UQ and Model Error Estimation for Machine Learning Interatomic Potentials

Khachik Sargsyan,
Logan Williams, Habib N. Najm

Sandia National Laboratories, Livermore, CA

FMUQ Meeting
June 6, 2022

Acknowledgements

- Aidan Thompson, Mary Alice Cusentino, Mitchell Wood, Ember Sikorski (Sandia, 1400)
- DOE, Office of Science,
  - Fusion Energy Sciences (FES)
  - Advanced Scientific Computing Research (ASCR)

U.S. DEPARTMENT OF ENERGY

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.

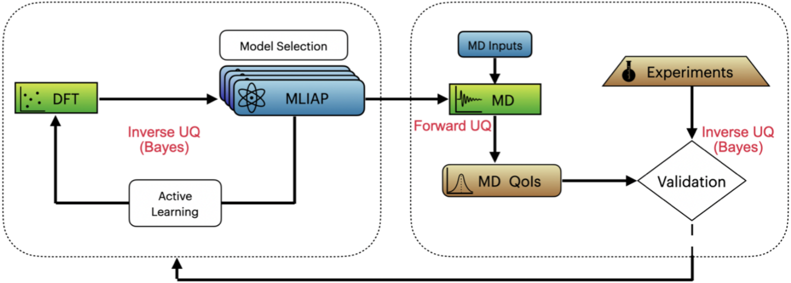
Outline

- Motivation: potential energy surface approximation
  - Machine learning for interatomic potentials (MLIAP)

- Bayesian estimation of MLIAPs
  - Linear regression models: Spectral Neighbor Analysis Potential (SNAP)
  - Importance of noise model, model error estimation
  - Complex (aka NN) models: UQ options, work in progress

- Active learning
  - What do we want from prediction uncertainties
  - How to measure ‘extrapolation’ - lack of training data

Overall Workflow (today's focus on the left box)



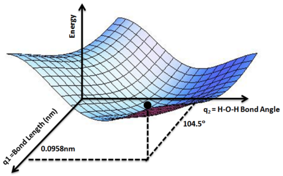
Target: Potential energy surface (PES) approximation

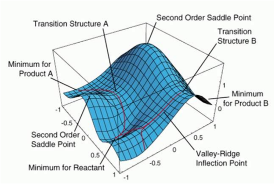
$$E = f(x)$$

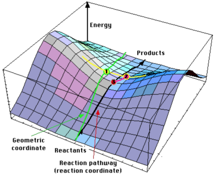
x represents coordinates/descriptors

E is energy

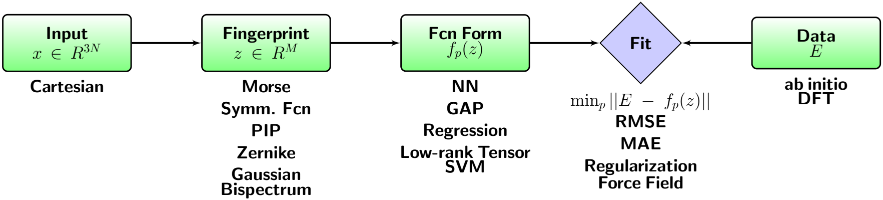
- Accurate and fast surrogates for PES to replace quantum mechanical computations for studies requiring many PES inquiries
  - saddle point search, transition paths, barrier heights
  - rapid assessment of reaction characteristics
  - automate the discovery of reactive pathways







ML Interatomic Potentials (MLIAP): supervised ML



Input $x \in R^{3N}$ → **Fingerprint** $z \in R^{M}$ → **Fcn Form** $f_p(z)$ → **Fit** ← **Data** E

Cartesian

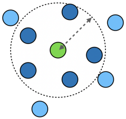
Morse
Symm. Fcn
PIP
Zernike
Gaussian
Bispectrum

NN
GAP
Regression
Low-rank Tensor
SVM

$\min_p ||E - f_p(z)||$
RMSE
MAE
Regularization
Force Field

ab initio
DFT

- Partition the interatomic interaction energy into individual contributions of the atoms $E_{\text{total}} = \sum_{i=1}^{N} E_i$
- Assume flexible functional forms of each such contribution
  - Function of positions of the neighboring atoms
  - $O(100)$ parameters



- Require the energy, forces and/or stresses predicted by a MLIAP to be close to those obtained by a quantum mechanical model on some atomic configurations (a.k.a. training set)

MLIAP - desired features

- Good input descriptors
- Accurate, fast-to-evaluate, analytic derivatives
- High-dimensional, flexible functional form
- Transferable/generalizable to unseen atomic configurations
- Account for physics:
  - invariant with respect to translation,rotation, and reflection of the space, and also permutation of chemically equivalent atoms
- Locality (depend on surrounding atoms only within a finite cut-off radius), but remain smooth with respect to atoms entering and leaving the local neighborhood
- **Equipped with uncertainty estimate**
  - for active learning, for MD propagation, ...

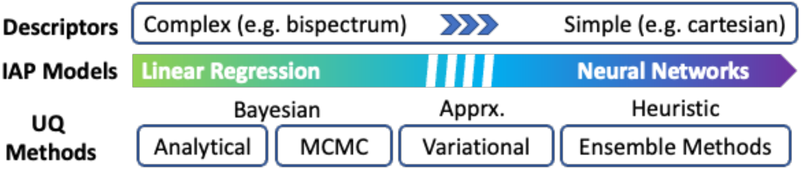
ML for PES, (growing) literature:

uncertainty estimation is largely lacking

- Weighted interpolation [Ischtwan 1994; Dowes, 2007-09; Maisuradze, 2009]
- Permutationally invariant polynomials [Xie, 2010]
- Gaussian processes [Bartok, Csanyi 2010-15; Mills, 2012; Rupp, 2013; Cui, 2016; Uteva, 2017; Guan, 2018; Schmitz, 2018]
- Low-rank tensor expansions [Jackle, 1996; Baranov, 2015; Rai, 2017, 2018]
- Support vector machines, kernel regression [Le, 2009; Balabin, 2011; Dral, 2017]
- Neural networks (NN) [Blank, 1995; Tai No, 1997; Prudente, 1998; Lorenz, 2004; Witkoskie, 2005; Manzhos, 2006-09; Malshe, 2008; Le, 2009] [Behler, 2010-16; Handley, 2010, 2014; Jiang, 2013; Li, 2013; Dolgirev, 2016; Khorshidi, 2016; Peterson, 2016; Carr, 2016; Kolb, 2016; Shao, 2016; Chmiela, 2017; Cubuk, 2017; McGibbon, 2017; Smith, 2017; Schutt, 2017; Yao, 2017; Hajinazar, 2017; Bereau, 2018; Lubbers, 2018; Unke, 2018; Wang, 2018; Natarajan, 2018; Zhang, 2018; Onat, 2018]

Enabling parametric fits with uncertainties

$$y \approx f_c(x)$$



Descriptors Complex (e.g. bispectrum) ⟫⟫ Simple (e.g. cartesian)

IAP Models Linear Regression → Neural Networks

UQ Methods

Bayesian: Analytical, MCMC

Apprx.: Variational

Heuristic: Ensemble Methods

Uncertainty estimation options

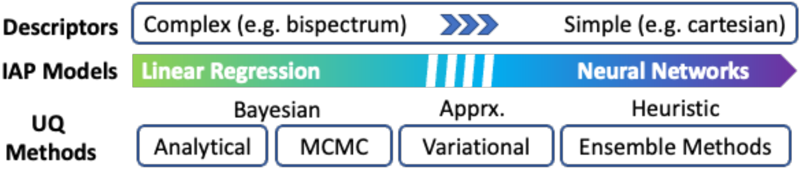
$$y \approx f_c(x)$$

- Bayesian inference: $\overbrace{P(c|y)}^{\text{Posterior}} \propto \overbrace{P(y|c)}^{\text{Likelihood}} \overbrace{P(c)}^{\text{Prior}}$
    - Markov chain Monte Carlo sampling of posterior PDF

- Variational methods: $c \sim N(\mu, \Sigma)$ and optimize μ, Σ.
    - Largely, this is also called Bayesian Neural Networks
    - Stochastic gradient descent to minimize evidence lower bound

- Ensemble methods: many flavors.
    - Deep ensembles
    - Query-by-committee
    - Boosting/bagging

Focus on SNAP (Left end of the figure)



Descriptors: Complex (e.g. bispectrum) »» Simple (e.g. cartesian)

IAP Models: Linear Regression → Neural Networks

UQ Methods: Bayesian (Analytical, MCMC) | Apprx. (Variational) | Heuristic (Ensemble Methods)

- [Thompson et al., 2015] “Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials”, *J Comp Phys*, 2015.

$$f(q)=\sum_{k=0}^{K} c_k B_k(q)$$

- Linear expansion in parameters c.
- Bayesian inference: both MCMC and analytical posterior PDFs are feasible

(Bayesian) Parameter Inference

- Given a model $f(x,c)$ and data $y_i = y(x_i)$, calibrate parameters c.
  - Linear model $y \approx Ac$ with coefficients c
  - NN model $y \approx NN_c(x)$ with weights/biases c

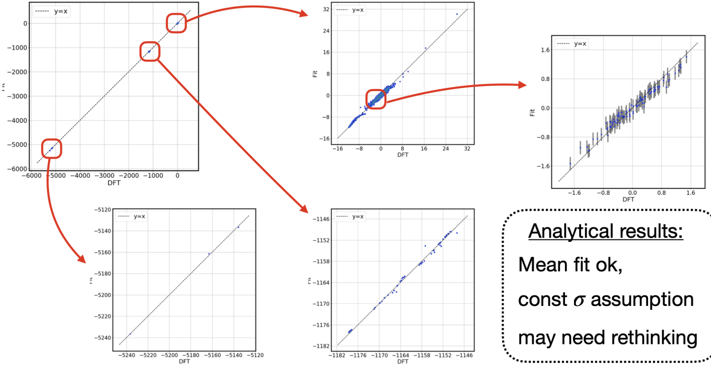
- Bayesian least-squares fit:
  $p(c|y) \propto p(y|c)p(c) \propto \prod_{i=1}^N \exp\left(-\frac{(f(x_i,c)-y_i)^2}{2\sigma_i^2}\right)$

- ... corresponding data model $y_i = f(x_i,c) + \sigma_i \underbrace{\epsilon_i}_{\mathcal{N}(0,1)}$

SNAP uncertainty with Tantalum data set

$$f(q) = \sum_{k=0}^{K} c_k B_k(q)$$

- Employed FitSNAP *https://github.com/FitSNAP/FitSNAP*
- Exact analytical Bayesian answer:
  $c \sim \mathcal{N}\left((B^T B)^{-1} B^T y, \sigma^2 (B^T B)^{-1}\right)$
  - ... if Gaussian i.i.d. likelihood is used



Analytical results:

Mean fit ok,

const σ assumption

may need rethinking

- assumptions baked in likelihood form are crucial

Elephant in the room: model is assumed to be *the* correct model behind data

$$y_i = \underset{\text{Truth}}{\overset{\text{Model}}{f(x_i, c)}} + \overset{\text{Data err.}}{\sigma_i \epsilon_i}$$

Model $\neq$ Truth

- One gets biased estimates of parameters c (crucial if the model is physical, and/or c is propagated through other models)
- More data leads to overconfident predictions (we become more and more certain about the wrong values of the data)
- More evident when there is no (observational/experimental) data error: e.g. DFT is data, and MLIAP is model

Posterior pushed-forward uncertainty does not capture true discrepancy

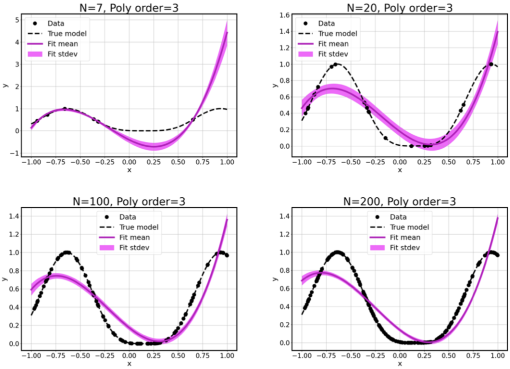
Synthetic data

$$y(x) = \sin^4(2x - 0.3)$$

Cubic fit

$$y_i \approx \sum_{k=0}^{3} c_k B_k(x)$$

More data leads to overconfident prediction



Capturing model error in data model (a.k.a. likelihood)

External correction (Kennedy-O'Hagan):

$$y_i = f(x_i, c) + \delta(x_i) + \sigma_i \epsilon_i$$

- Kennedy, O'Hagan, "Bayesian Calibration of Computer Models". *J Royal Stat Soc: Series B (Stat Meth)*, 63: 425-464, 2001.

Internal correction (embedded model error):

$$y_i = f(x_i, c + \delta(x_i)) + \sigma_i \epsilon_i$$

- Allows meaningful usage of calibrated model
- 'Leftover' noise term even with no data error
- Respects physics (not too relevant in our context)

- Sargsyan, Najm, Ghanem, "On the Statistical Calibration of Physical Models". *Int. J. Chem. Kinet.*, 47: 246-276, 2015.
- Sargsyan, Huan, Najm, "Embedded Model Error Representation for Bayesian Model Calibration". *Int. J. Uncert. Quantif.*, 9(4): 365-394, 2019.

- Typically requires uncertainty propagation in the likelihood computation
- For linear regression, we can take some shortcuts (see next)

Embedded Model Error for Linear Regression Models

Conventional (i.i.d. error term):

$$y_i \approx \sum_{k=0}^{P} c_k B_k(x) + \sigma_i \epsilon_i$$

Embed uncertainty in all or selected coefficients:

$$y_i \approx \sum_{k=0}^{P} (c_k + d_k \xi_k) B_k(x) = \overbrace{\sum_{k=0}^{P} c_k B_k(x)}^{\text{Model}} + \overbrace{\sum_{k=0}^{P} d_k B_k(x) \xi_k}^{\text{Model Error}}$$

Note:

No formal distinction between internal and external corrections, but internal allows for interpretation and model-informed error.

Embedded Model Error: Joint MCMC Inference

Conventional:

$$y_i \approx \sum_{k=0}^{P} c_k B_k(x) + \sigma_i \epsilon_i \qquad p(c|y) \propto \prod_{i=1}^{N} \exp\left(-\frac{(\sum_{k=0}^{P} c_k B_k(x_i) - y_i)^2}{2\sigma_i^2}\right)$$

Embedded:

$$y_i \approx \sum_{k=0}^{P} (c_k + d_k \xi_k) B_k(x) = \overbrace{\sum_{k=0}^{P} c_k B_k(x)}^{\text{Model}} + \overbrace{\sum_{k=0}^{P} d_k B_k(x) \xi_k}^{\text{Model Error}}$$

$$p(c, d|y) \propto \underbrace{p(y|c, d)}_{\text{Likelihood}} \underbrace{p(c, d)}_{\text{Prior}}$$

Both likelihood and prior selection are challenging.

Embedded Model Error: Two Approximate Likelihood Options

$$y_i \approx \sum_{k=0}^{P}(c_k + d_k\xi_k)\, B_k(x) \quad = \quad \sum_{k=0}^{P} c_k B_k(x) + \sum_{k=0}^{P} d_k B_k(x)\xi_k$$

Option 1: IID

$$p(c,d|y) \propto \prod_{i=1}^{N} \exp\left(-\frac{(\sum_{k=0}^{P} c_k B_k(x_i) - y_i)^2}{2\sum_{k=0}^{K} d_k^2 B_k(x_i)^2}\right)$$

Option 2: ABC

$$p(c,d|y) \propto \prod_{i=1}^{N} \exp\left(-\frac{(\sum_{k=0}^{P} c_k B_k(x_i) - y_i)^2 + (\sqrt{\sum_{k=0}^{P} d_k^2 B_k^2(x_i)} - \alpha|\sum_{k=0}^{P} c_k B_k(x_i) - y_i|)^2}{2\epsilon^2}\right)$$

Does not have to be MCMC: simply optimize the posterior for (c,d)

Pushed forward predictive uncertainty captures the true discrepancy from the data

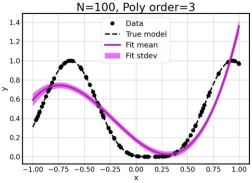
Synthetic data

$$y(x) = \sin^4(2x - 0.3)$$

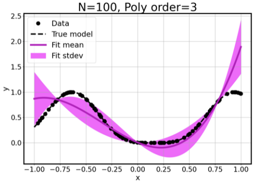
Cubic fit

$$y_i \approx \sum_{k=0}^{3} c_k B_k(x)$$

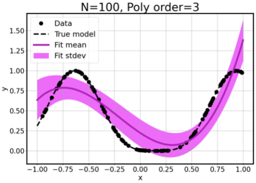
Classical case



Model error, IID likelihood

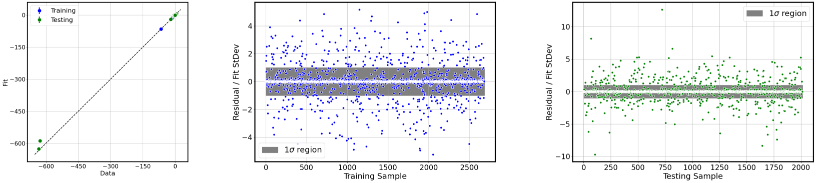


Model error, ABC likelihood

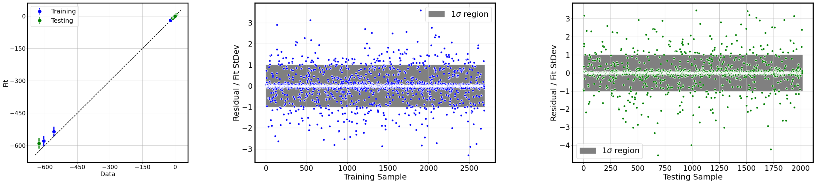


Uncertainty validation: W-ZrC Dataset

Uncertainty without model error



Uncertainty with model error



Uncertainty validation: two examples

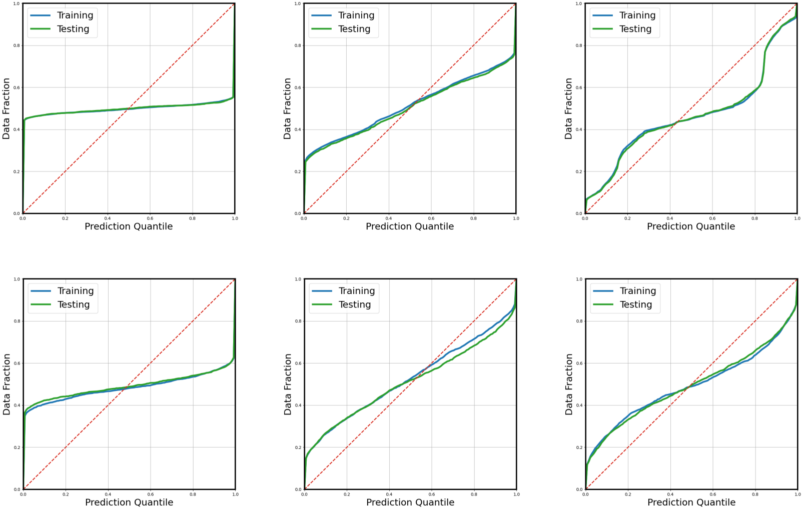
Conventional

Embedded, IID Lik.

Embedded, ABC Lik.

Ta

W-ZrC



Model Error Wrapup: several challenges and choces

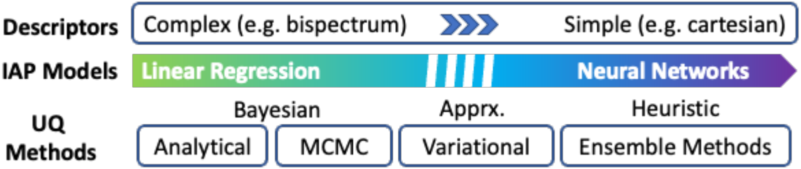
- Embedding type, e.g. additive/multiplicative

$$y_i \approx \sum_{k=0}^{P} (c_k + d_k \xi_k) B_k(x) \qquad \text{or} \qquad y_i \approx \sum_{k=0}^{P} (c_k + c_k d_k \xi_k) B_k(x)$$

- Degenerate (Gaussian) likelihoods: resort to approximate Bayesian computation (ABC) or independent (IID) assumptions
- Difficult posterior PDFs for MCMC, choice of priors for embedding parameters
- Which coefficients to embed the model error in?
- Connect predictive uncertainty and the residual error with an extrapolation metric
- Weighting between energies, forces and stresses

Enabling parametric fits with uncertainties

$$y \approx f_c(x)$$



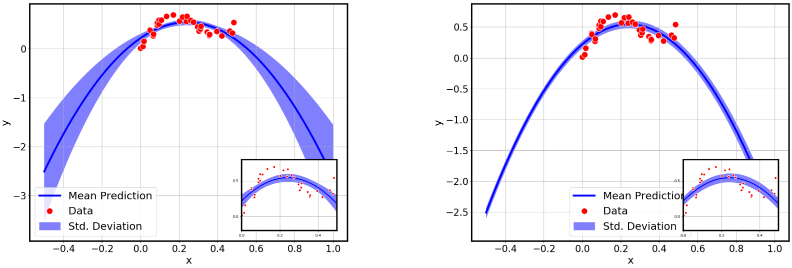
Note the connection between variational inference and embedded model error

- Variational methods: $w \sim N(\mu, \Sigma)$ and optimize μ, Σ.
  - Largely, this is also called Bayesian Neural Networks
  - Minimize evidence lower bound via SGD
- Embedded model error: $w \sim N(\mu, \Sigma)$ and optimize μ, Σ.
  - Minimize Gaussian approximation of output predictions (IID), or
  - Minimize statistics/moment matching criterion (ABC)

Next:
Toy example demonstrating issues of mean-field variational inference outside training support.

Polynomial fit: Extrapolation scenario

Order=2



True Posterior

Variational Posterior

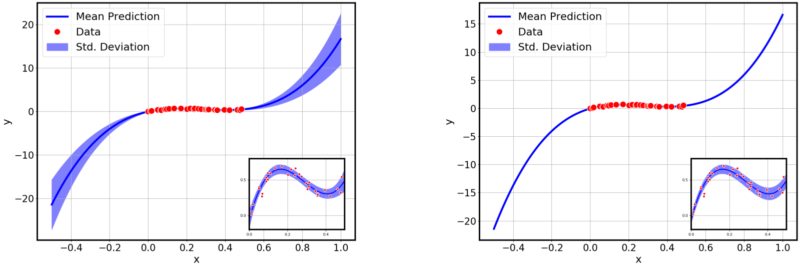
Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Extrapolation scenario

Order=3

True Posterior

Variational Posterior



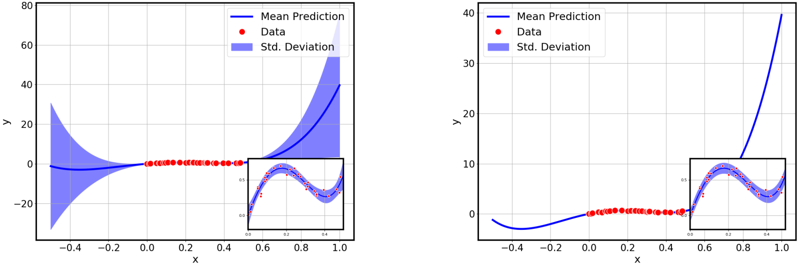
Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Extrapolation scenario

Order=4

True Posterior

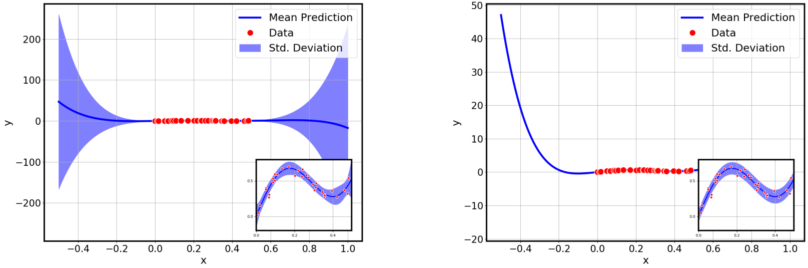
Variational Posterior



Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Extrapolation scenario

Order=5



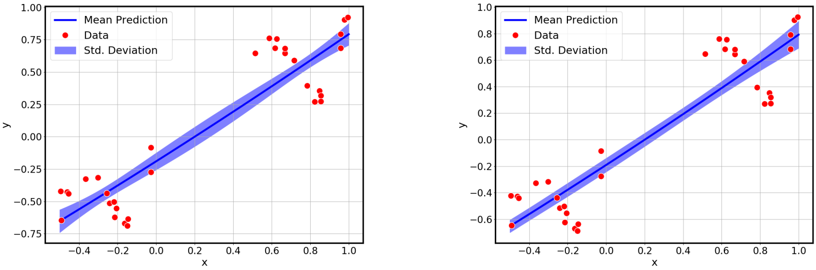
True Posterior

Variational Posterior

Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Interpolation scenario

Order=2



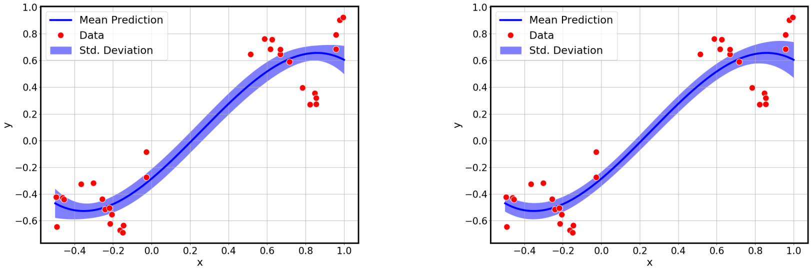
True Posterior

Variational Posterior

Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Interpolation scenario

Order=3



True Posterior

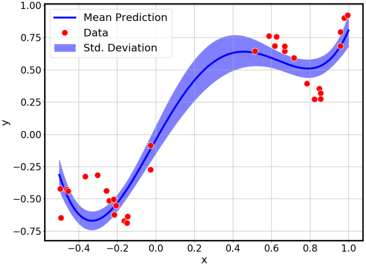
Variational Posterior

Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

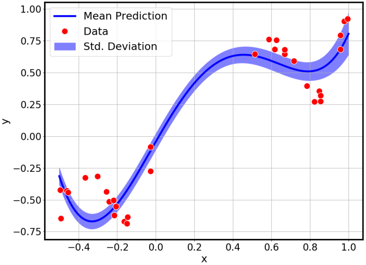
Polynomial fit: Interpolation scenario

Order=4

True Posterior



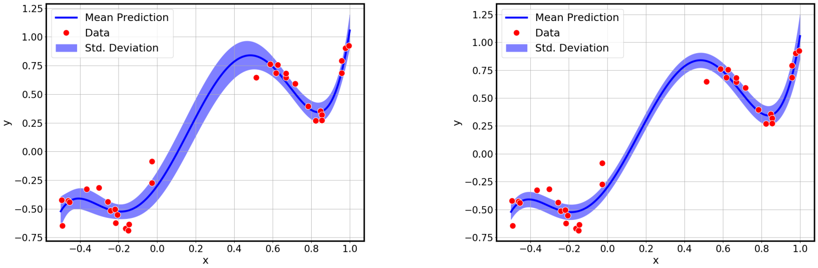
Variational Posterior



Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Interpolation scenario

Order=5


True Posterior

Variational Posterior

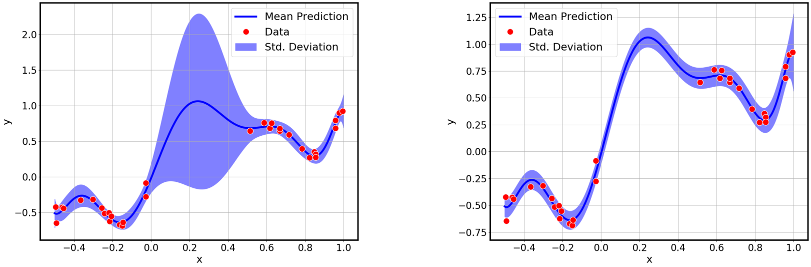
Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Polynomial fit: Interpolation scenario

Order=10

True Posterior

Variational Posterior



Variational posterior predictions heavily underestimate both interpolative and extrapolative errors.

Summary and Future Work Thank you!

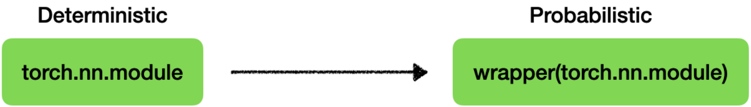
- UQ and ML for interatomic potential models

- Embedded model error for Bayesian inference of linear MLIAPs
  - Leads to data model with baked-in uncertainty
  - Meaningful model-error uncertainty capturing the true residual
  - Choices to make: priors, likelihoods, MCMC sampler, where to embed...

- Nonlinear MLIAPs, uncertainty estimation options
  - Bayesian inference: careful with likelihood assumptions; does not always work
  - Variational methods: underestimate/homogenize the uncertainty, parallel with embedded model error
  - Ensemble learning: mostly empirical, but they work!

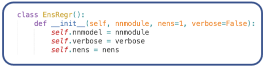
Additional Material

Uncertainty-enabling wrappers over PyTorch modules



Option 1: ensemble NN

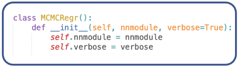
```
nn_ens = EnsRegr(torch.nn.module, nens=111)
```



```
class EnsRegr():
    def __init__(self, nnmodule, nens=1, verbose=False):
        self.nnmodel = nnmodule
        self.verbose = verbose
        self.nens = nens
```

Option 2: NN learning with MCMC

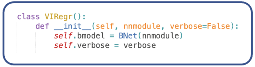
```
nn_mcmc = MCMCRegr(torch.nn.module)
```



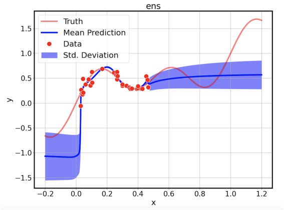
```
class MCMCRegr():
    def __init__(self, nnmodule, verbose=True):
        self.nnmodule = nnmodule
        self.verbose = verbose
```

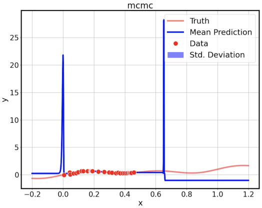
Option 3: NN learning with VI

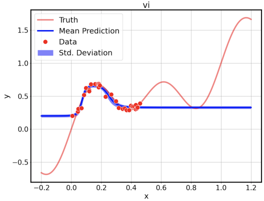
```
nn_vi = VIRegr(torch.nn.module)
```



```
class VIRegr():
    def __init__(self, nnmodule, verbose=False):
        self.bmodel = BNet(nnmodule)
        self.verbose = verbose
```

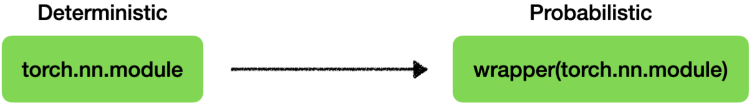






- MCMC struggles with complex NNs; VI underestimates; Ensembles do well

Uncertainty-enabling wrappers over PyTorch modules



Deterministic

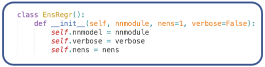
torch.nn.module

Probabilistic

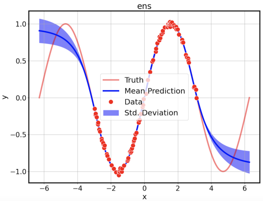
wrapper(torch.nn.module)

Option 1: ensemble NN

```
nn_ens = EnsRegr(torch.nn.module, nens=111)
```

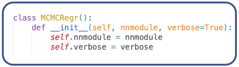


```
class EnsRegr():
    def __init__(self, nnmodule, nens=1, verbose=False):
        self.nnmodel = nnmodule
        self.verbose = verbose
        self.nens = nens
```

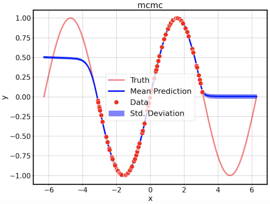


Option 2: NN learning with MCMC

```
nn_mcmc = MCMCRegr(torch.nn.module)
```

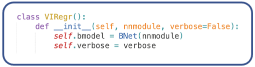


```
class MCMCRegr():
    def __init__(self, nnmodule, verbose=True):
        self.nnmodule = nnmodule
        self.verbose = verbose
```

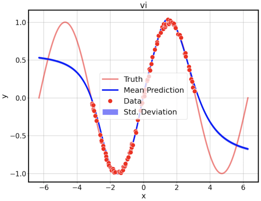


Option 3: NN learning with VI

```
nn_vi = VIRegr(torch.nn.module)
```



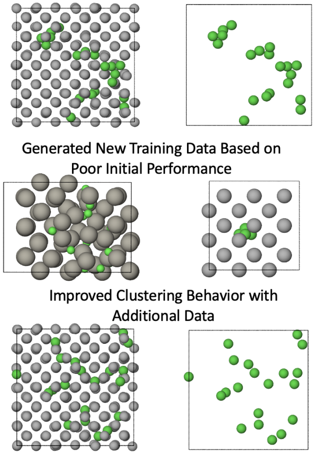
```
class VIRegr():
    def __init__(self, nnmodule, verbose=False):
        self.bmodel = BNet(nnmodule)
        self.verbose = verbose
```



- MCMC struggles with complex NNs; VI underestimates; Ensembles do well

Training set selection is crucial

- Configurations chosen for training data influence results
- Example: W-H (tungsten/hydrogen) IAPs
- Initial IAPs resulted in hydrogen clusters in bulk tungsten, which should not occur
- Additional training data was generated and put into the training set
- Including these specific configurations prevented unphysical hydrogen clustering



Grey: Tungsten **Green:** Hydrogen

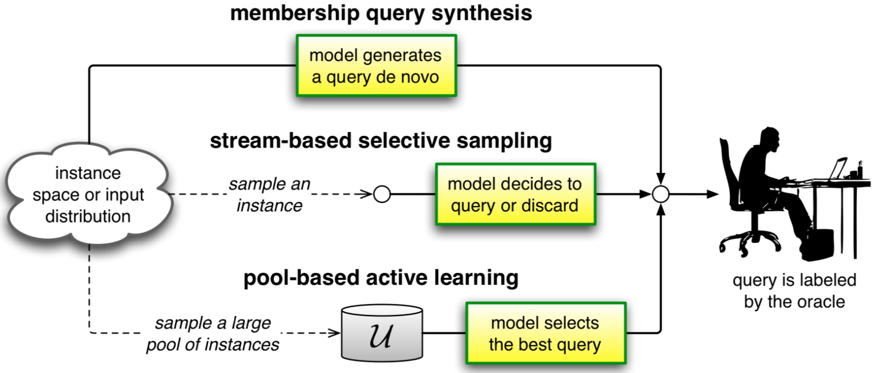
Initial Poor Hydrogen Clustering Behavior

Generated New Training Data Based on Poor Initial Performance

Improved Clustering Behavior with Additional Data

Results from Mary Alice Cusentino (SNL), using LAMMPS software.

Active Learning: selection of training configurations



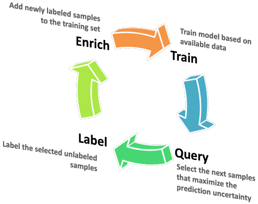
membership query synthesis
model generates a query de novo
stream-based selective sampling
instance space or input distribution
sample an instance
model decides to query or discard
pool-based active learning
sample a large pool of instances
$\mathcal{U}$
model selects the best query
query is labeled by the oracle

[B. Settles, “Active learning literature survey”, Computer Sciences Technical Report 1648, University of Wisconsin-Madison, 2009]

Active Learning: selection of training configurations

Greater test accuracy with fewer training samples

- Two flavors of the challenge:
  - Interpolation: developing a reliable problem-specific MLIAP that would accurately interpolates within the training domain is nontrivial
  - Extrapolation: prediction outside the training domain is even harder


Add newly labeled samples to the training set
Enrich
Train model based on available data
Train
Query
Select the next samples that maximize the prediction uncertainty
Label
Label the selected unlabeled samples

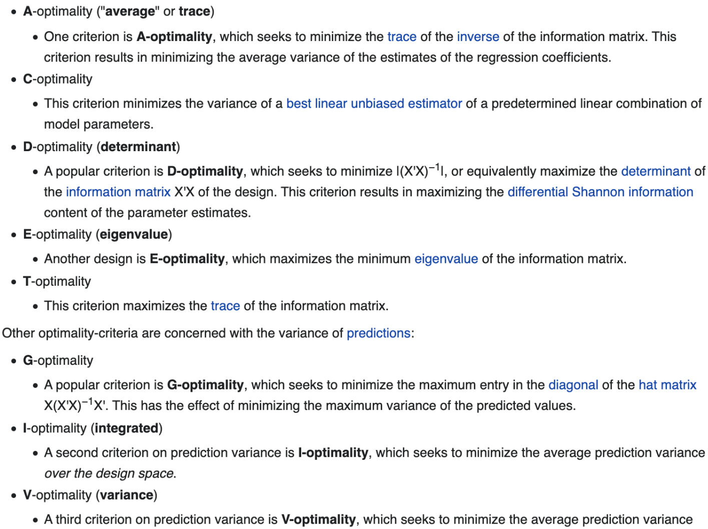
- Key: *query strategy*, whether to query high-fidelity quantum mechanical (QM) simulation or not.
  - If such decision can be made reliably, then one does not need to start with a very good training set

Query Strategies: almost all rely on some form of uncertainty estimate

- **Uncertainty sampling:** an active learner queries the instances about which it is least certain how to label.
- **Query-by-committee:** committee of competing models, and pick a query about which they most disagree. Need a measure of disagreement.
- **Expected model change:** which query would lead to greatest model change, e.g. largest gradient length.
- **Variance Reduction and Fisher Information Ratio:** minimizing the variance component of generalization error estimate (via Fisher Information)
- **Estimated error reduction:** Estimate the expected future error that would result if some new instance x is labeled and added to training set, and then select the instance that minimizes that expectation.

Optimality options

Straight out of wiki...

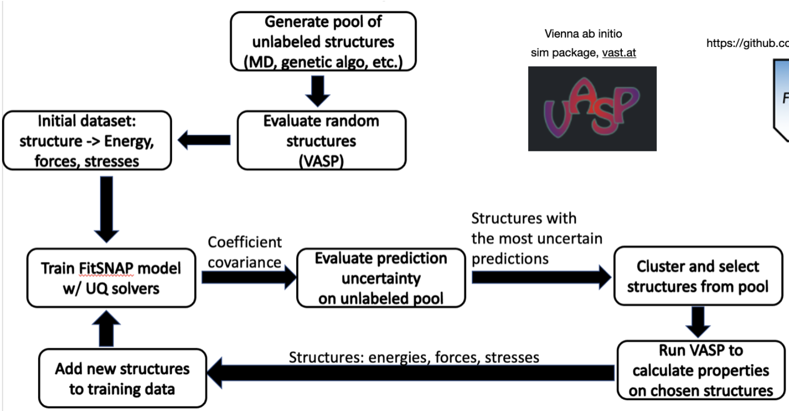


- **A**-optimality ("**average**" or **trace**)
  - One criterion is **A-optimality**, which seeks to minimize the trace of the inverse of the information matrix. This criterion results in minimizing the average variance of the estimates of the regression coefficients.
- **C**-optimality
  - This criterion minimizes the variance of a best linear unbiased estimator of a predetermined linear combination of model parameters.
- **D**-optimality (**determinant**)
  - A popular criterion is **D-optimality**, which seeks to minimize $|(X'X)^{-1}|$, or equivalently maximize the determinant of the information matrix X'X of the design. This criterion results in maximizing the differential Shannon information content of the parameter estimates.
- **E**-optimality (**eigenvalue**)
  - Another design is **E-optimality**, which maximizes the minimum eigenvalue of the information matrix.
- **T**-optimality
  - This criterion maximizes the trace of the information matrix.

Other optimality-criteria are concerned with the variance of predictions:

- **G**-optimality
  - A popular criterion is **G-optimality**, which seeks to minimize the maximum entry in the diagonal of the hat matrix $X(X'X)^{-1}X'$. This has the effect of minimizing the maximum variance of the predicted values.
- **I**-optimality (**integrated**)
  - A second criterion on prediction variance is **I-optimality**, which seeks to minimize the average prediction variance *over the design space*.
- **V**-optimality (**variance**)
  - A third criterion on prediction variance is **V-optimality**, which seeks to minimize the average prediction variance

Active Learning: current workflow



Generate pool of unlabeled structures (MD, genetic algo, etc.)

Evaluate random structures (VASP)

Initial dataset: structure -> Energy, forces, stresses

Train FitSNAP model w/ UQ solvers

Coefficient covariance

Evaluate prediction uncertainty on unlabeled pool

Structures with the most uncertain predictions

Cluster and select structures from pool

Run VASP to calculate properties on chosen structures

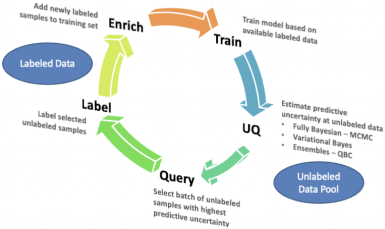
Structures: energies, forces, stresses

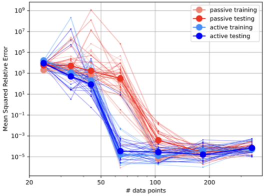
Add new structures to training data

Vienna ab initio
sim package, vast.at

https://github.com/FitSNAP/FitSNAP

Active Learning: Query Options


Add newly labeled samples to training set
Enrich
Train
Train model based on available labeled data
Labeled Data
UQ
Estimate predictive uncertainty at unlabeled data
• Fully Bayesian – MCMC
• Variational Bayes
• Ensembles – QBC
Label
Label selected unlabeled samples
Query
Select batch of unlabeled samples with highest predictive uncertainty
Unlabeled Data Pool



Query-by-Committee (QBC)

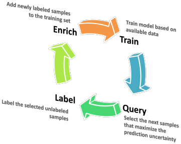
- Launch K learners, each with fN training points (f=0.8)
- Evaluate the learners’ performance at all points in the pool
- Select training points from the pool that correspond to the highest ’disagreement’ and add them to the training set

Bayesian Uncertainty

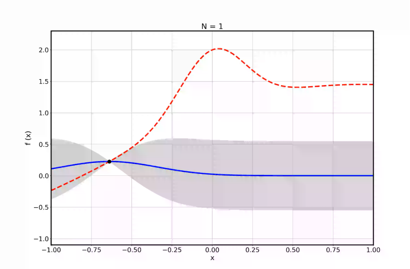
- Launch a single learner
- Evaluate its performance at all points in the pool
- Select training points from the pool that correspond to the highest posterior uncertainty and add them to the training set

Demonstration of AL

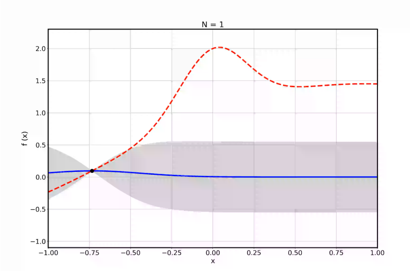
- Selecting one point at a time given the current uncertainty estimate


Add newly labeled samples to the training set
Enrich
Train model based on available data
Train
Query
Select the next samples that maximize the prediction uncertainty
Label
Label the selected unlabeled samples

Naïve approach

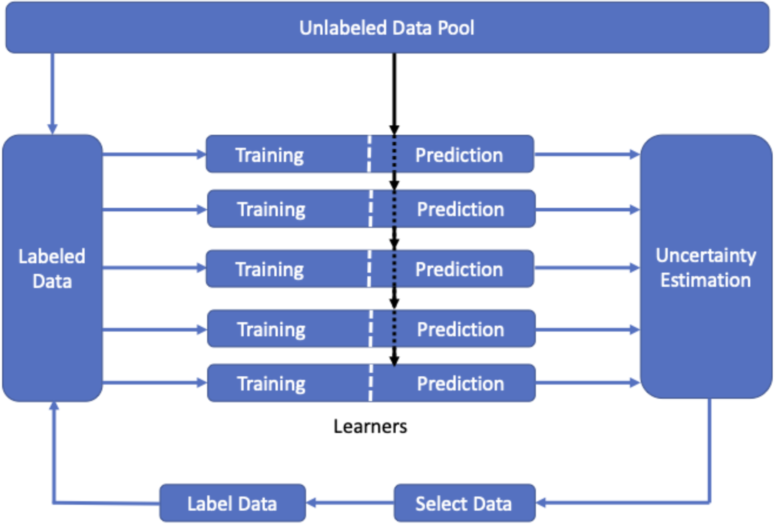


Active approach



- Next: how to reliably estimate uncertainty?

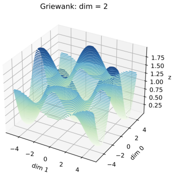
Query-by-Committee (QBC): algorithm sketch



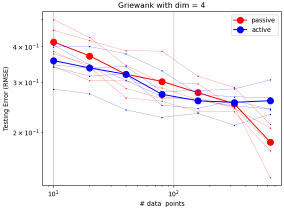
Query-by-Committee (QBC): algorithm outline

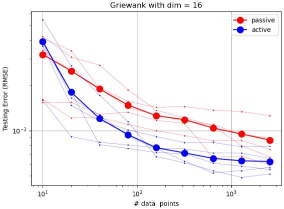
- Start with a large pool of P unlabeled points
- Select a training set of N points from the pool
- Launch K learners, each with fN randomly-chosen training points
    - Random sampling with replacement
    - Selection of fraction f determines data size per learner
        - diversity vs data size tradeoff
- Evaluate the learners' performance at all points in the pool
- Select M points from the pool, having highest 'disagreement', & add them to the training set
    - M choice, size of batch added per query, low error vs optimal choice
    - K-means clustering to discover geometry of selected data
    - Distribute data from clusters evenly among learners
        - Add fM points per learner with replacement
- Re-train, and repeat query to evaluate learners performance on prediction of unlabeled data in pool

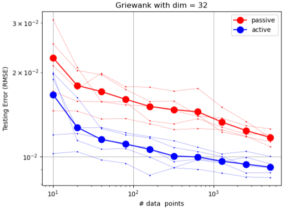
QBC: Griewank test function



$$f(\boldsymbol{x}) = 1 + \sum_{i=1}^{d} \frac{x_i^2}{4000} - \prod_{i=1}^{d} \cos\left(\frac{x_i}{\sqrt{i}}\right)$$

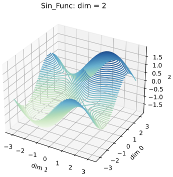




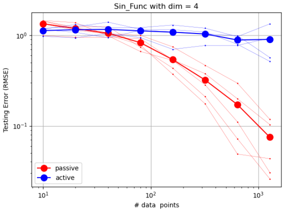


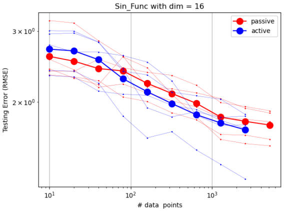
- Efficiency of active learning improves with higher dimension.

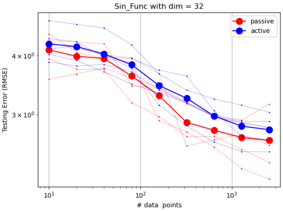
QBC: Sine test function



$$f(\boldsymbol{x}) = \sin\left(\sum_{i=1}^{d} x_i\right)$$







- In low-d, large pool size causes newly selected points to cluster.
- Potential solution: sample according to PDF $e^{-std(x)}$ to concentrate new points near high uncertainty region, but select elsewhere, too.

Literature

Model error embedding

- [Sargsyan et al., 2019] “Embedded model error representation for Bayesian model calibration”, *Int. J. Uncertain. Quantif.*, 9(4), 2019.

MLIAPs

- [Thompson et al., 2015] “Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials”, *J Comp Phys*, 2015.
- [J. Behler, 2014] “Representing potential energy surfaces by high-dimensional neural network potentials”, *J. Phys.: Condens. Matter*, 26, 2014.

Active learning

- [B. Settles, 2009] “Active learning literature survey”, *Comp Sci Tech Report 1648*, University of Wisconsin-Madison, 2009.

Active learning for MLIAPs

- [E. Podryabinkin, A. Shapeev, 2017] “Active learning of linearly parametrized interatomic potentials”, *Comp Mat Sci*, 140, 2017.
- [J. Vandermause et al., 2020] “On-the-fly active learning of interpretable Bayesian force fields for atomistic rare events”, *npj Computational Materials*, 6, 2020.