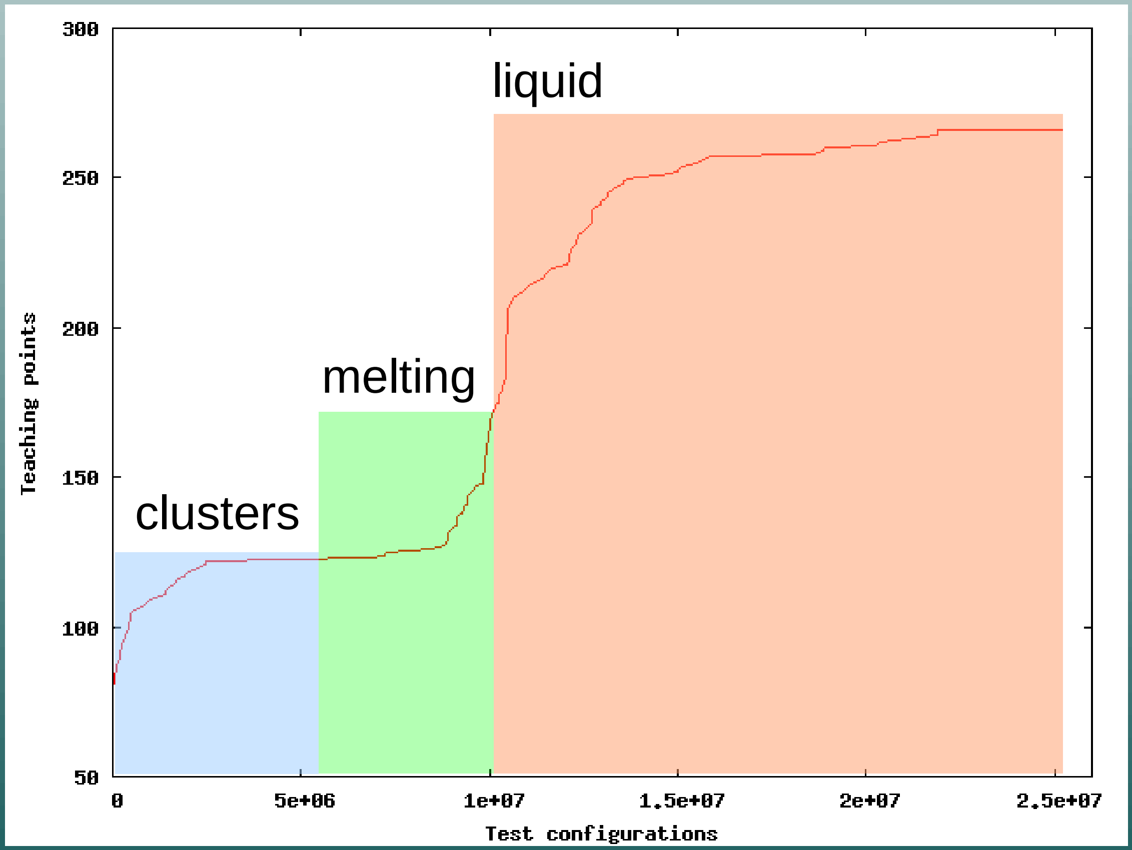
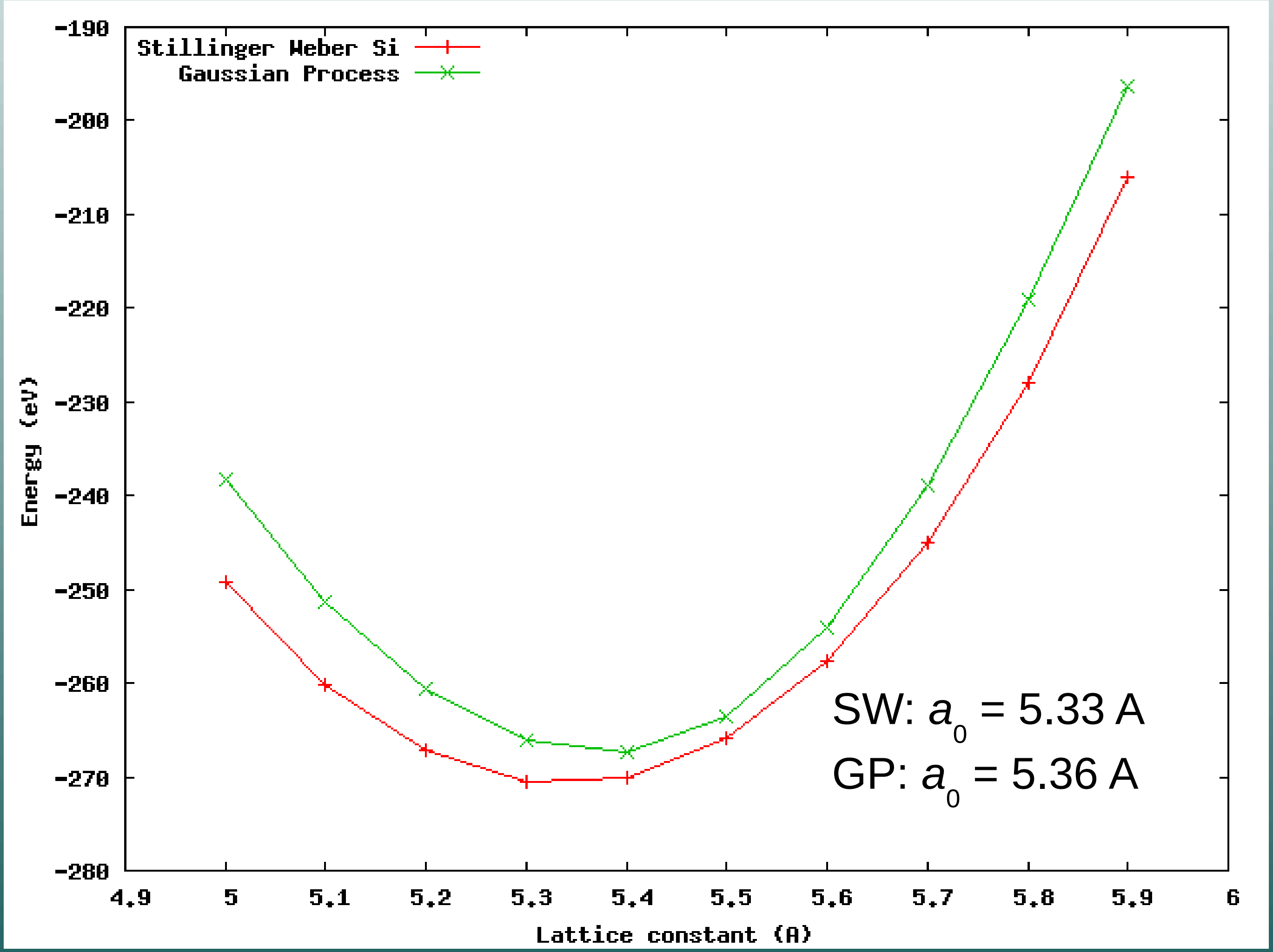


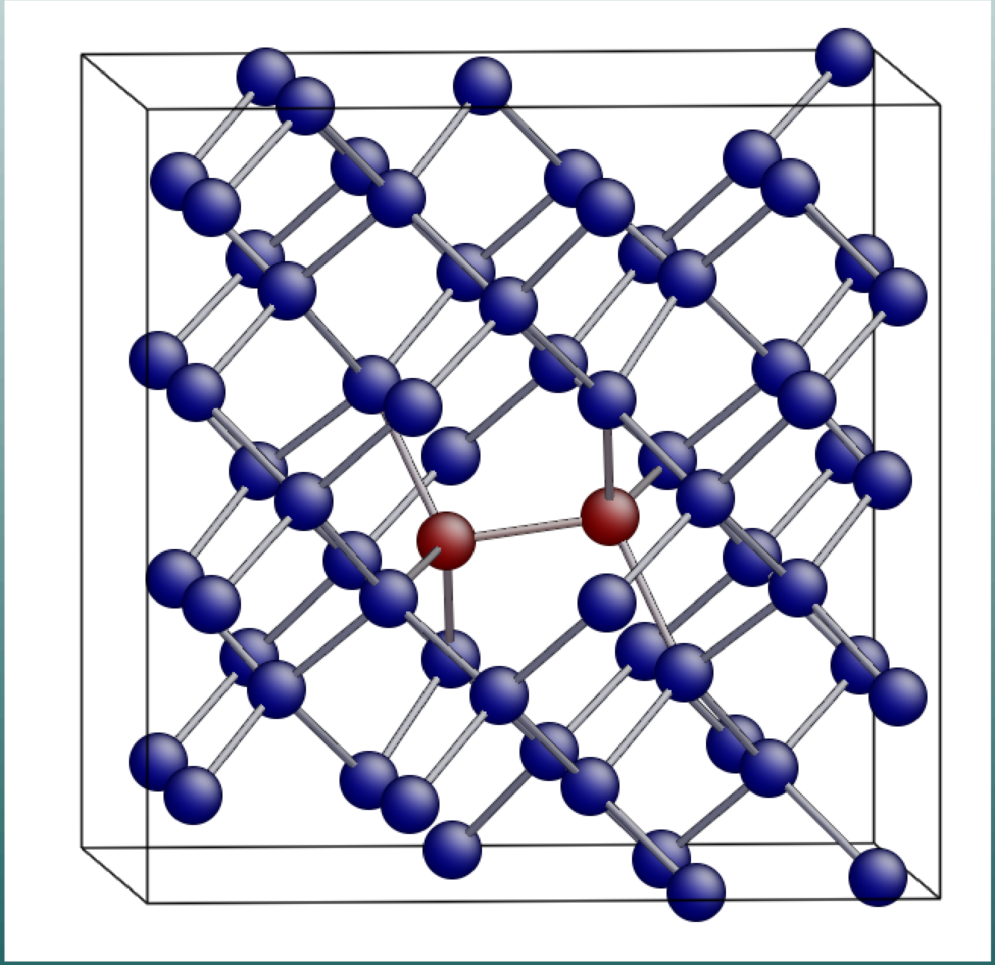
Results

Energy vs. lattice constant



Results

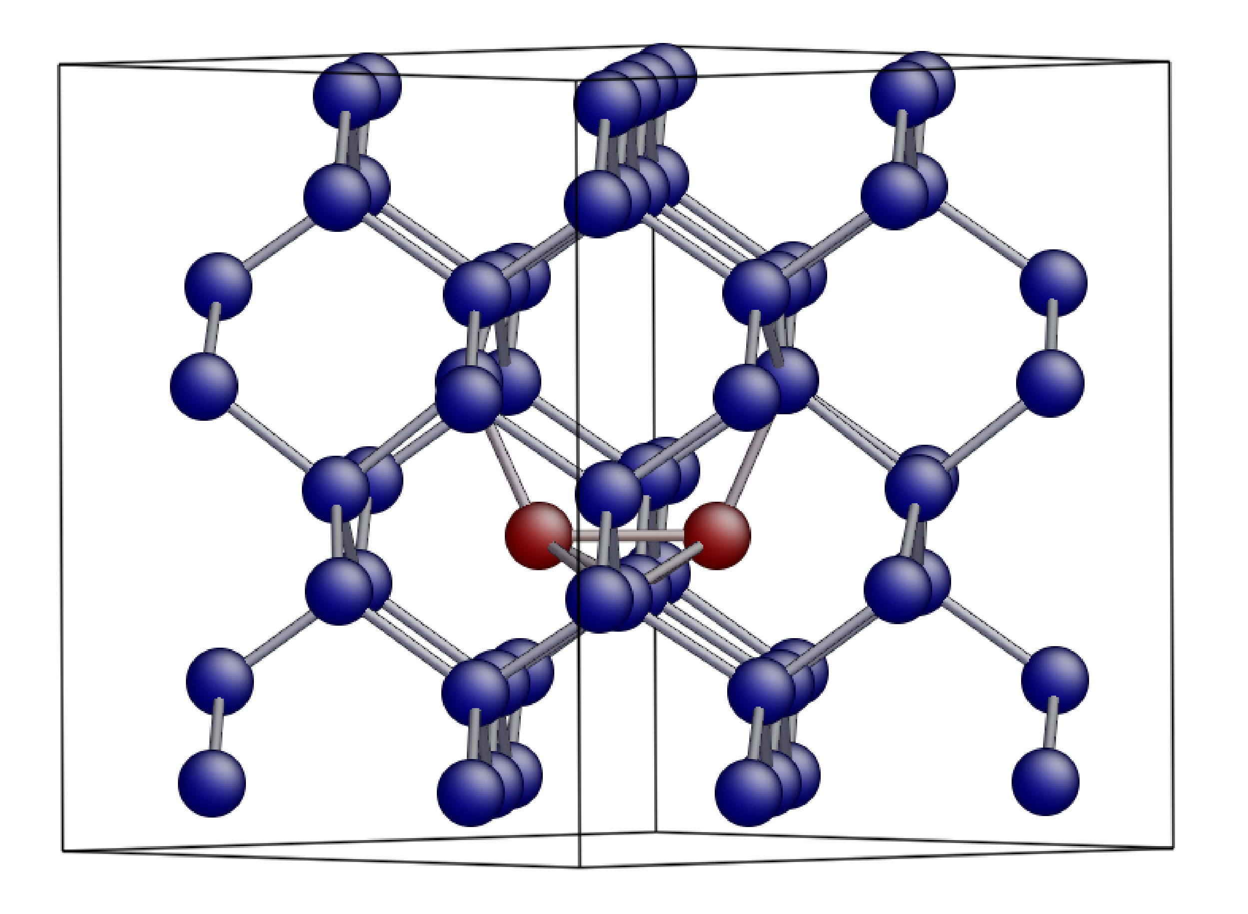
Four-fold coordinated defect



SW: 3.50 eV
GP: 2.43 eV

Results

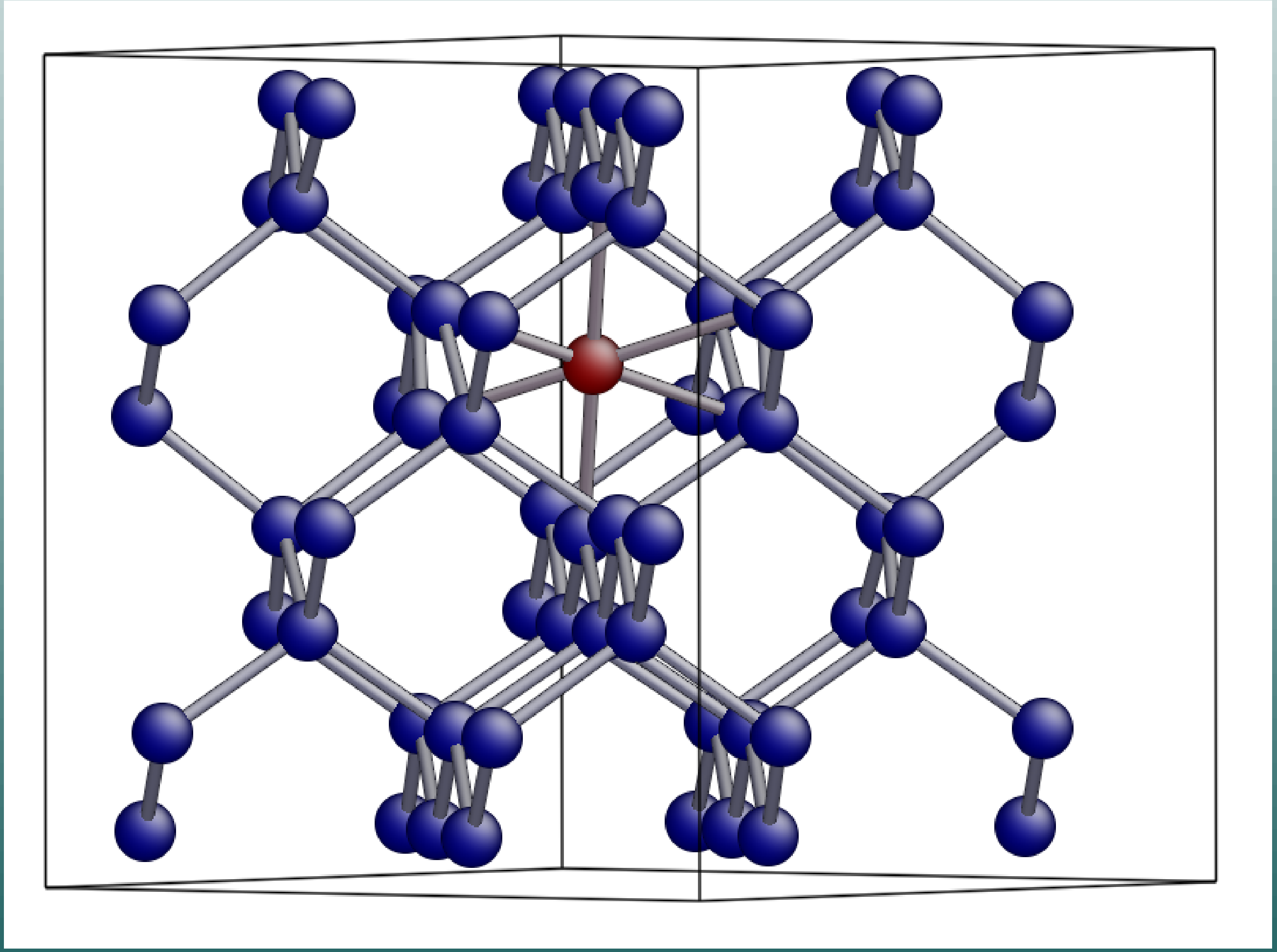
X interstitial



SW: 4.62 eV
GP: 3.98 eV

Results

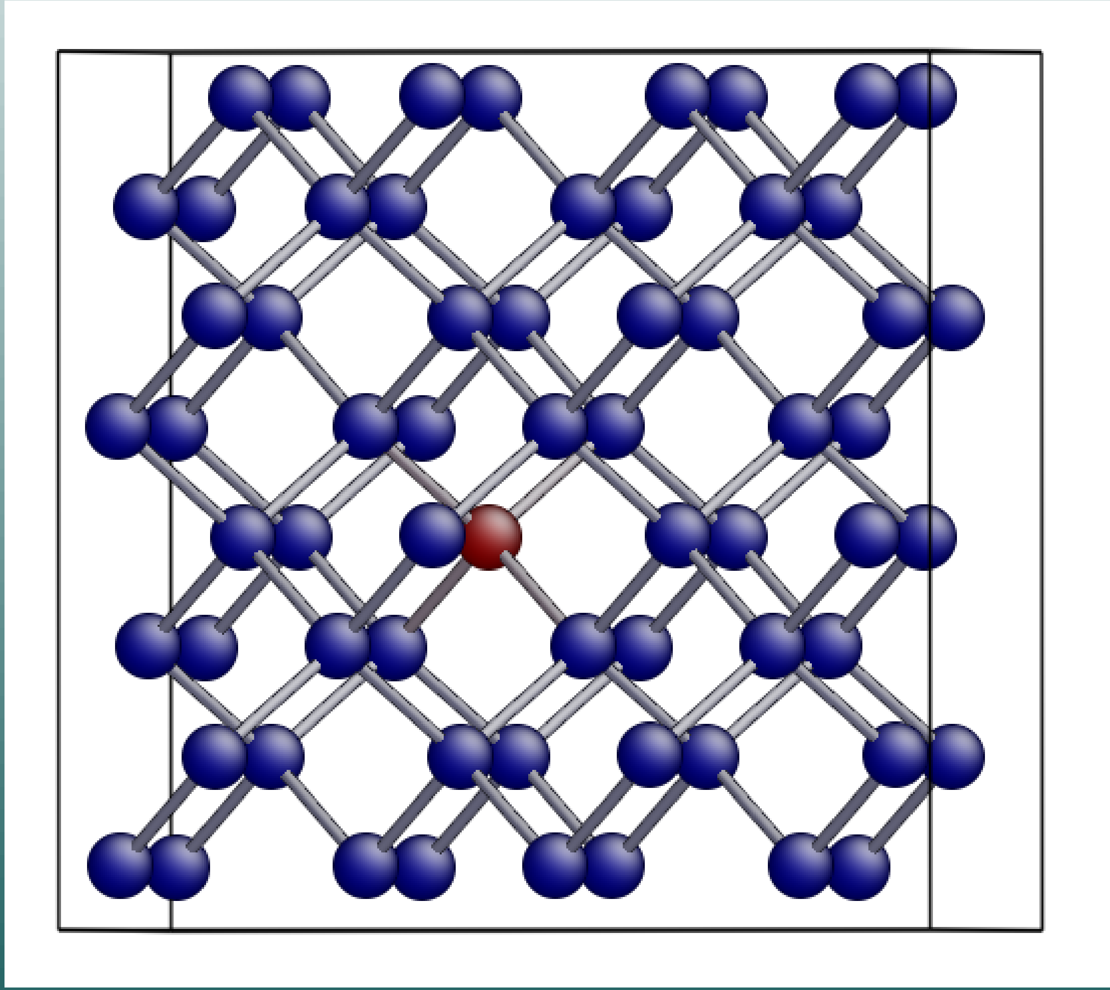
H interstitial



SW: 6.97 eV
GP: 5.89 eV

Results

Vacancy

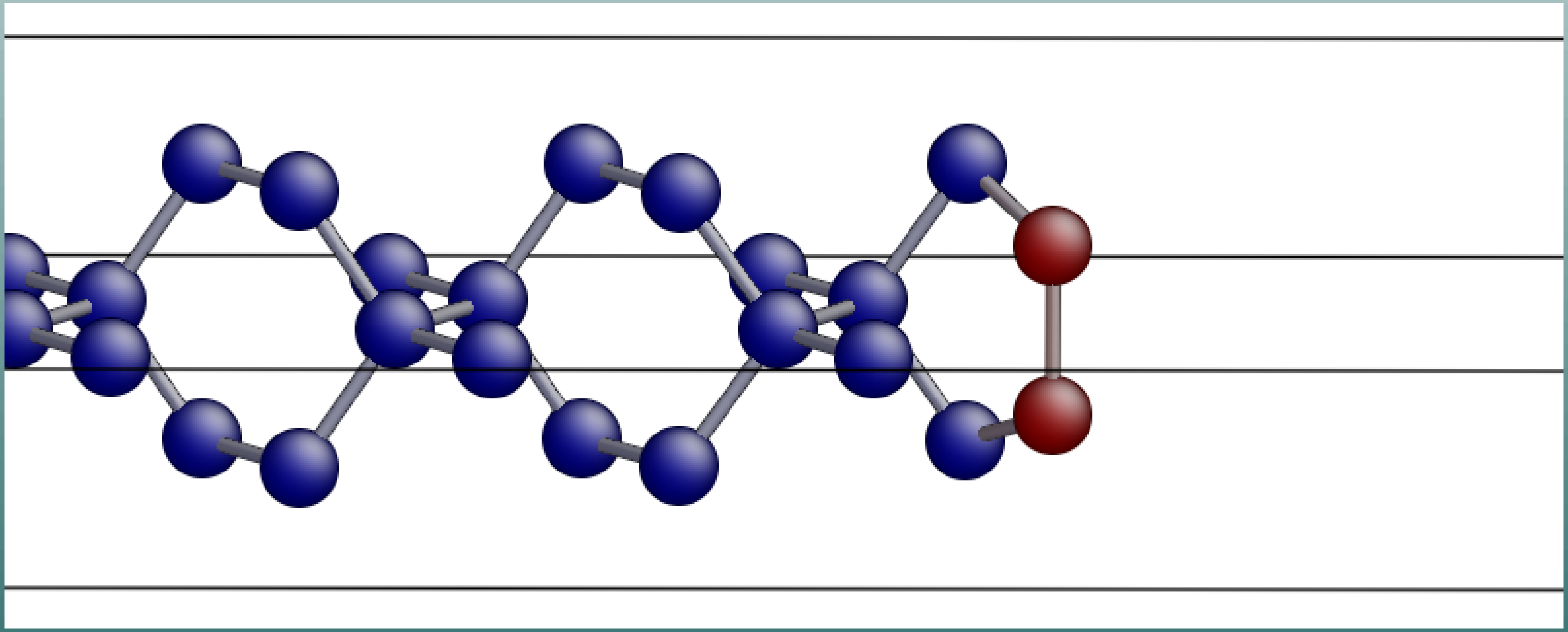


SW: 3.97 eV
GP: 3.71 eV

Results

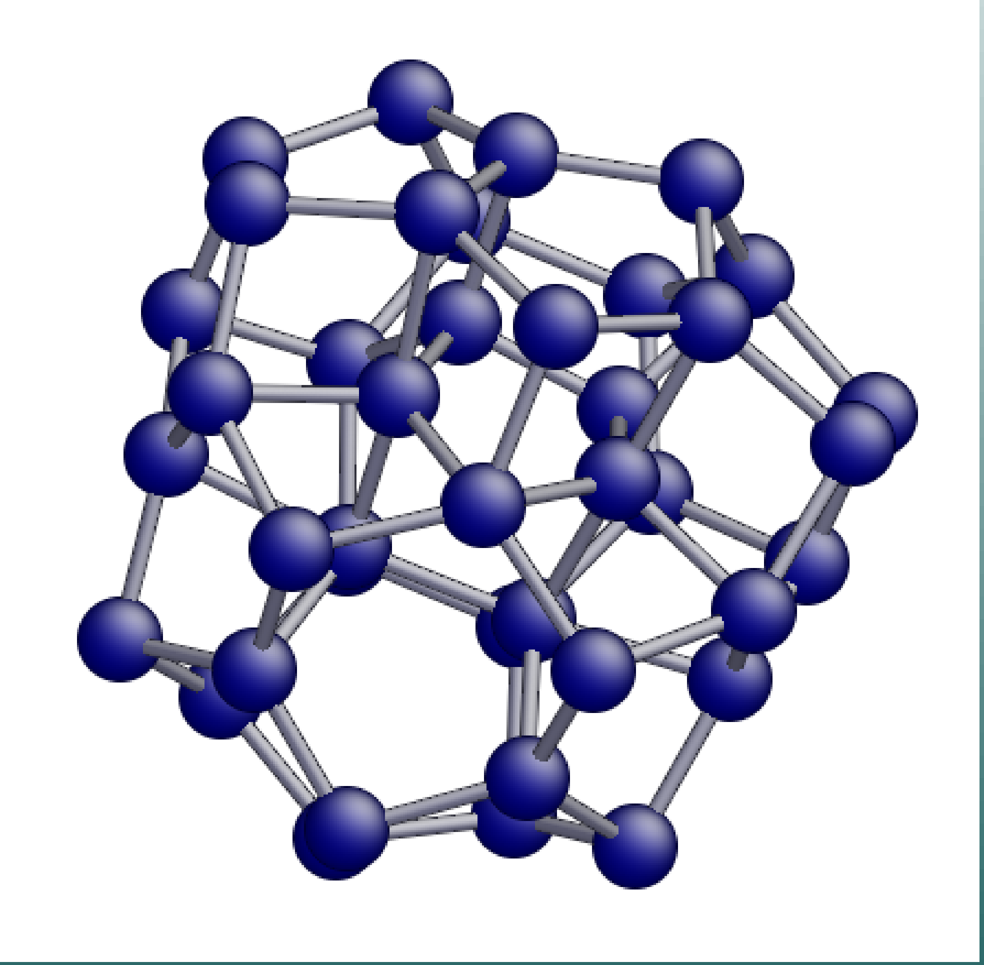
(100) surface reconstruction

SW: -1.78 eV
GP: -1.66 eV



Results

Si_{40} cluster



SW: 37.26 eV
GP: 38.95 eV

Summary

- interpolation in high dimensional space
- target potential: high level quantum calculation
- very reasonable number of teaching points
- adaptive method
- current teaching set: too few points
- aim: smaller error limit, more teaching points
- next step: TB local energies