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Active Learning of SNAP Potentials using Bayesian Uncertainty Estimation

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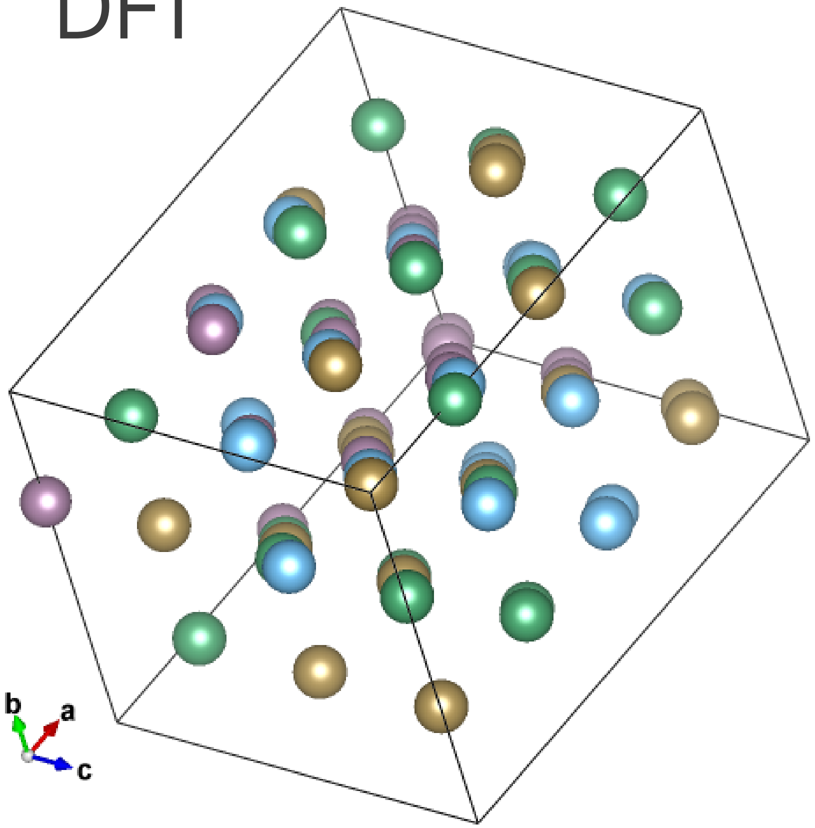
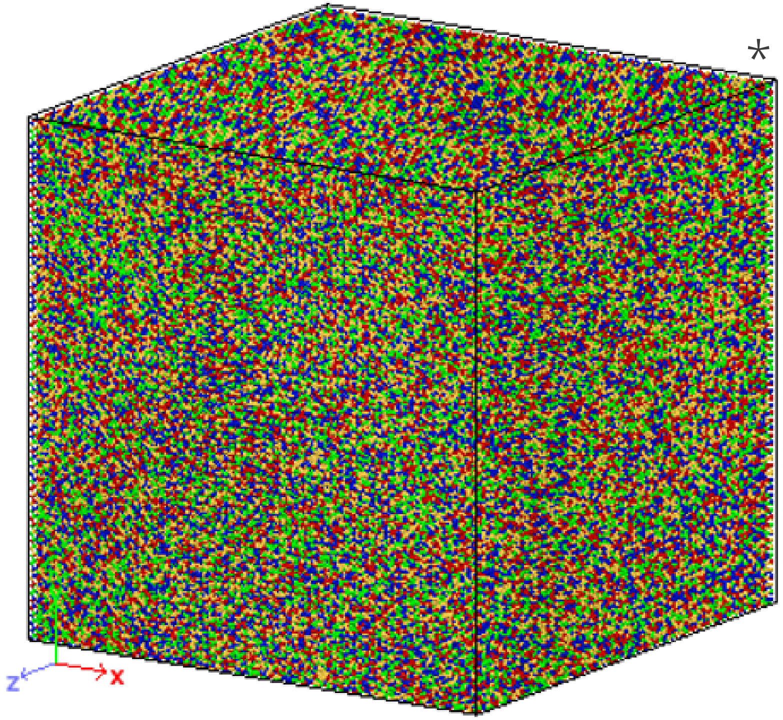
Efficient atomistic computation allows scaling

DFT



System size limit: ~1000 atoms

MD



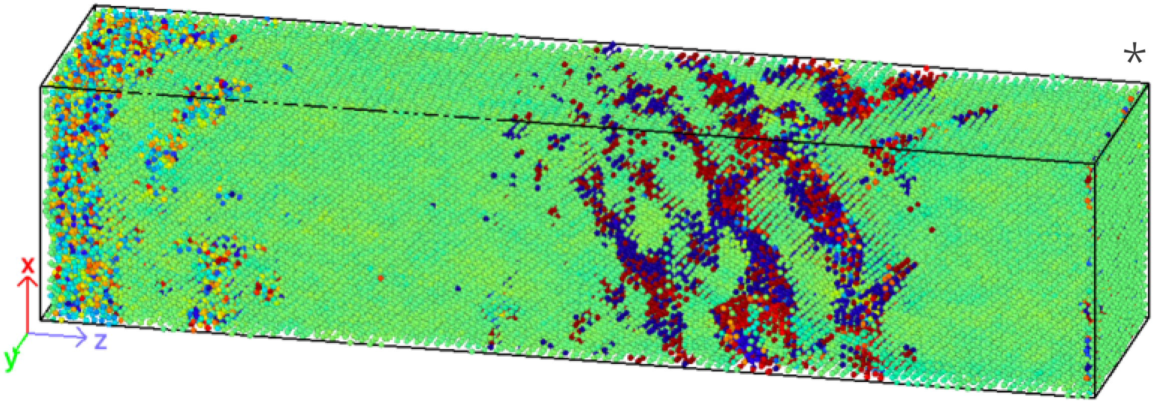
System size limit: ~1,000,000,000 atoms

* Image credit to Megan McCarthy

Uncertainty Quantification (UQ) gives reliability of results

- MD simulations are (almost) always outside the size limit of DFT
- Often studying behavior that can not be replicated at smaller scale
- Trust in the model results is required – UQ allows for building that

MD



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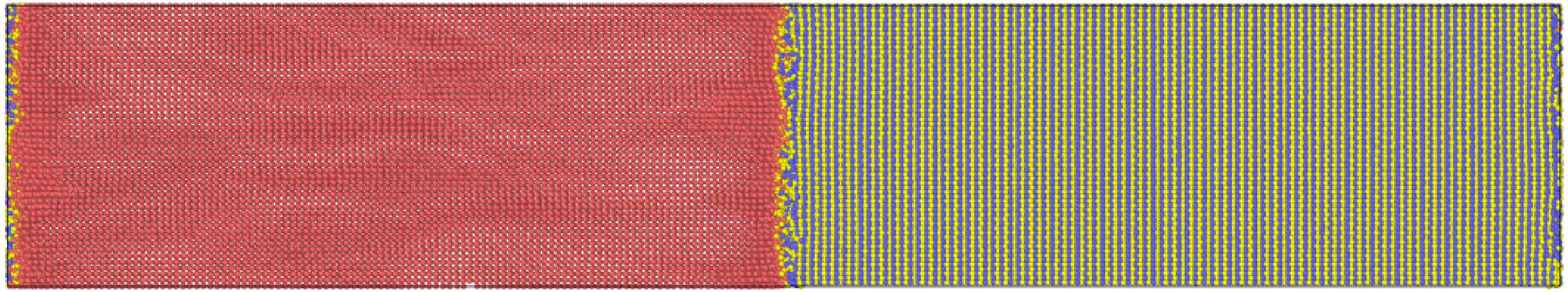


* Image credit to Megan McCarthy
** Image credit to Ember Sikorski

Bayes Rule

- For a model f(x, c) and data $y_i = y(x_i)$, calibrate the model parameters, c.

Likelihood

Prior probability

$$p(c|y) = \frac{p(y|c)\,p(c)}{p(y)} \;\longrightarrow\; p(c|y) \propto p(y|c)\,p(c)$$

Posterior probability

Evidence

Bayesian Parameter Inference

- For a model f(x, c) and data $y_i = y(x_i)$, calibrate the model parameters, c.

Likelihood

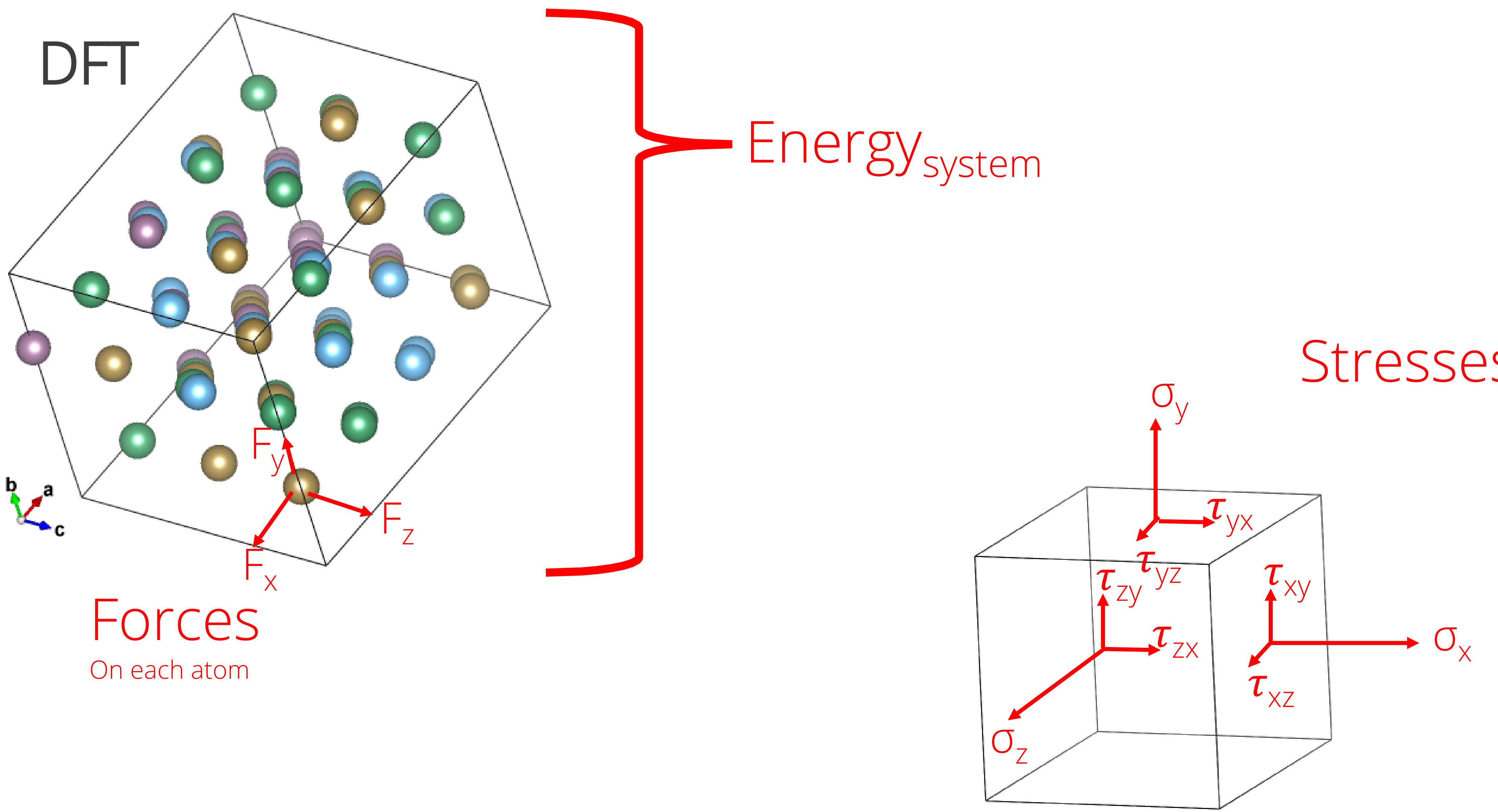
$$p(c|y) = \frac{p(y|c)p(c)}{p(y)}$$

Make some assumptions about noise: $y_i = f(x_i, c) + \sigma_i \epsilon_i, \text{ where } \epsilon_i \sim \mathcal{N}(0, 1)$

Likelihood

$$p(y|c) \propto \prod_{i=1}^{N} exp(-\frac{(f(x_i, c) - y_i)^2}{2\sigma_i^2})$$

DFT provides training data



Bispectrum – formulation of the descriptor

Density around an atom

$$\rho_i(\mathbf{r}) = \delta(\mathbf{r}) + \sum_{r_{ii'} < R_{cut}} f_c(r_{ii'}) w_{i'} \delta(\mathbf{r} - \mathbf{r}_{ii'})$$

Represent different atom types

Switching function, smooth to zero approaching R_{cut}

Modified polar coordinates

$$\theta_0 = \theta_0^{max} \frac{r}{R_{cut}}$$

Points near south pole are excluded

Switch to 4D hyperspherical harmonics

$$\rho(\mathbf{r}) = \sum_{j=0,\frac{1}{2},\ldots}^{\infty} \sum_{m=-j}^{j} \sum_{m'=-j}^{j} u^j_{m,m'} U^j_{m,m'}(\theta_0, \theta, \phi)$$

Hyperspherical harmonics basis functions

where

$$u^j_{m,m'} = U^j_{m,m'}(0,0,0) + \sum_{r_{ii'} < R_{cut}} f_c(r_{ii'}) w_i U^j_{m,m'}(\theta_0, \theta, \phi)$$

Complex valued expansion coefficients

Convert to the real valued, rotation invariant scalar triple products

$$B_{j_1,j_2,j} = \sum_{m_1,m'_1=-j_1}^{j_1} \sum_{m_2,m'_2=-j_2}^{j_2} \sum_{m,m'=-j}^{j} (u^j_{m,m'})^* H^{jmm'}_{\substack{j_1m_1m'_1 \\ j_2m_2m'_2}} u^{j_1}_{m_1,m'_1} u^{j_2}_{m_2,m'_2}$$

Bispectrum components

Coupling coefficients (constants)

($j = 0, 1/2, 1, \ldots$ and $m, m' = -j, -j+1, \ldots, j-1, j$)

Linear SNAP model for interatomic potential – single element model

$$E_{SNAP}(\mathbf{r}^N) = N\beta_0 + \boldsymbol{\beta} \cdot \sum_{i=1}^{N} \mathbf{B}^i$$

Linear coefficients to be fit

Constant energy contribution

$\mathbf{B}^i$ = (flattened) Bispectrum components
= F(atomic positions and geometry)
Defined for each atom in structure

$$\mathbf{F}^j_{SNAP} = -\nabla_j E_{SNAP} = -\boldsymbol{\beta} \cdot \sum_{i=1}^{N} \frac{\partial \mathbf{B}^i}{\partial \mathbf{r}_j}$$

$$\mathbf{W}_{SNAP} = -\sum_{j=1}^{N} \mathbf{r}_j \otimes \nabla_j E_{SNAP} = -\boldsymbol{\beta} \cdot \sum_{j=1}^{N} \mathbf{r}_j \otimes \sum_{i=1}^{N} \frac{\partial \mathbf{B}^i}{\partial \mathbf{r}_j}$$

Cartesian outer product

Example Descriptor Rows

Two elements (W, H). Higher 2J_max for W than for H. 55 non-zero descriptors for W, 14 non-zero descriptors for H

Energy of a W only structure = [34.50, -1.82, -2.94, 2.00, -3.32, -1.92, ... , 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, 0, ...]
Energy of a H only structure = [0, 0, 0, 0, 0, ..., 2.37, 1.77, -0.33, 1.12, -0.50, -1.87, 0, 0.87, 0, 0, 0, 2.87, 0, 0, 0, ...]
Energy of a W+H structure = [16.86, -0.98, -2.27, 4.15, -2.31, -0.46, ..., 0.53, -0.17, 0, 0.09, 0, 0, 0, -0.04, 0, 0, 0, ...]

Force follow the same zero-nonzero patterns based on what atoms surround the atom of interest

	Crystal Structure 1
1 row	$\sum_{i=1}^{N_s} \mathbf{B}^i$
3N rows	$-\sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_i^\alpha}$
6 rows	$-\sum_{j=1}^{N_s} r_j^\alpha \sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\beta}$

Model fitting – linear regression problem

Ab=y

Crystal Structure 1

1 row	$\sum_{i=1}^{N_s} \mathbf{B}^i$
3N rows	$-\sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\alpha}$
6 rows	$-\sum_{j=1}^{N_s} r_j^\alpha \sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\beta}$

Group 1

Group 2

Group 3

Group ...

Group M

$$\cdot \begin{bmatrix} \beta_0 \\ \boldsymbol{\beta} \end{bmatrix} = \begin{bmatrix} \vdots \\ E_s^{qm} - E_s^{ref} \\ \\ F_{j,\alpha}^{qm} - F_{j,\alpha}^{ref} \\ \vdots \\ W_{\alpha\beta,s}^{qm} - W_{\alpha\beta,s}^{ref} \\ \vdots \end{bmatrix}$$

Weighted linear regression

$$Ab=y \quad \Rightarrow \quad (w\odot A)b = (w\odot y)$$

$$\tilde{A}b = \tilde{\mathbf{y}}$$

Group 1	weight 1
Group 2	weight 2
Group 3	weight 3
Group ...	weight ...
Group M	weight M

FitSNAP default solver

$$L2\ loss\ function = \sum_{i=1}^{n} (y_{true} - y_{predicted})^2$$

Ab=y

$$\begin{bmatrix} \sum_{i=1}^{N_s} \mathbf{B}^i \\ -\sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\alpha} \\ -\sum_{j=1}^{N_s} r_j^\alpha \sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\beta} \end{bmatrix} \cdot \begin{bmatrix} \beta_0 \\ \boldsymbol{\beta} \end{bmatrix} = \begin{bmatrix} \vdots \\ E_s^{qm} - E_s^{ref} \\ \vdots \\ F_{j,\alpha}^{qm} - F_{j,\alpha}^{ref} \\ \vdots \\ W_{\alpha\beta,s}^{qm} - W_{\alpha\beta,s}^{ref} \\ \vdots \end{bmatrix}$$

b* = best fit solution of least-squares problem: $min(|\tilde{\boldsymbol{y}} - \tilde{A}\boldsymbol{b}|_2)$

(with a vector of group weights)

$(w \odot A)b = (w \odot y)$

$\tilde{A}b = \tilde{y}$

Trained model:

Ab*= predictions

UQ-FitSNAP = Analytical Bayesian solver

$$L2\ loss\ function = \sum_{i=1}^{n} (y_{true} - y_{predicted})^2$$

$$SNAP\ model\ fit = (\tilde{A}^T \cdot \tilde{A})^{-1} \cdot (\tilde{A}^T \cdot \tilde{\boldsymbol{y}}) = \boldsymbol{b}^*$$

(identical to standard least squares regression fit)

$$SNAP\ fit\ covariance = \frac{(\tilde{\boldsymbol{y}} - \tilde{A}\boldsymbol{b}^*)^T \cdot (\tilde{\boldsymbol{y}} - \tilde{A}\boldsymbol{b}^*)}{N - k - 2} \cdot (\tilde{A}^T \cdot \tilde{A})^{-1} = \Sigma$$

$$\boldsymbol{b} = MVN(\boldsymbol{b}^*, \Sigma)$$

Can draw samples / do statistics!

Ab=y

$$\begin{bmatrix} \sum_{i=1}^{N_s} \mathbf{B}^i \\ -\sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\alpha} \\ -\sum_{j=1}^{N_s} r_j^\alpha \sum_{i=1}^{N_s} \frac{\partial \mathbf{B}^i}{\partial r_j^\beta} \end{bmatrix} \cdot \begin{bmatrix} \beta_0 \\ \boldsymbol{\beta} \end{bmatrix} = \begin{bmatrix} \vdots \\ E_s^{qm} - E_s^{ref} \\ \vdots \\ F_{j,\alpha}^{qm} - F_{j,\alpha}^{ref} \\ \vdots \\ W_{\alpha\beta,s}^{qm} - W_{\alpha\beta,s}^{ref} \\ \vdots \end{bmatrix}$$

(with a vector of group weights)

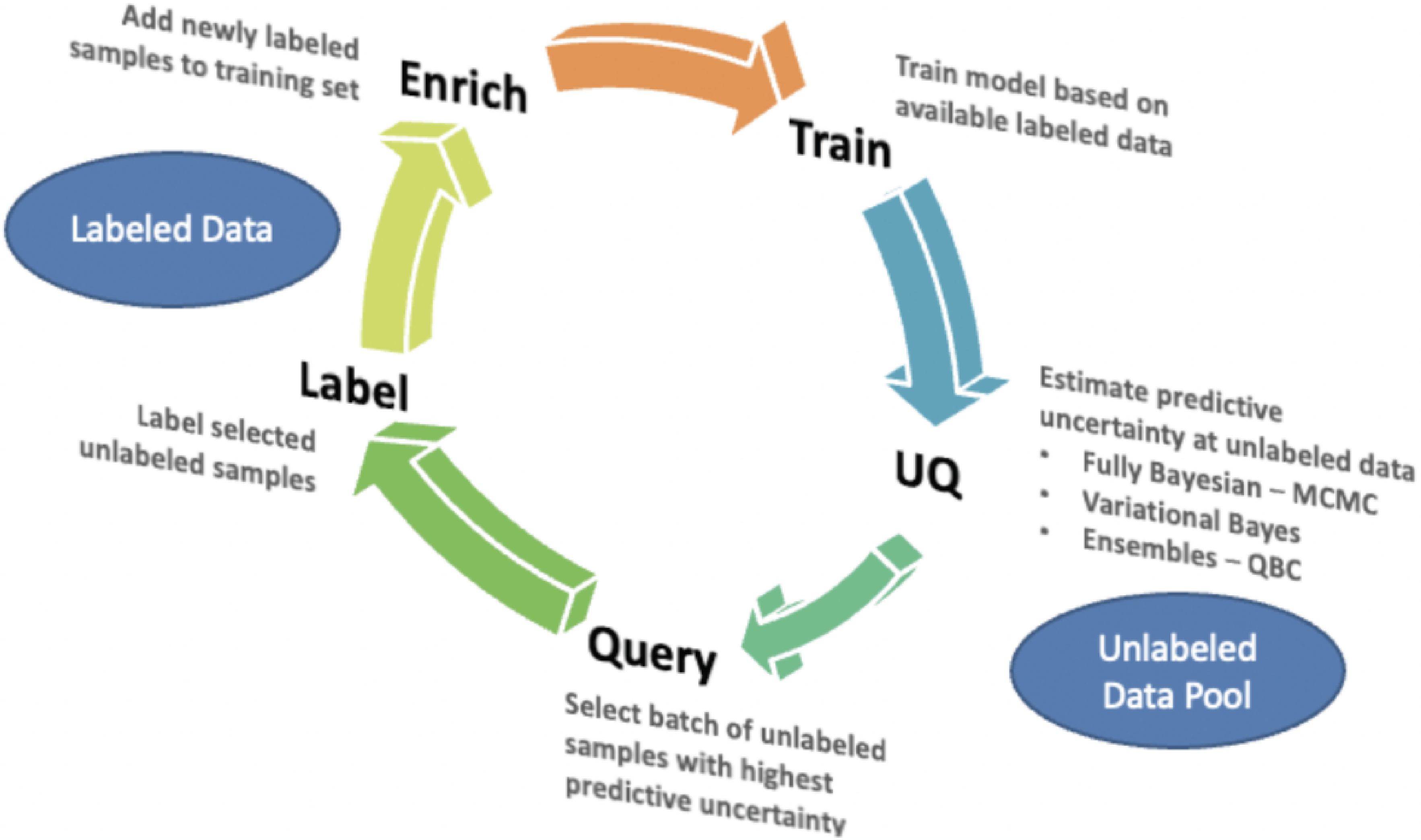
$(w \odot A)b = (w \odot y)$

$\tilde{A}b = \tilde{y}$

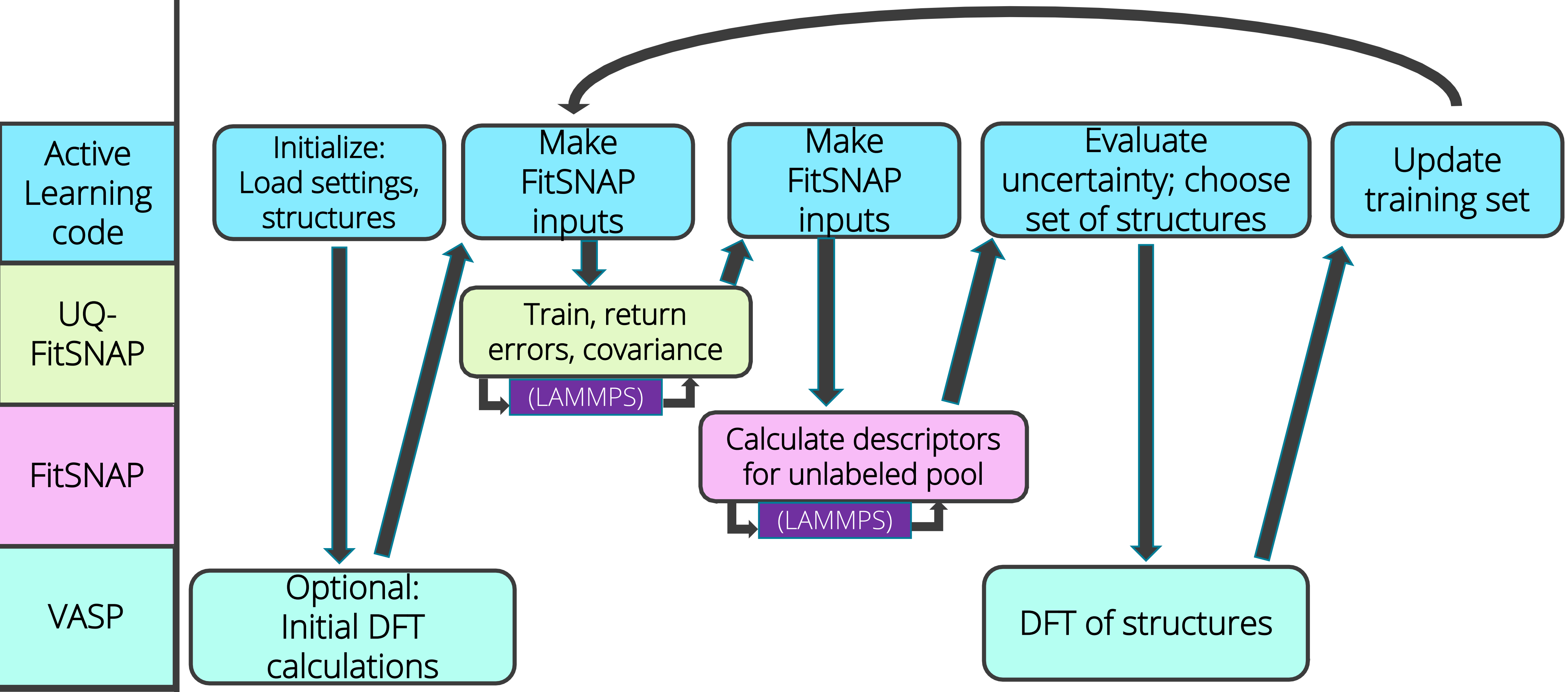
Trained model:

Ab^* = predictions $\mathrm{diag}(\mathbf{A \Sigma A^T})$ = prediction variance

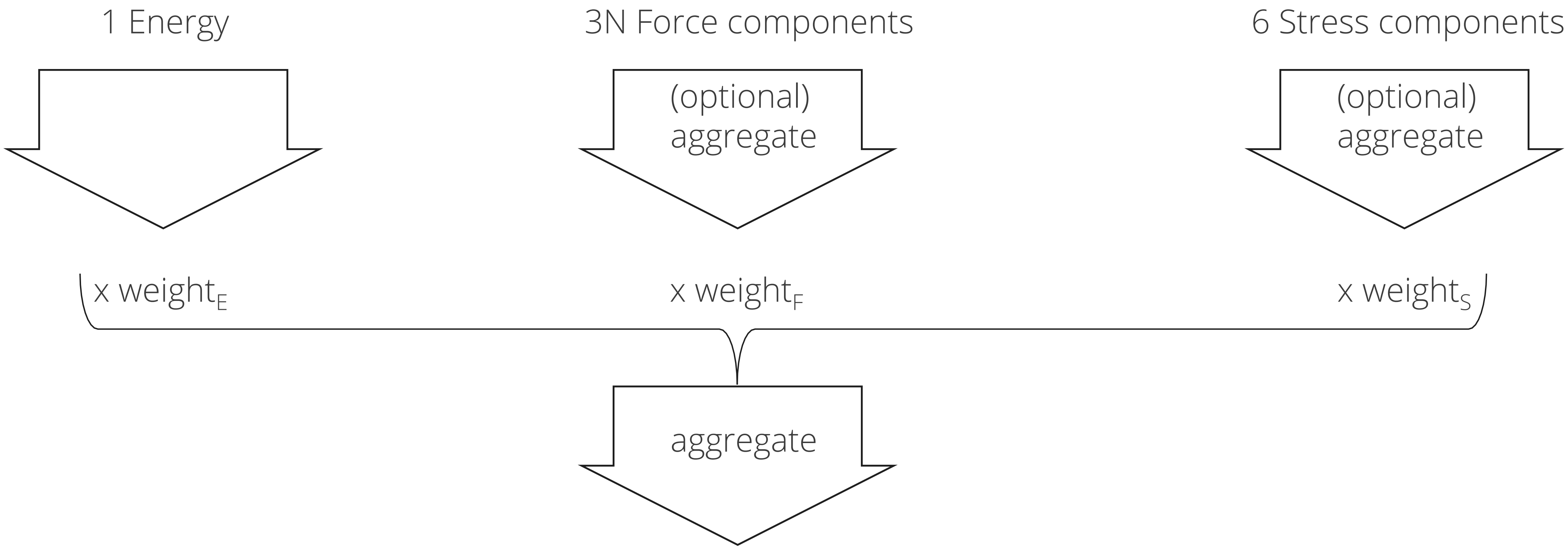
Pool based active learning – helping automate interatomic potential creation



Active Learning Code Structure



Active Learning – structure selection by prediction uncertainties



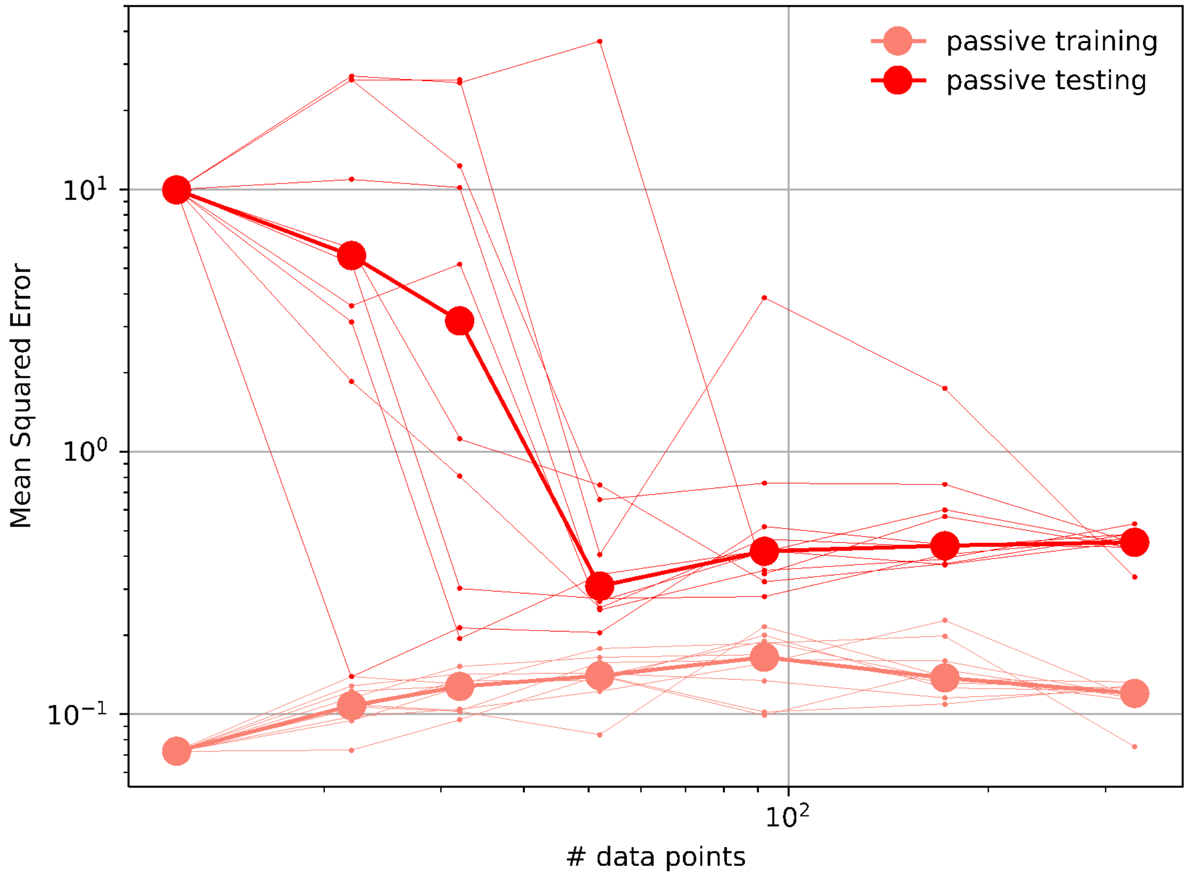
Each aggregate can be: max, mean, median, etc.

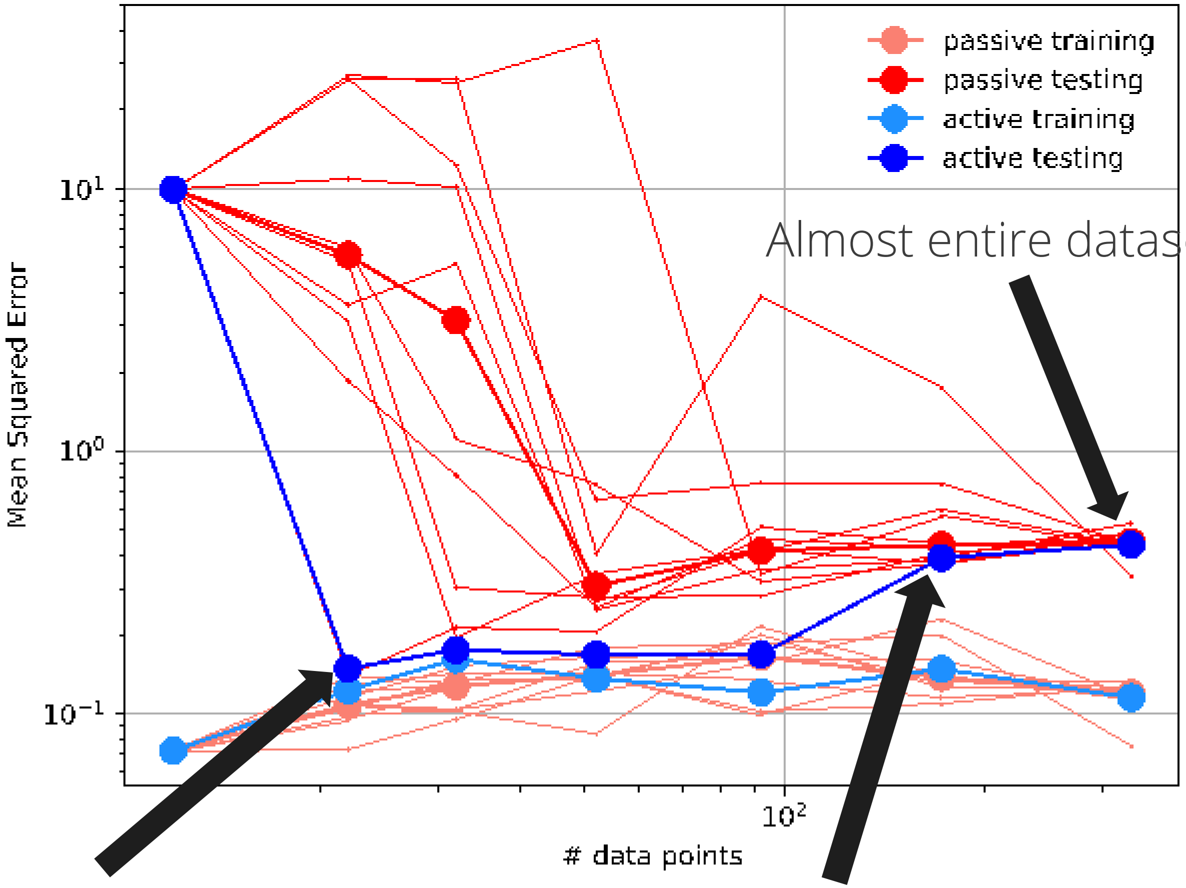
Case Studies

General settings

- Using the example datasets from FitSNAP
  - Begin with a small subset as training set
  - Test set is separated at the start, equally sampled from all defined groups, and held constant
- Use the DAKOTA pre-determined weights from the example files
- Using the ‘anl’ form for UQ approximation

Ta dataset



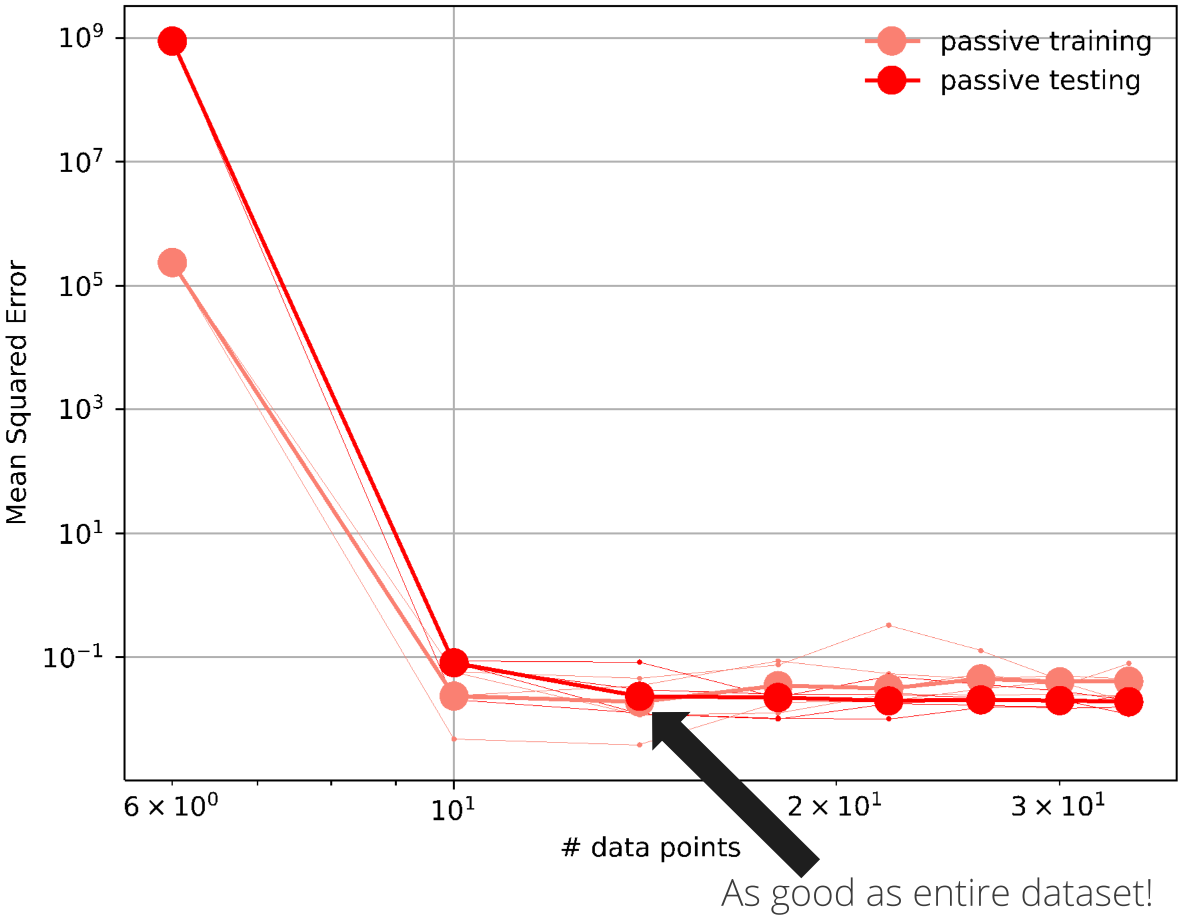


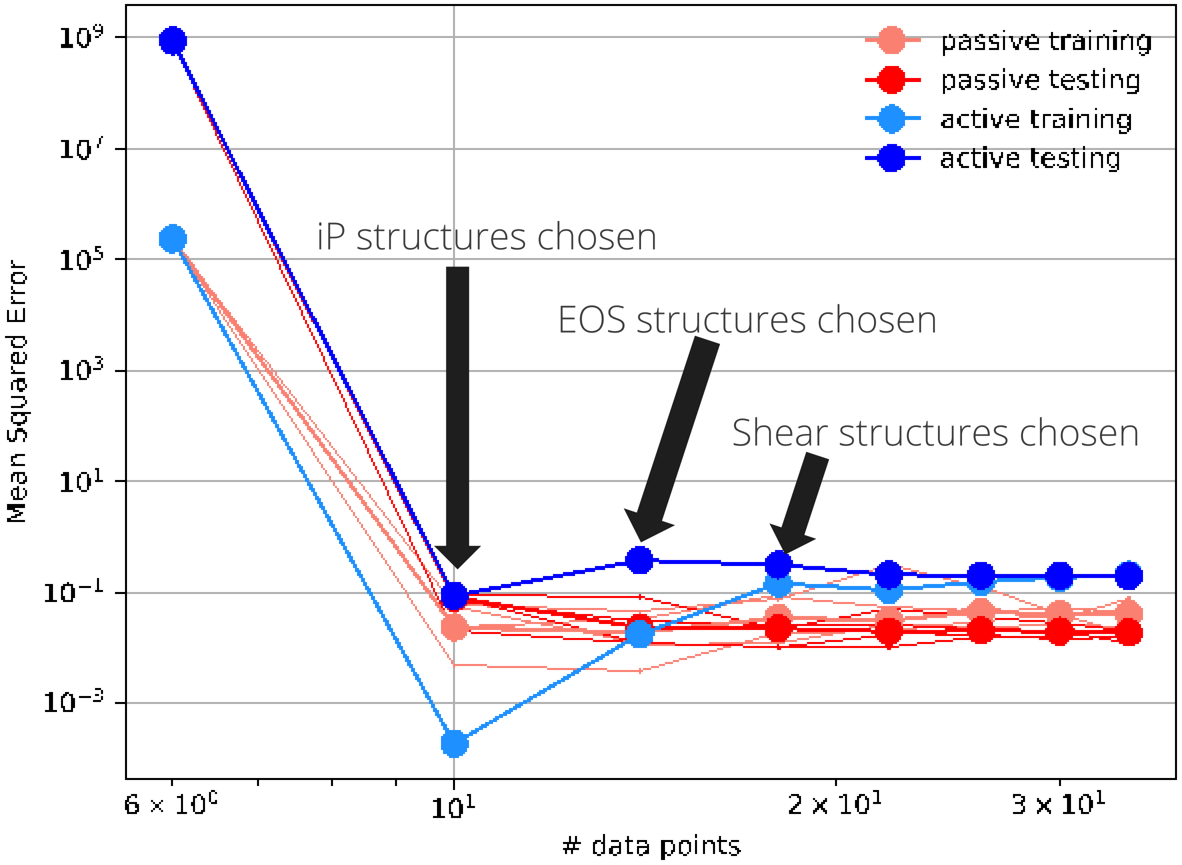
Almost entire dataset

EOS structures chosen

other structures chosen

InP dataset – minimal starting data; models plateau with very few structures

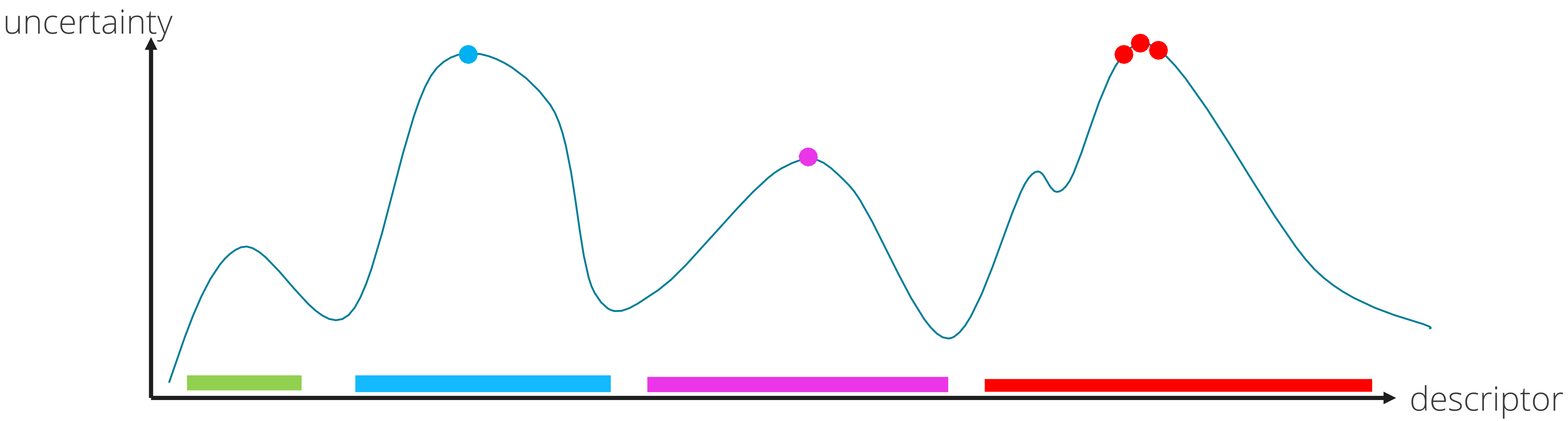




*Passive (random) mostly picks s_iP, Shear, and S_iIn due to dataset group sizes

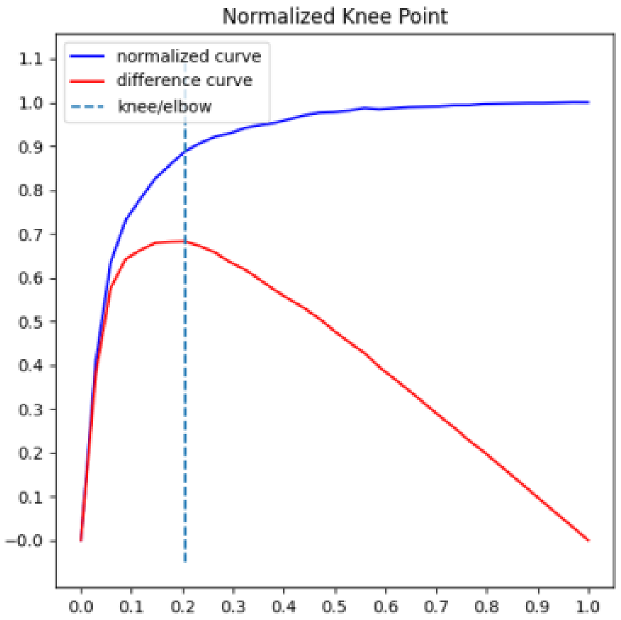
Active learning – motivation for clustering

- Minimal benefit to taking many samples that are very similar
- More efficient to get multiple samples per active learning loop
  - Want to encourage diverse selections

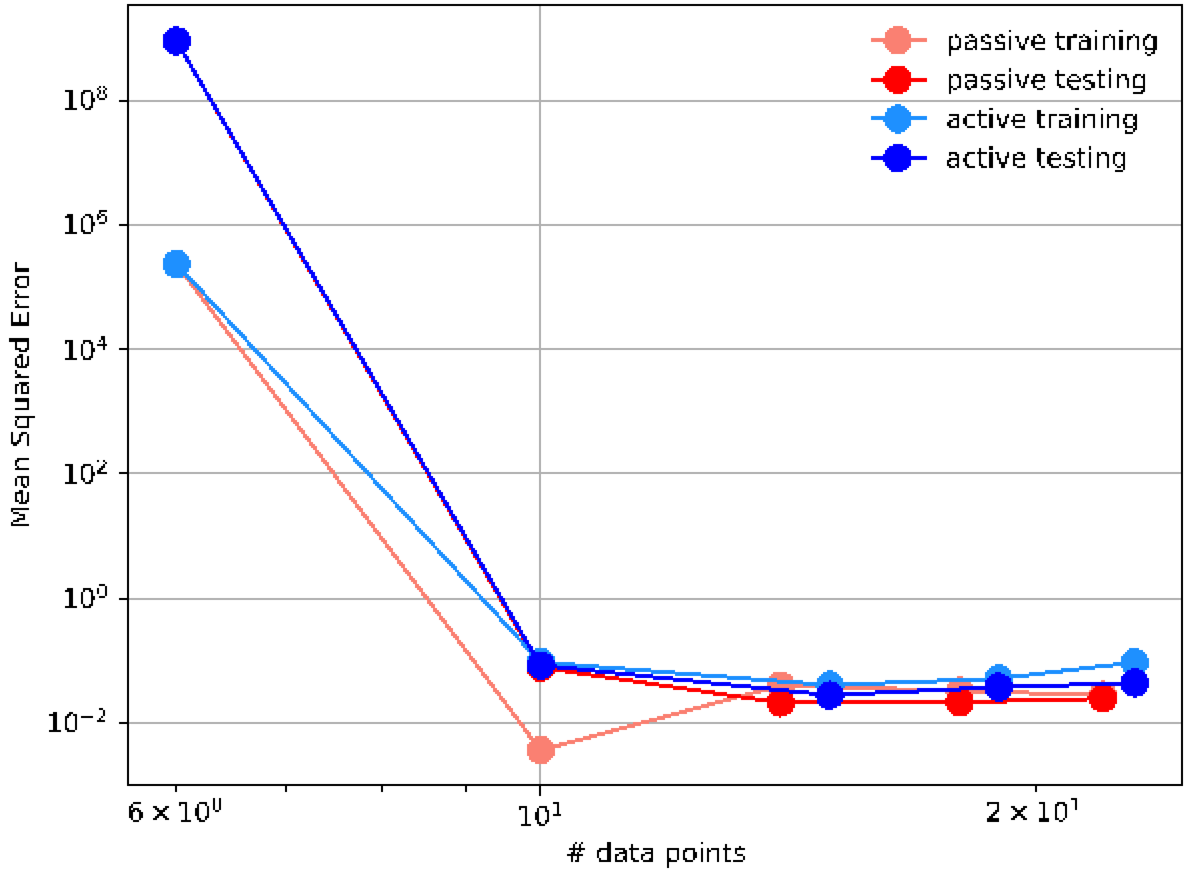


Active learning – clustering

- Kmeans clustering
  - requires assumed # of clusters
  - optimal # of clusters determined by 'knee' method
    - determine sum of squared distances from cluster centers for each # of clusters
    - select point at which the second derivative becomes negative



InP dataset – minimal starting data – clustering



Questions?

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