Training machine learned potentials on the fly

Jonathan Vandermause

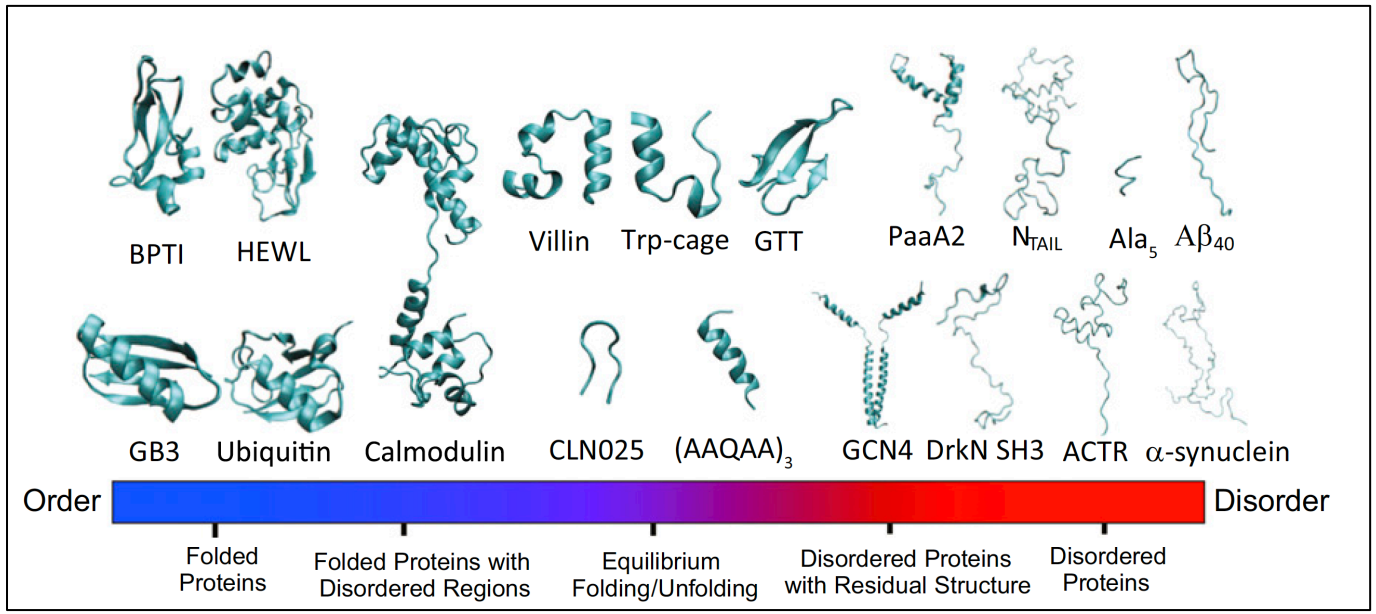
Department of Physics, Harvard University

Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

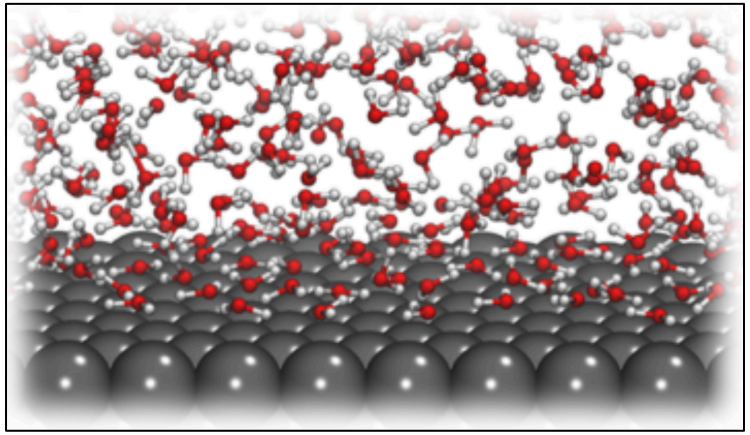
Atomistic simulation

protein folding



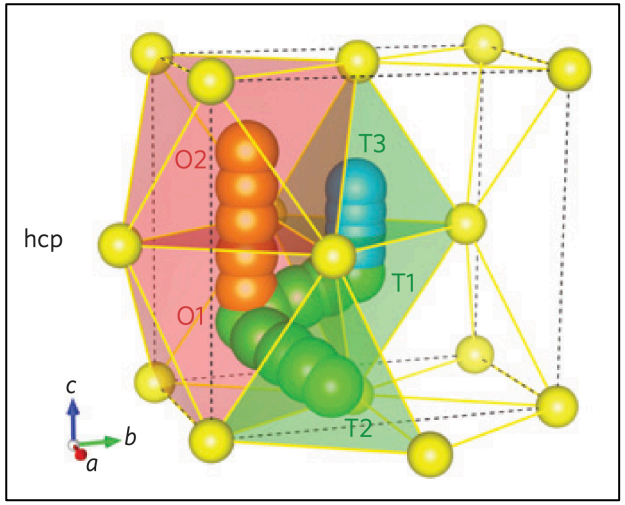
Robustelli, Paul, Stefano Piana, and David E. Shaw. *PNAS* 115.21 (2018): E4758-E4766.

heterogeneous catalysis



Steinmann, Stephan N., et al. *JCTC* 14.6 (2018): 3238-3251.

lithium ion diffusion



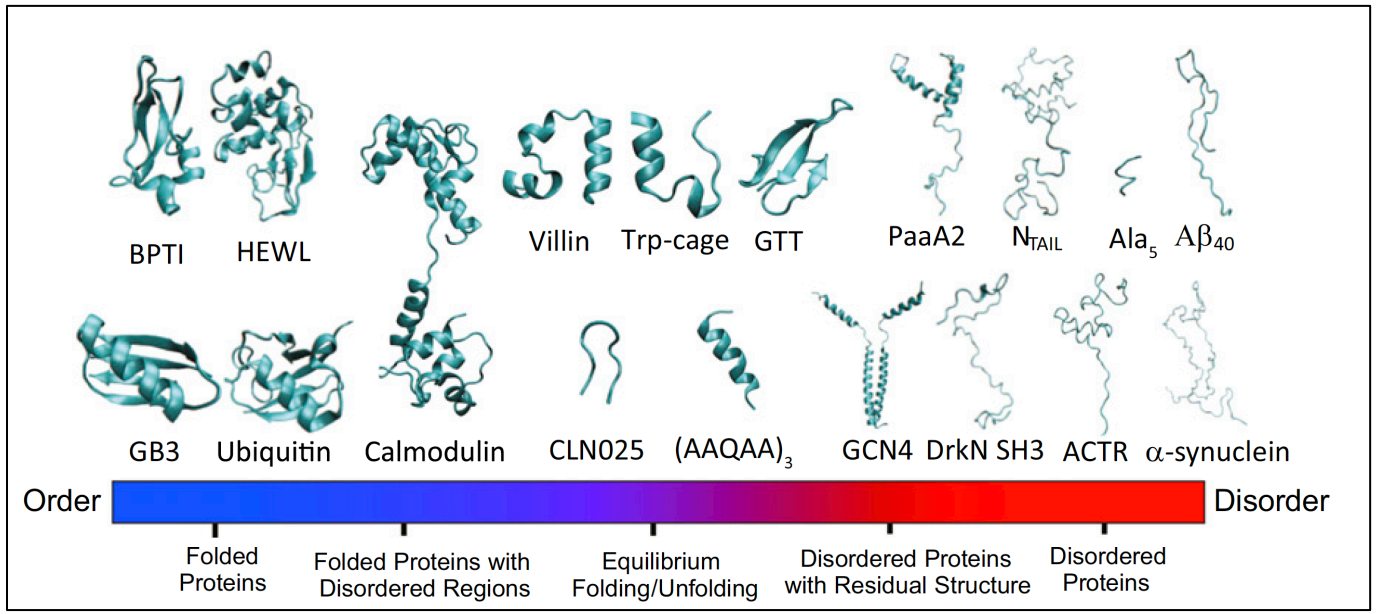
Wang, Yan, et al. *Nature materials* 14.10 (2015): 1026.

Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

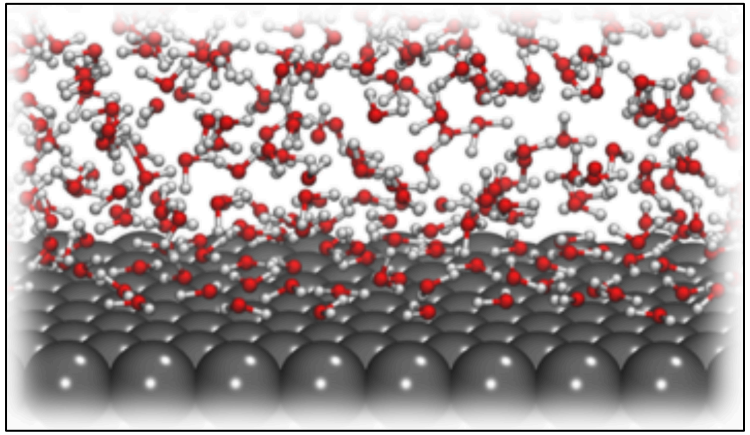
Atomistic simulation

protein folding



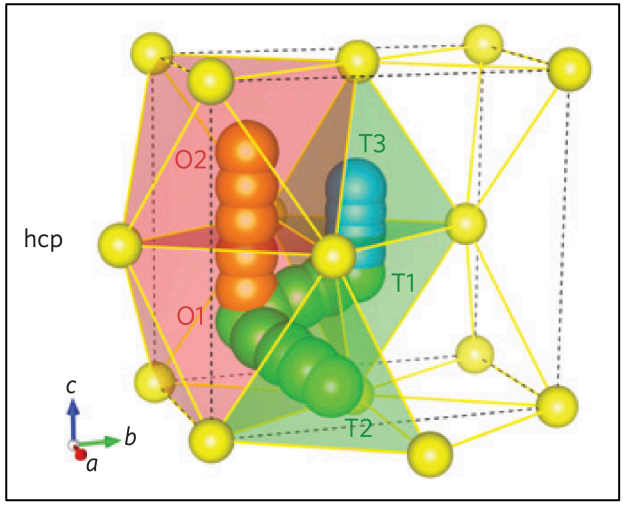
Robustelli, Paul, Stefano Piana, and David E. Shaw. *PNAS* 115.21 (2018): E4758-E4766.

heterogeneous catalysis



Steinmann, Stephan N., et al. *JCTC* 14.6 (2018): 3238-3251.

lithium ion diffusion



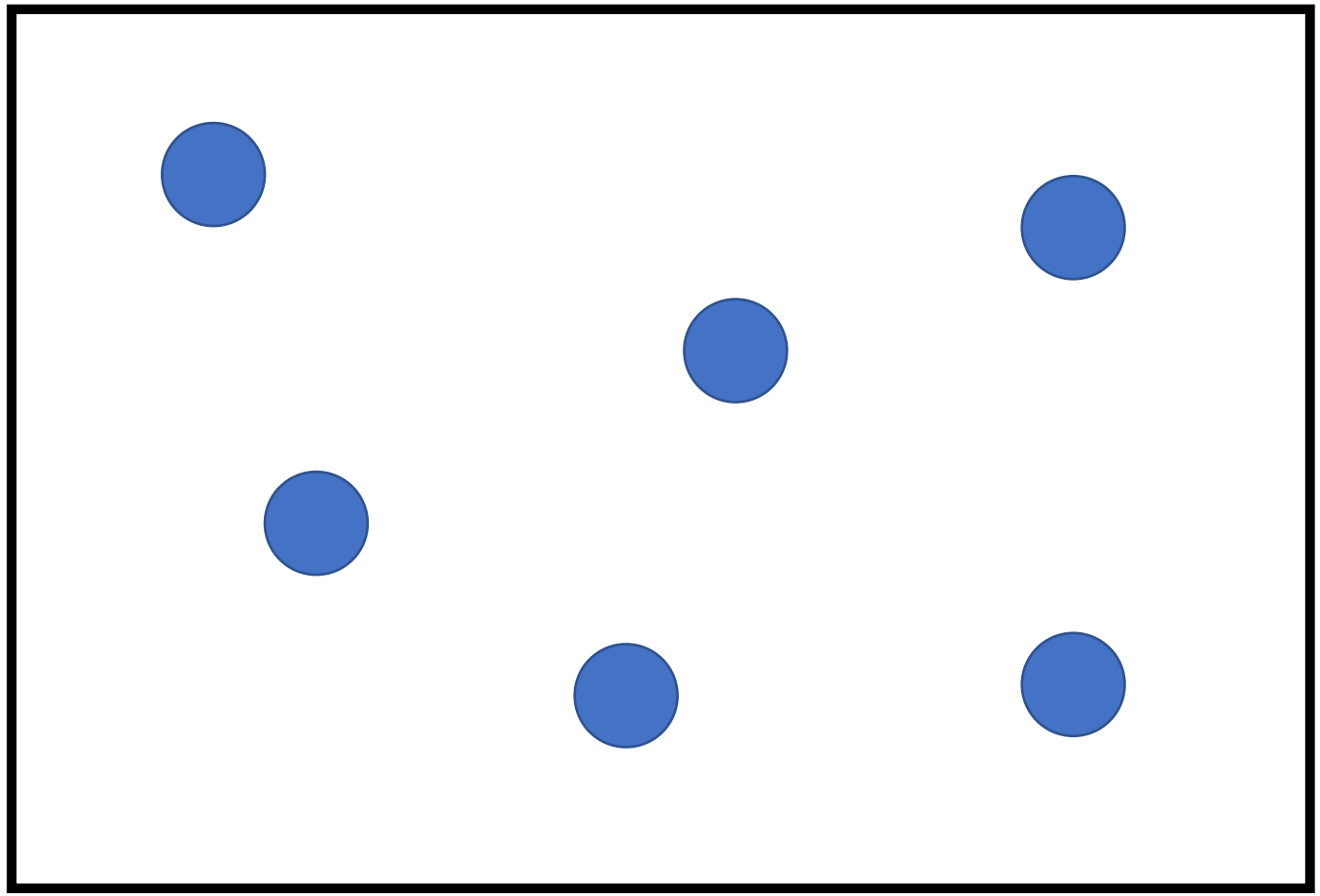
Wang, Yan, et al. *Nature materials* 14.10 (2015): 1026.

***Wanted**: cheap force fields for complex systems with the accuracy of quantum mechanics.*

Materials Intelligence Research

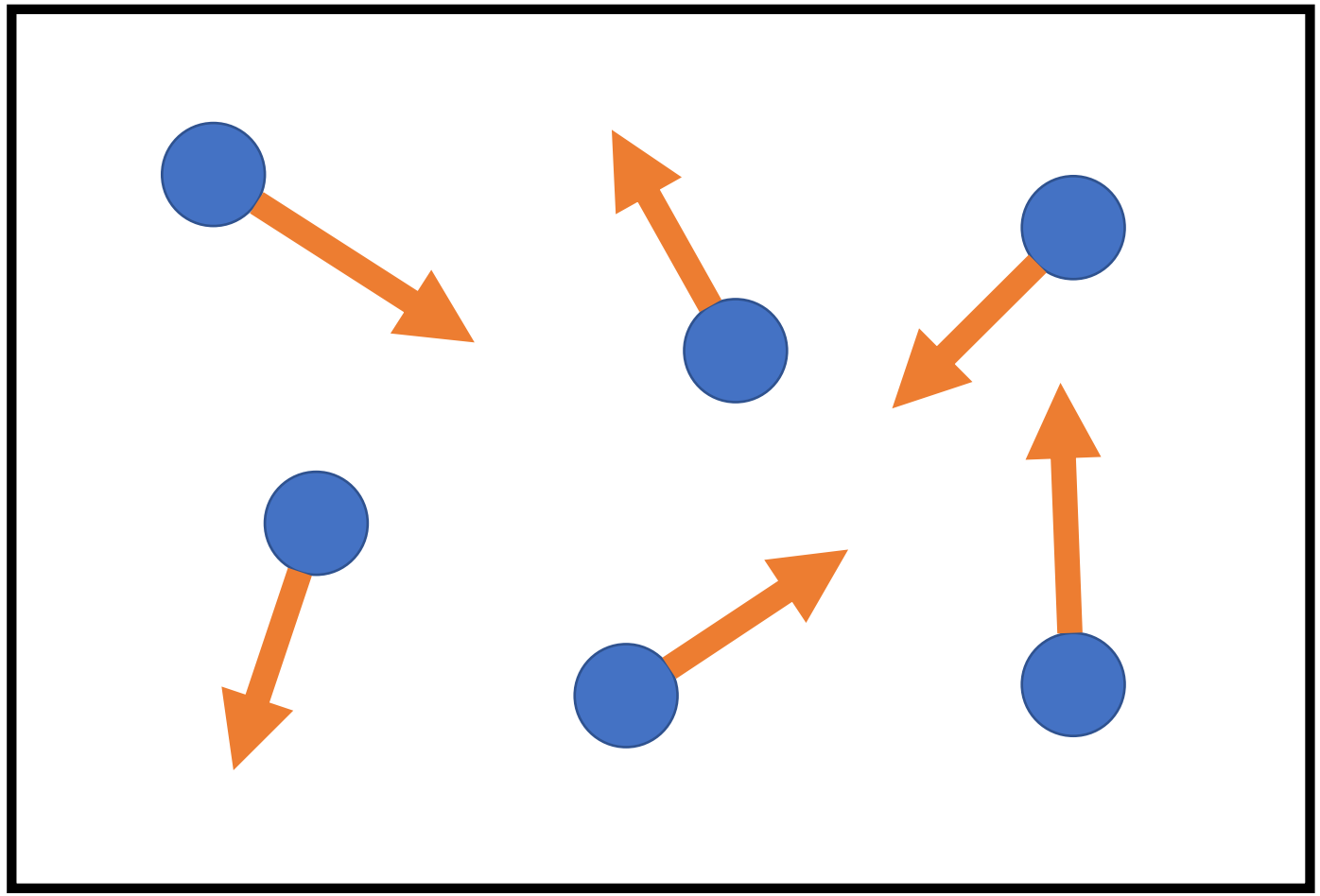
HARVARD School of Engineering and Applied Sciences

Molecular dynamics



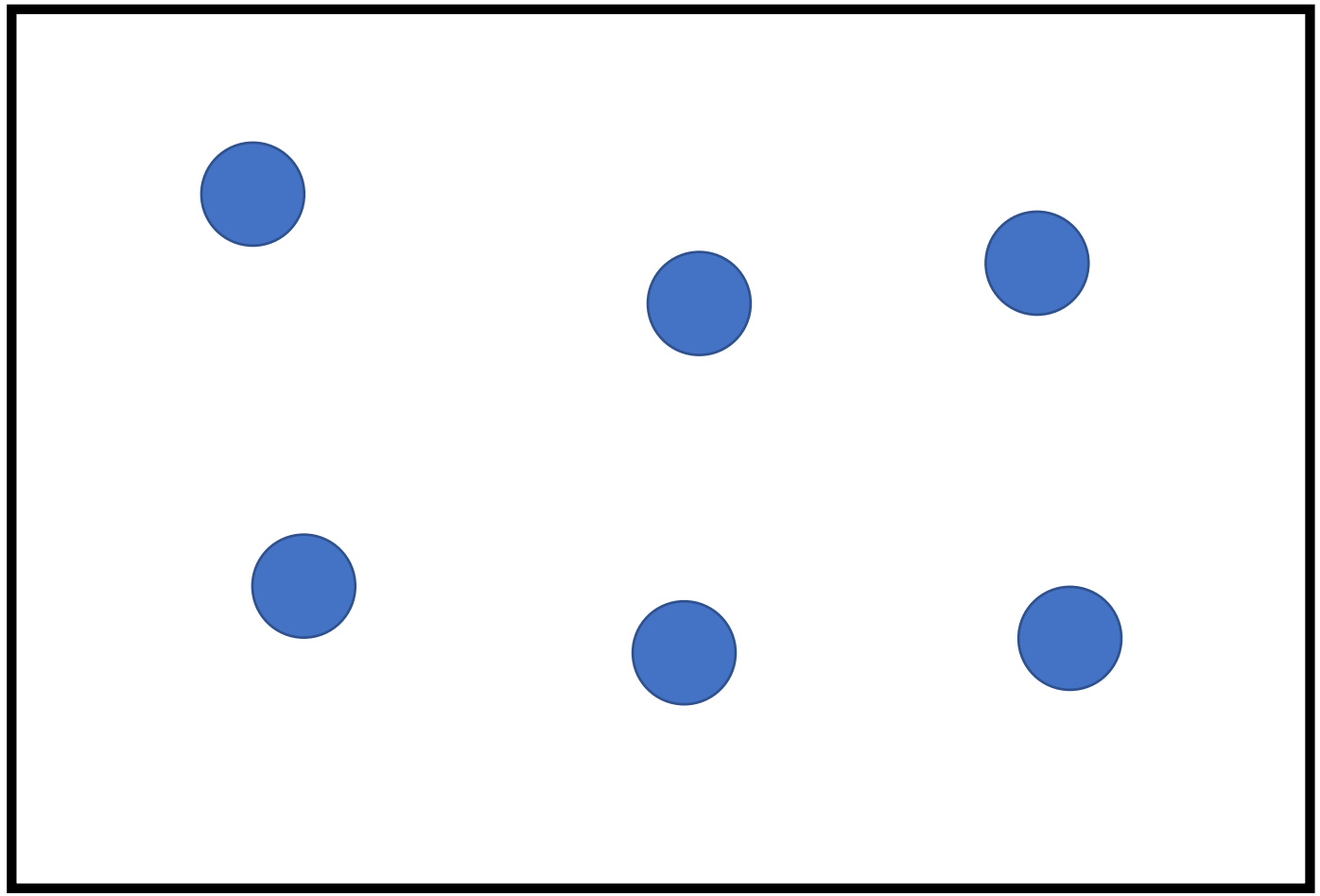
1. initialize

Molecular dynamics



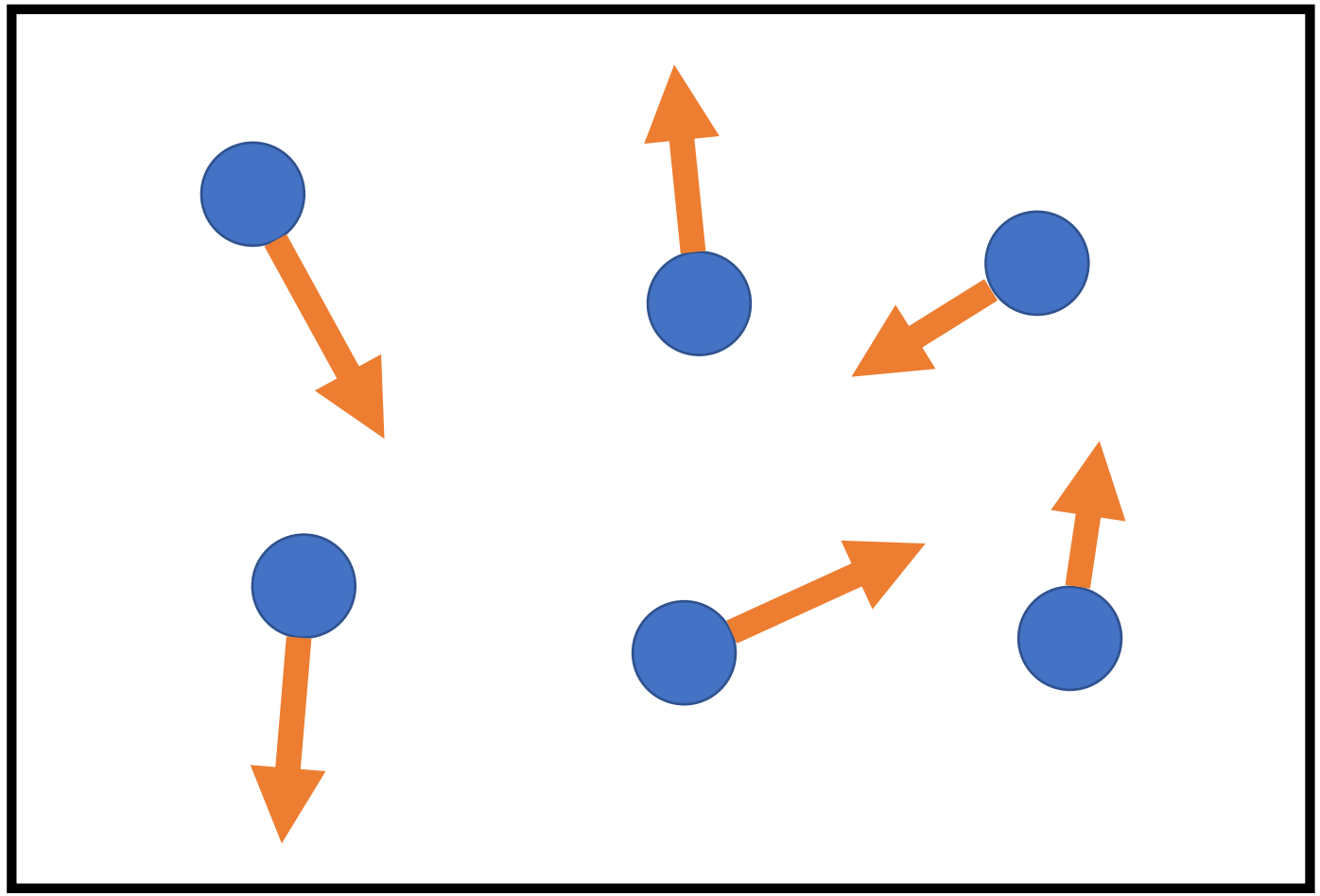
1. initialize
2. compute forces

Molecular dynamics



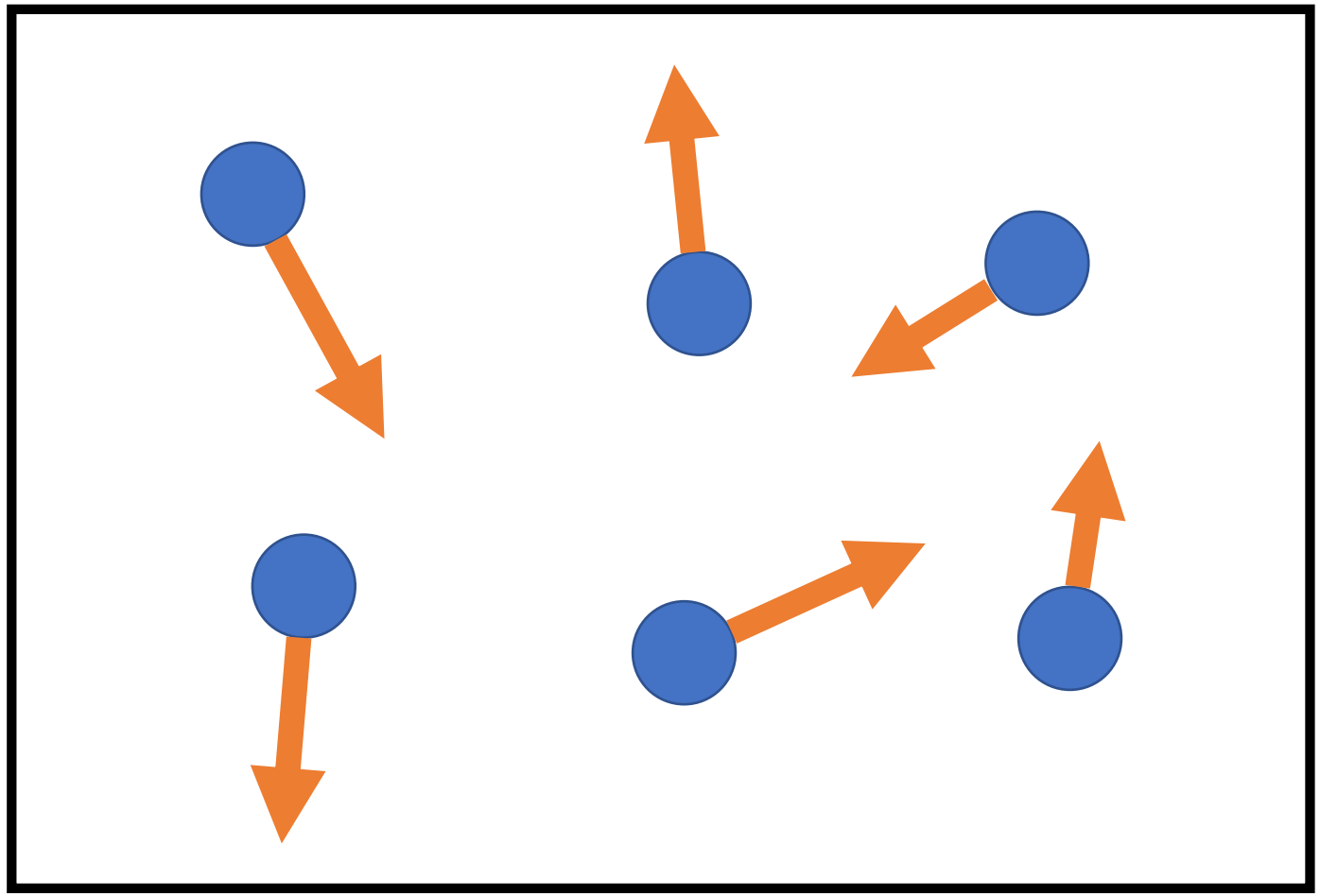
1. initialize
2. compute forces
3. update positions

Molecular dynamics



1. initialize
2. compute forces
3. update positions
4. repeat

Molecular dynamics



1. initialize
2. ***compute forces***
3. update positions
4. repeat

To scale up, forces need to be computed cheaply and accurately.

Interatomic potentials

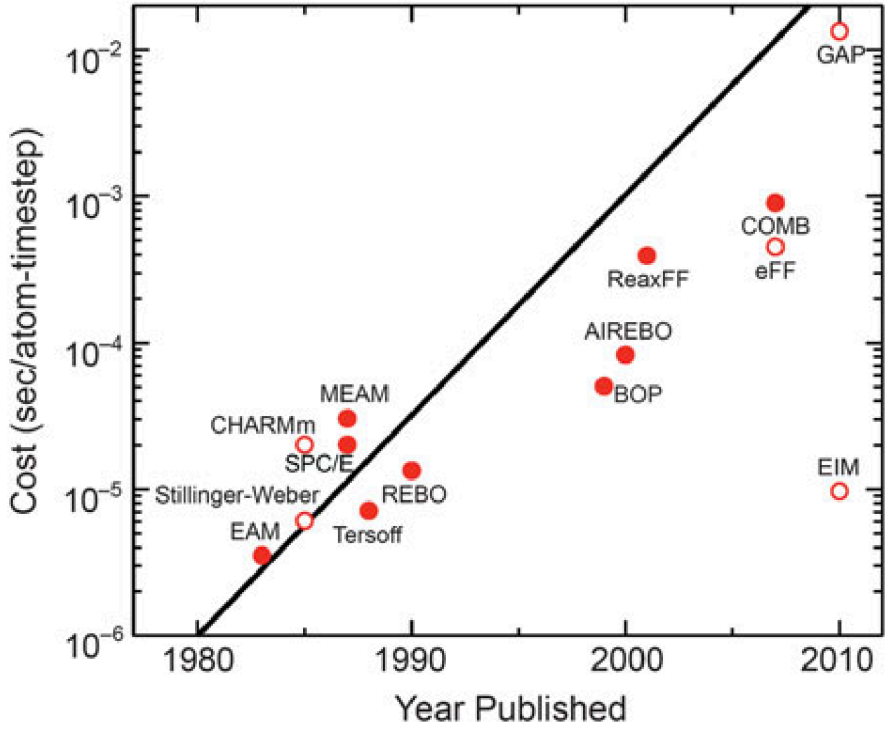
Density functional theory (DFT) is accurate, but too slow.

# atoms	# electrons	sec/atom/step
192	1024	252
432	3456	1344

Steven J. Plimpton and Aidan P. Thompson. *MRS bulletin* 37.5 (2012): 513-521.

Interatomic potentials

Interatomic potentials are several orders of magnitude faster than DFT.



Steven J. Plimpton and Aidan P. Thompson. *MRS bulletin* 37.5 (2012): 513-521.

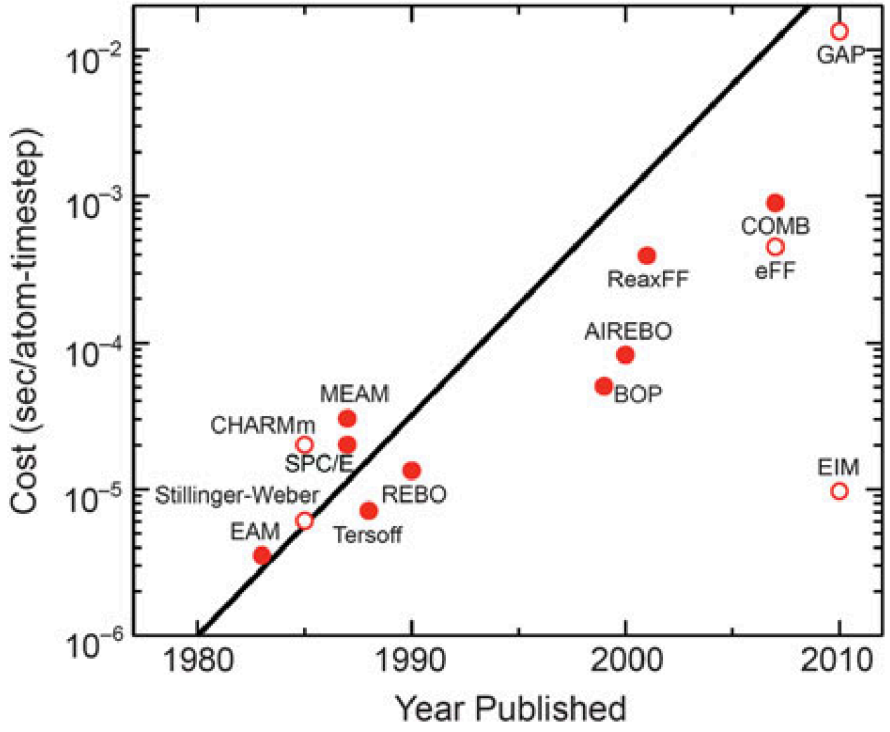
Density functional theory (DFT) is accurate, but too slow.

# atoms	# electrons	sec/atom/step
192	1024	252
432	3456	1344

Steven J. Plimpton and Aidan P. Thompson. *MRS bulletin* 37.5 (2012): 513-521.

Interatomic potentials

Interatomic potentials are several orders of magnitude faster than DFT.



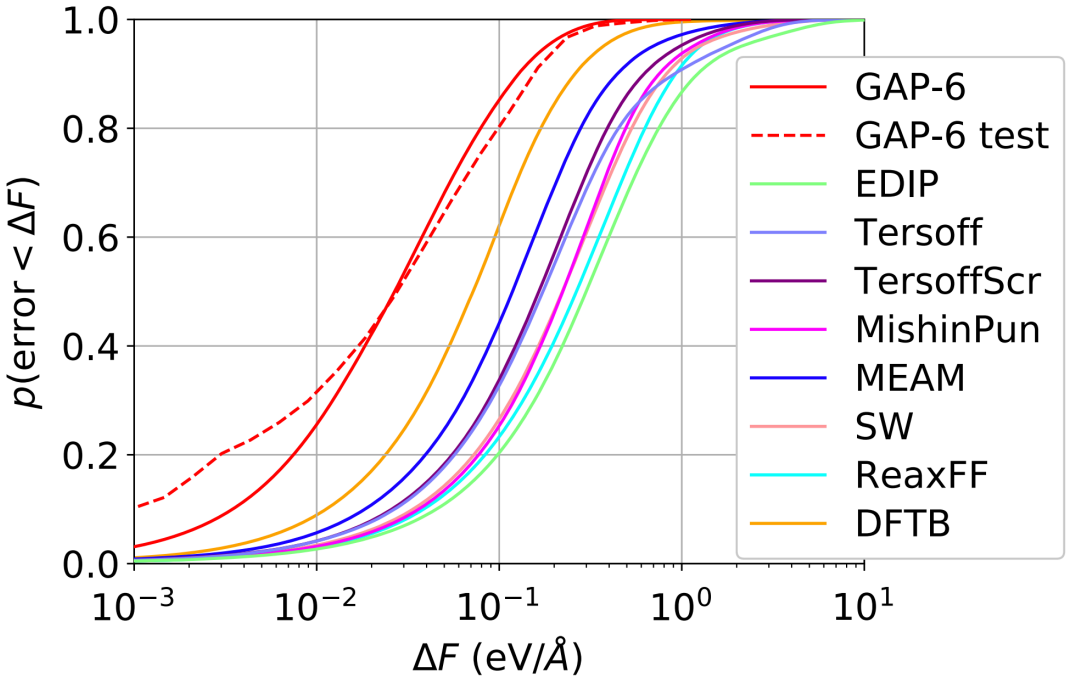
Steven J. Plimpton and Aidan P. Thompson. *MRS bulletin* 37.5 (2012): 513-521.

Density functional theory (DFT) is accurate, but too slow.

# atoms	# electrons	sec/atom/step
192	1024	252
432	3456	1344

Steven J. Plimpton and Aidan P. Thompson. *MRS bulletin* 37.5 (2012): 513-521.

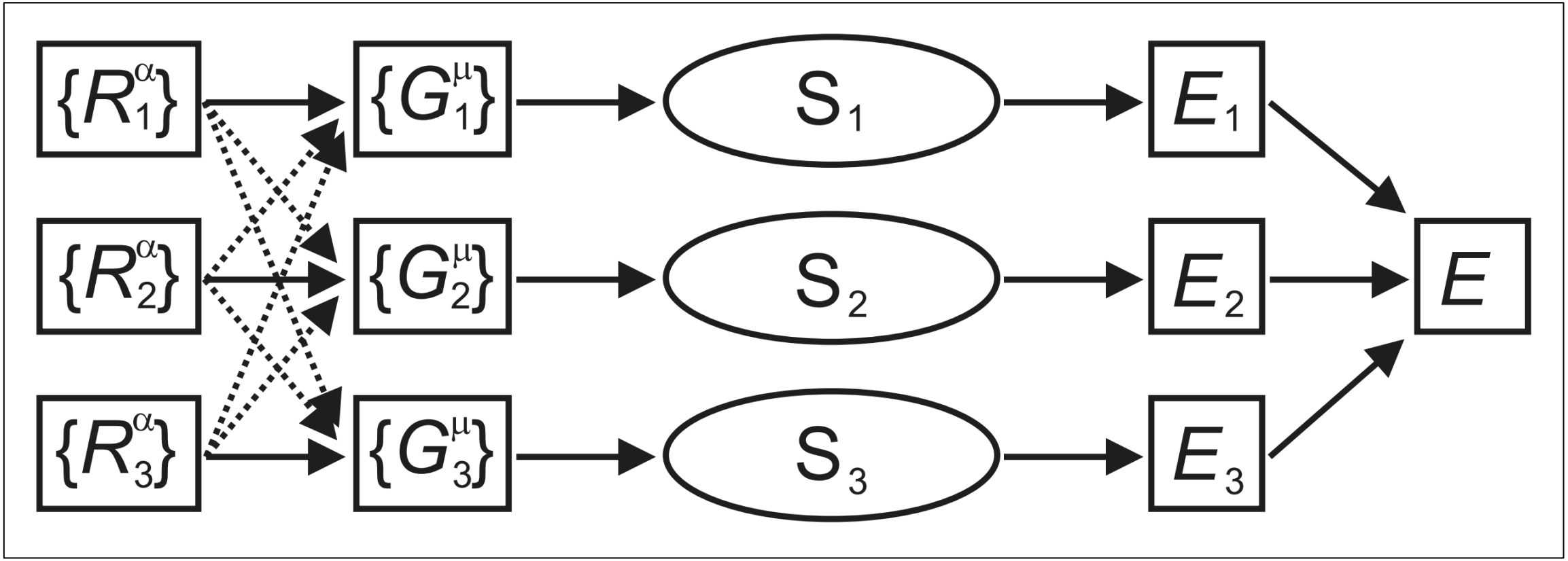
Flexible potentials can approach the accuracy of DFT.



Bartók, Albert P., Kermode, J., Bernstein, N., & Csányi, G.. *PRX* 8.4 (2018): 041048.

Machine learned potentials

Approach #1: Train a flexible black box model on a large database of DFT data.



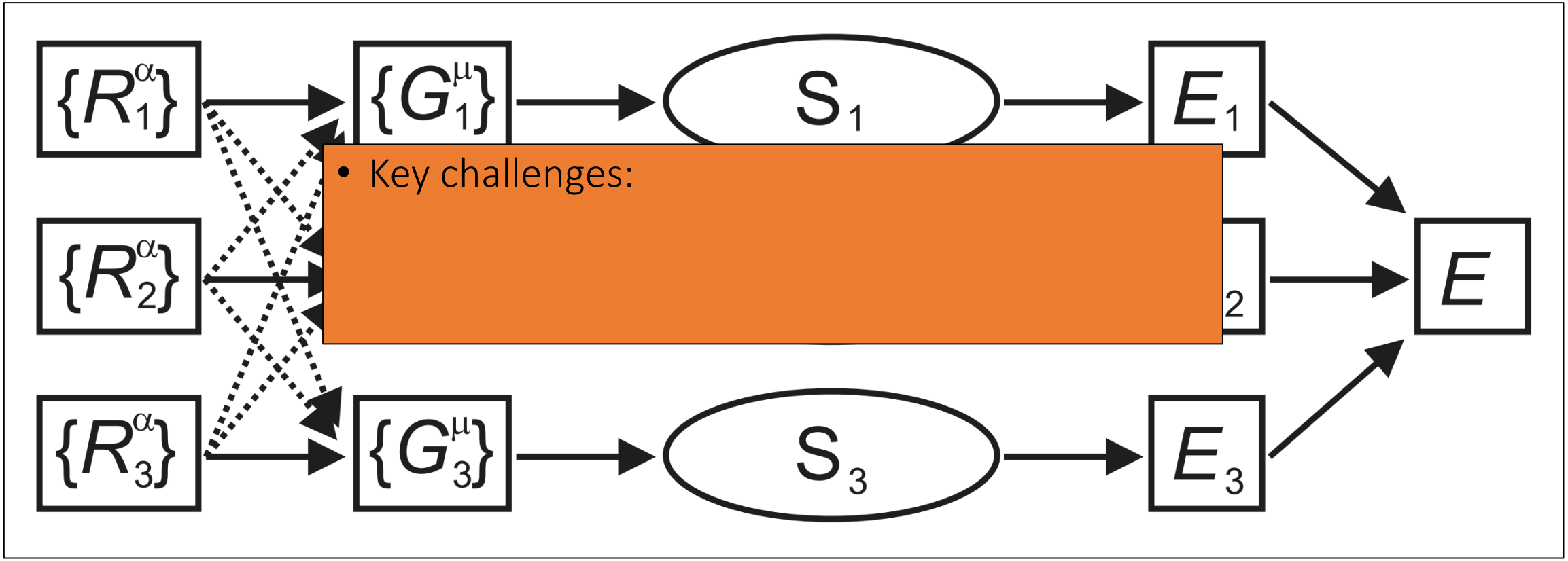
Behler, Jörg, and Parrinello, Michele. *PRL* 98.14 (2007): 146401.

Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

Machine learned potentials

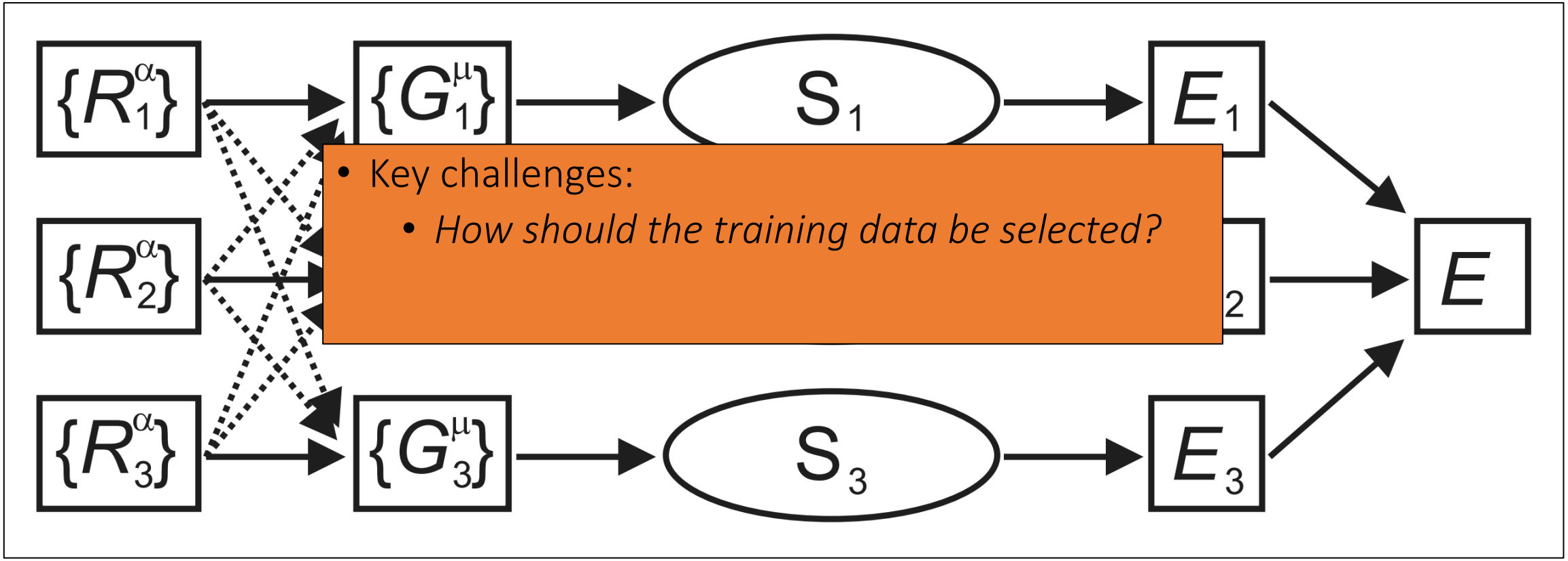
Approach #1: Train a flexible black box model on a large database of DFT data.



Behler, Jörg, and Parrinello, Michele. *PRL* 98.14 (2007): 146401.

Machine learned potentials

Approach #1: Train a flexible black box model on a large database of DFT data.

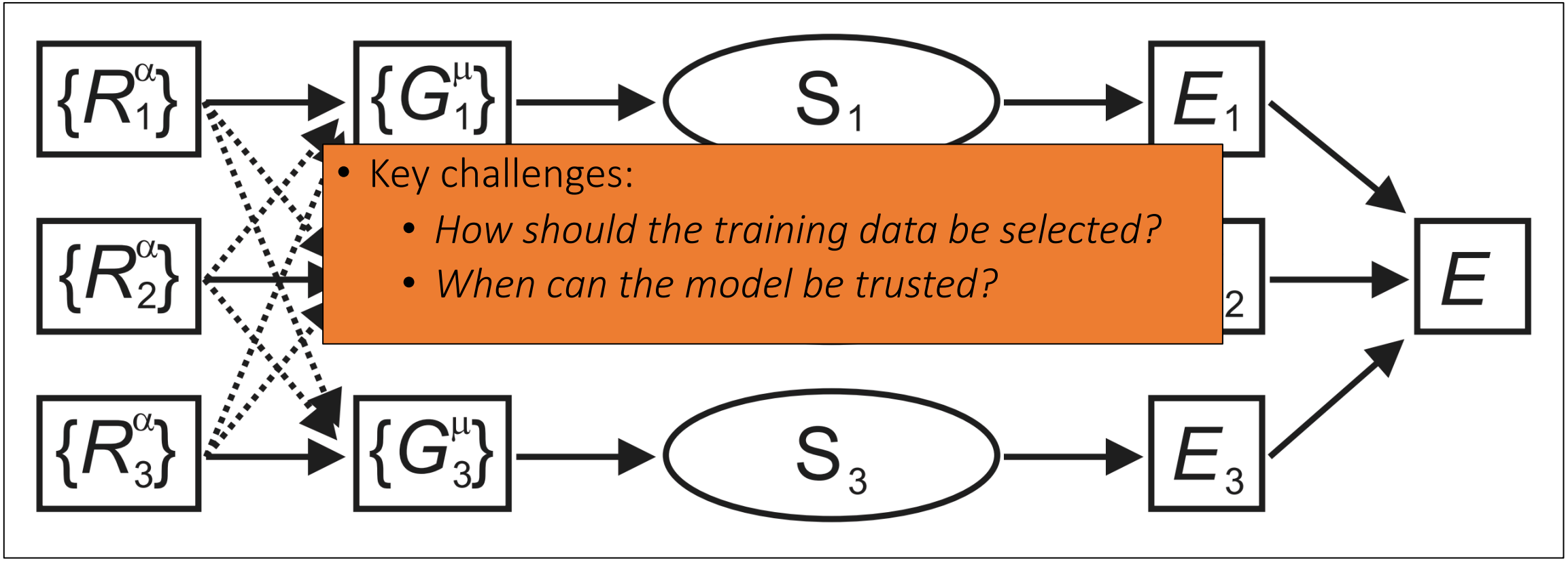


- Key challenges:
  - *How should the training data be selected?*

Behler, Jörg, and Parrinello, Michele. *PRL* 98.14 (2007): 146401.

Machine learned potentials

Approach #1: Train a flexible black box model on a large database of DFT data.



- Key challenges:
  - *How should the training data be selected?*
  - *When can the model be trusted?*

Behler, Jörg, and Parrinello, Michele. *PRL* 98.14 (2007): 146401.

Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

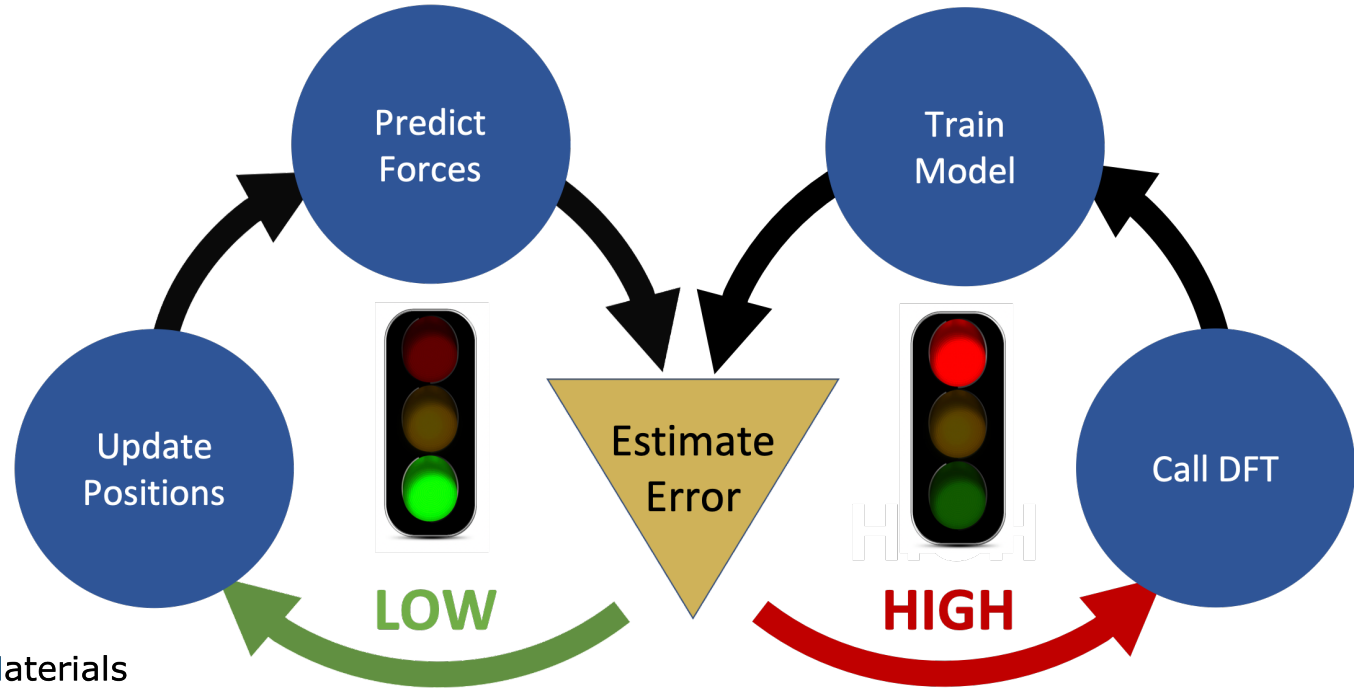
Machine learned potentials

Approach #2: Grow the training set adaptively during an MD simulation.

Machine learned potentials

Approach #2: Grow the training set adaptively during an MD simulation.

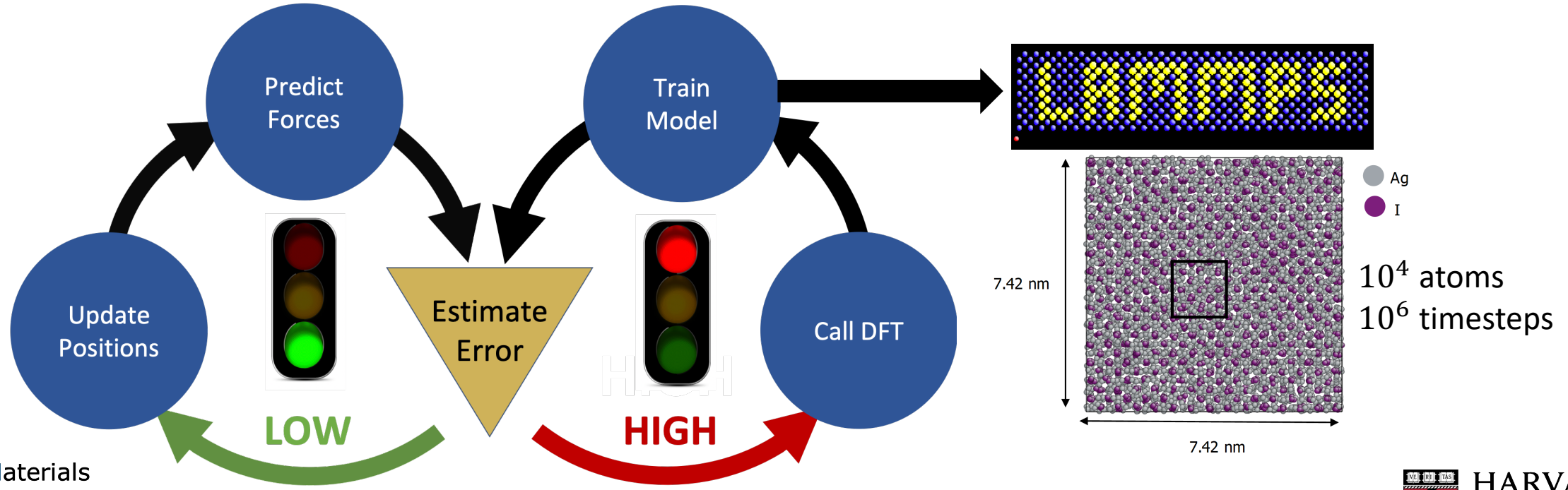
Goal: Automatically learn fast, accurate, multi-element force fields for large-scale MD.
Tool: Bayesian inference with Gaussian process regression.



Machine learned potentials

Approach #2: Grow the training set adaptively during an MD simulation.

Goal: Automatically learn fast, accurate, multi-element force fields for large-scale MD.
Tool: Bayesian inference with Gaussian process regression.



On-the-fly learning is on the rise

On-the-fly learning method	ML Model	Descriptor / Kernel	Extrapolation Detection	Low dimensional?
Li, Kermode, and De Vita (2015) [1]	Gaussian process	Internal vectors	True error	No
Podryabinkin and Shapeev (2017) [2]	Linear regression	Moment tensor polynomials	D-optimality criterion	No
Bernstein, Csányi, and Deringer (2019) [3]	Gaussian process	Smooth overlap of atomic positions (SOAP) [4]	Random structure search	No
Jinnouchi *et al.* (2019) [4]	Bayesian linear regression	Multi-element SOAP	Bayesian uncertainty	No
Fast Learning of Atomistic Rare Events (FLARE) [5]	**Gaussian process**	**Covariant 2+3 body kernel**	**Bayesian uncertainty**	**Yes**

[1] Li, Zhenwei, James R. Kermode, and Alessandro De Vita. *Physical review letters* 114.9 (2015): 096405.
[2] Podryabinkin, Evgeny V., and Alexander V. Shapeev. *Computational Materials Science* 140 (2017): 171-180.
[3] Bernstein, Noam, Gábor Csányi, and Volker L. Deringer. *npj Computational Materials* (2019): 99.
[4] Jinnouchi, Ryosuke, et al. *Physical Review Letters* 122.22 (2019): 225701.
[5] Vandermause, J., Torrisi, S. B., Batzner, S., Xie, Y., Sun, L., Kolpak, A. M., & Kozinsky, B. (2020 *npj Computational Materials*, *6*(1), 1-11.

Low-dimensional force fields

- The Born-Oppenheimer potential energy surface is high dimensional:

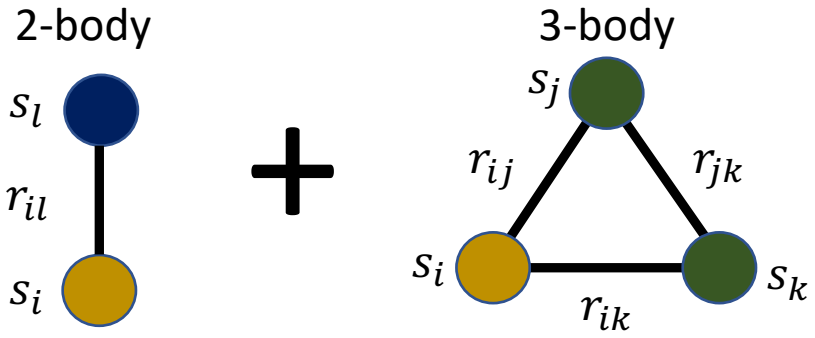
$$E = E(\vec{r}_1, \ldots, \vec{r}_N).$$

Low-dimensional force fields

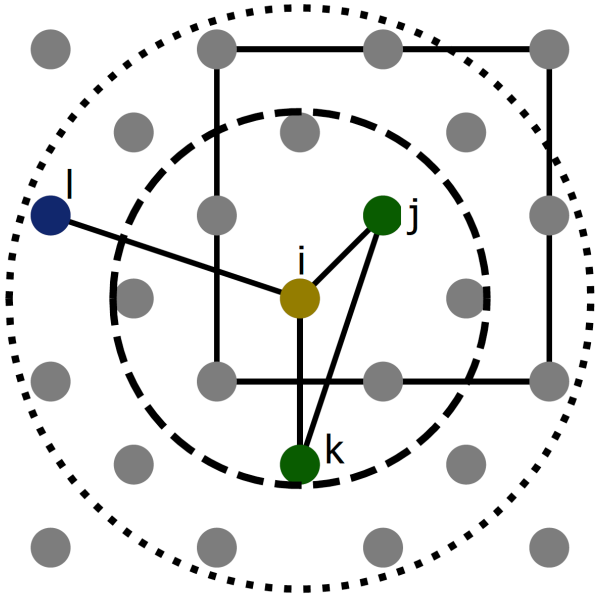
- The Born-Oppenheimer potential energy surface is high dimensional:

$$E = E(\vec{r}_1, \ldots, \vec{r}_N).$$

- **Simplifying assumption:** each cluster of 2 and 3 atoms in the local environment of atom i contributes to its local energy E_i.



Local environment of atom i

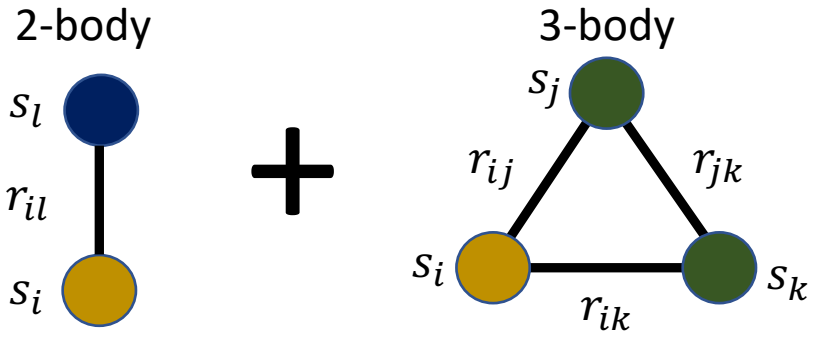


Low-dimensional force fields

- The Born-Oppenheimer potential energy surface is high dimensional:

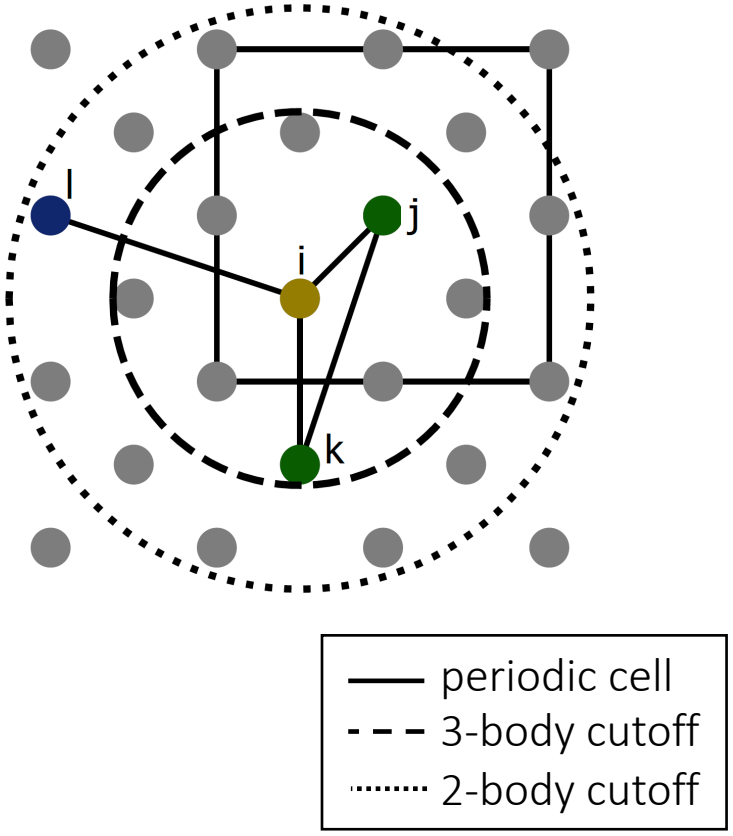
$$E = E(\vec{r}_1, \ldots, \vec{r}_N).$$

- **Simplifying assumption:** each cluster of 2 and 3 atoms in the local environment of atom i contributes to its local energy E_i.



- **Regression task:** learn $\varepsilon_{s_i, s_l}(r_{il})$ and $\varepsilon_{s_i,s_j,s_k}(r_{ij}, r_{ik}, r_{jk})$ from *ab initio* force data.

Local environment of atom i



Gaussian process regression

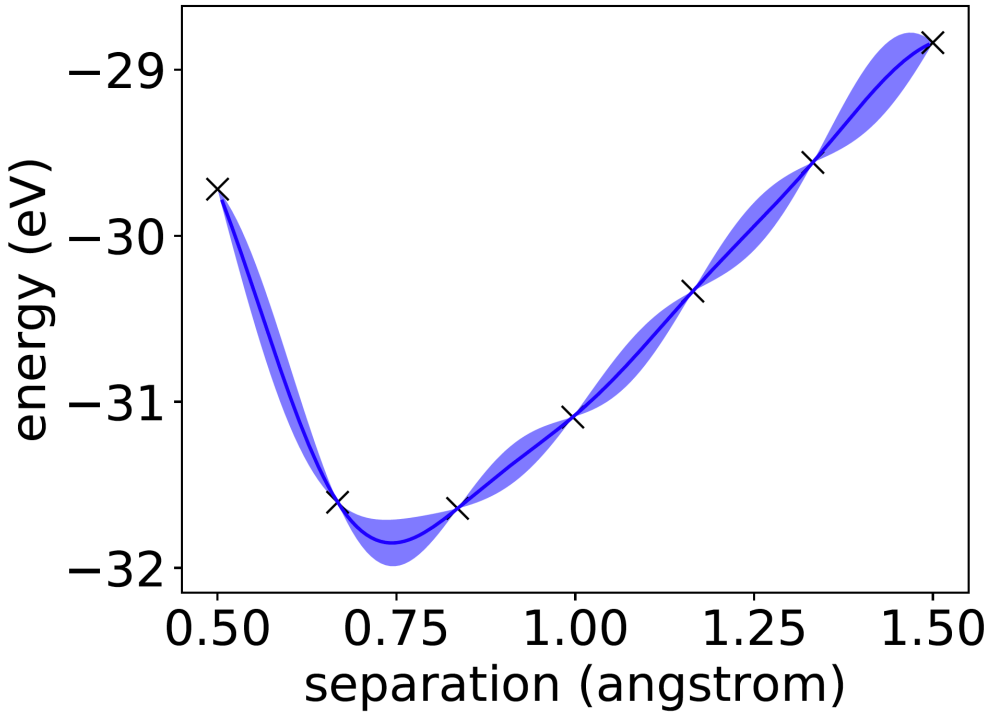
- Gaussian processes put a probability distribution over functions according to Bayes' rule.

$$p(\mathbf{f}, \mathbf{f}_* | \mathbf{y}) = \frac{p(\mathbf{f}, \mathbf{f}_*) p(\mathbf{y}|\mathbf{f})}{p(\mathbf{y})}$$

- The posterior is calculated *exactly* by assuming a Gaussian prior and likelihood.

$$p(\mathbf{f}, \mathbf{f}_*) = \mathcal{N}\left(0, \begin{bmatrix} K_{\mathbf{f},\mathbf{f}} & K_{*,\mathbf{f}} \\ K_{\mathbf{f},*} & K_{*,*} \end{bmatrix}\right)$$

$$p(\mathbf{y}|\mathbf{f}) = \mathcal{N}(\mathbf{f}, \sigma_n^2 I)$$



Materials Intelligence Research

Quiñonero-Candela, J., & Rasmussen, C. E. (2005). *Journal of Machine Learning Research*, *6*(Dec), 1939-1959.

HARVARD School of Engineering and Applied Sciences

Gaussian process force fields

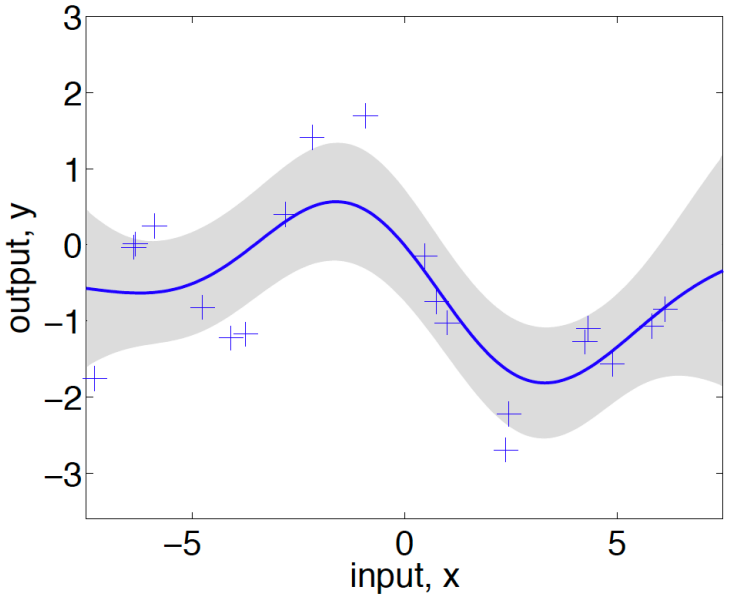
- We use Gaussian process regression to learn force fields from DFT force components [1].
- The GP model estimates force components $f_{i\alpha}$ on all atoms and their uncertainties $\sigma_{i\alpha}$.

Force prediction:

$$f_{i\alpha} = \sum_t \underbrace{k(\rho_i, \rho_t)}_{\text{comparison to training environments}} \alpha_t$$

Uncertainty estimate:

$$\sigma^2_{\text{tot},\, i\alpha} = \underbrace{k_{\alpha,\,\alpha}(\rho_i, \rho_i) - \bar{k}^T_{i\alpha}(K + \sigma_n^2 \mathrm{I})^{-1}\bar{k}_{i\alpha}}_{\text{epistemic uncertainty } \sigma^2_{i\alpha}} + \underbrace{\sigma_n^2}_{\text{noise}}$$

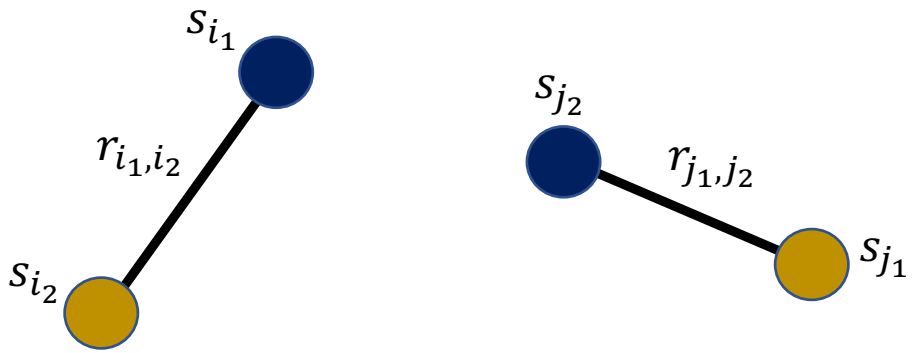


Williams, C. K., & Rasmussen, C. E. (2006). Cambridge, MA: MIT press.

[1] Bartók, A. P., Payne, M. C., Kondor, R., & Csányi, G. (2010). Gaussian approximation potentials: The accuracy of quantum mechanics, without the electrons. *Physical review letters*, *104*(13), 136403.

N-body covariant kernels

- Compares interatomic distances of two clusters if they're of the same type:



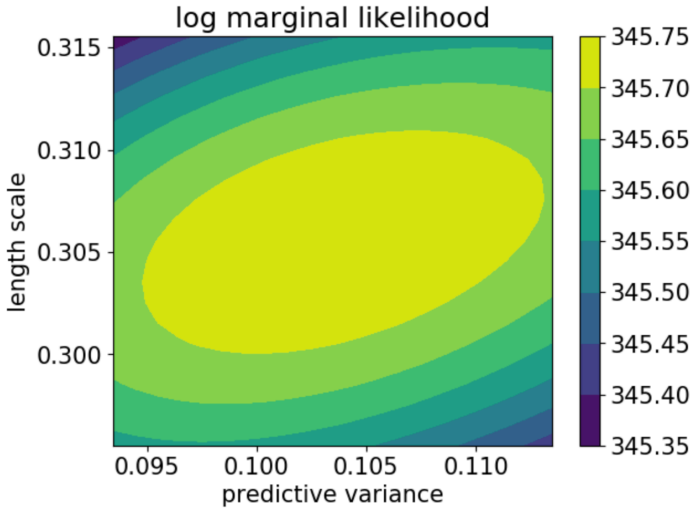
$$k_2 = \sigma^2 \delta_{s_{i_1},s_{j_1}} \delta_{s_{i_2},s_{j_2}} \exp\left(-\frac{(r_{i_1,i_2} - r_{j_1,j_2})^2}{2\,\ell^2}\right) f_{\text{cut}}(r_{i_1,i_2}, r_{j_1,j_2})$$

multi-element rotationally invariant smooth

- To model forces, we differentiate with respect to the Cartesian coordinates of the central atom.

N-body covariant kernels

- Hyperparameters are interpretable and can be tuned with an analytic likelihood.



Term	Interpretation
ℓ	Length scale of Gaussian prior
σ_s	Out-of-sample uncertainty
σ_n	Noise uncertainty

$$k_2 = \sigma^2 \delta_{s_{i_1}, s_{j_1}} \delta_{s_{i_2}, s_{j_2}} \exp\left(-\frac{(r_{i_1,i_2} - r_{j_1,j_2})^2}{2\,\ell^2}\right) f_{\text{cut}}(r_{i_1,i_2}, r_{j_1,j_2})$$

multi-element rotationally invariant smooth

GP uncertainties

- **Uncertainty estimate:** $\sigma^2_{\text{tot},\, i\alpha} = \sigma^2_{i\alpha} + \sigma^2_n$

uncertainty on force component α of atom i

epistemic

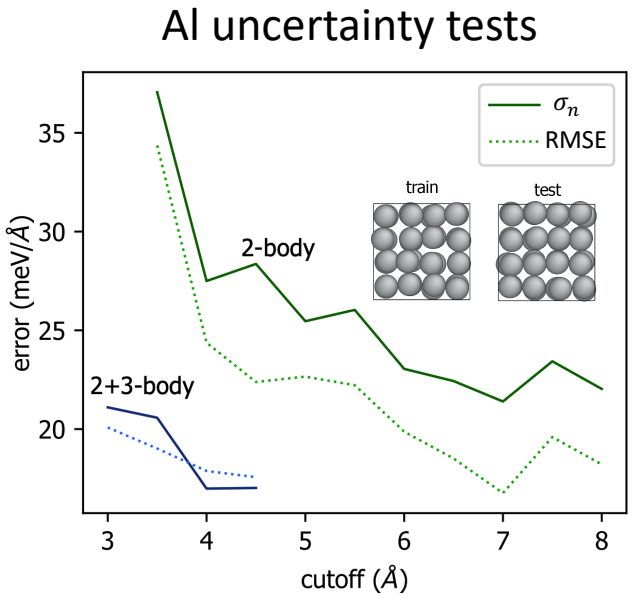
noise

GP uncertainties

- **Uncertainty estimate:** $\sigma^2_{\text{tot},\, i\alpha} = \sigma^2_{i\alpha} + \sigma^2_n$

  uncertainty on force component α of atom i — epistemic — noise

- **Noise uncertainty:** quantifies variation in the DFT forces that the model cannot capture.
  - Sources: long range forces, many-body interactions, unconverged DFT

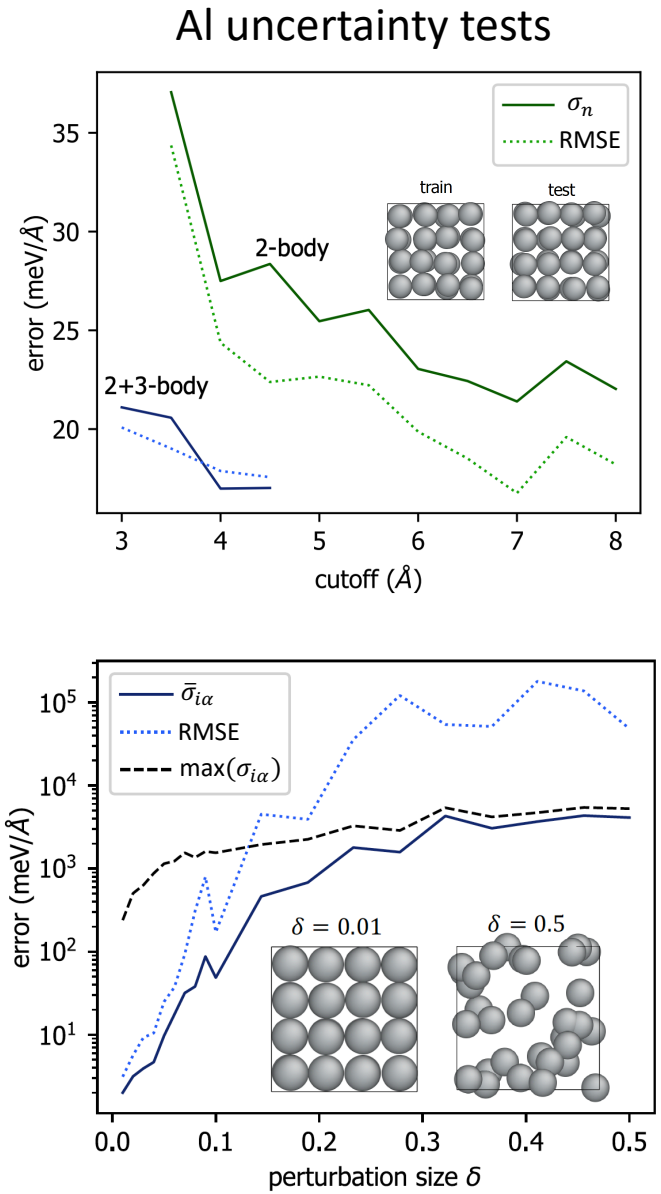


GP uncertainties

- **Uncertainty estimate:** $\sigma^2_{\text{tot},\,i\alpha} = \sigma^2_{i\alpha} + \sigma^2_n$

  uncertainty on force component α of atom i — epistemic — noise

- **Noise uncertainty:** quantifies variation in the DFT forces that the model cannot capture.
  - Sources: long range forces, many-body interactions, unconverged DFT
- **Epistemic uncertainty:** quantifies distance from the training set.

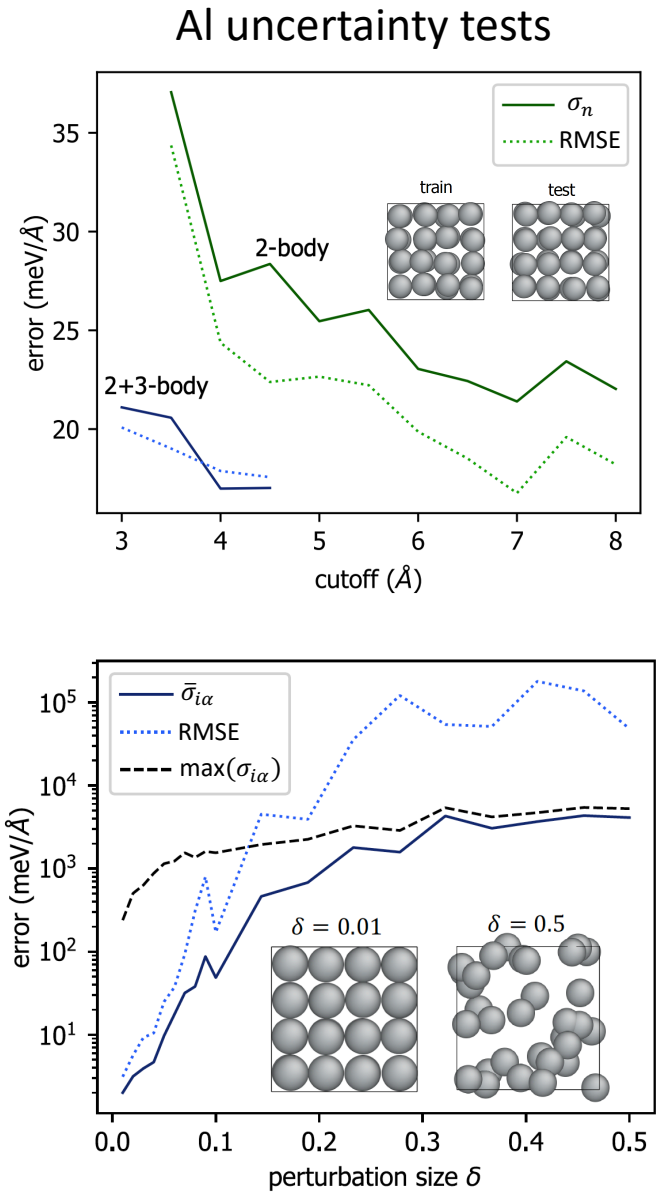


GP uncertainties

- **Uncertainty estimate:** $\sigma^2_{\text{tot},\,i\alpha} = \sigma^2_{i\alpha} + \sigma^2_n$

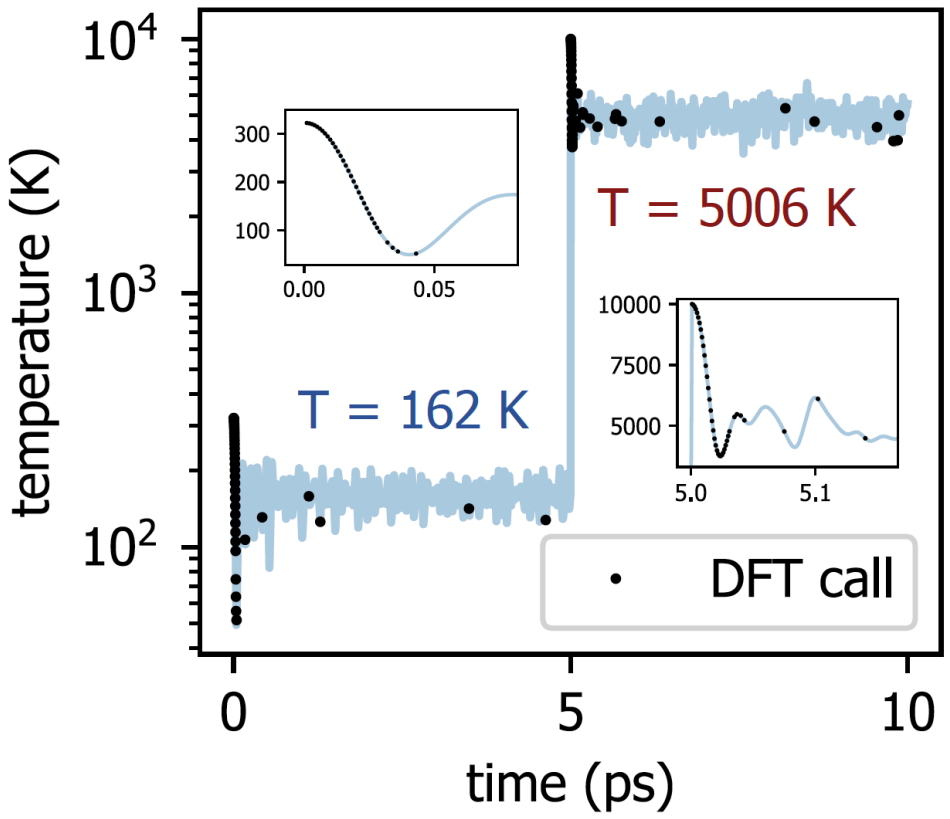
  uncertainty on force component α of atom i — epistemic — noise

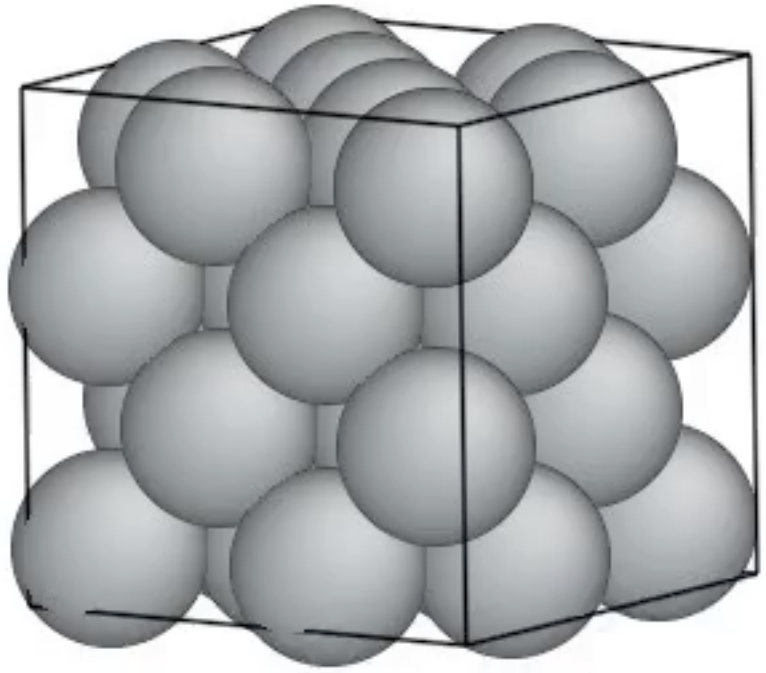
- **Noise uncertainty:** quantifies variation in the DFT forces that the model cannot capture.
  - Sources: long range forces, many-body interactions, unconverged DFT
- **Epistemic uncertainty:** quantifies distance from the training set.
- **Decision criterion:** When the epistemic uncertainty exceeds the noise uncertainty, more data is needed.



On-the-fly learning: aluminum melt

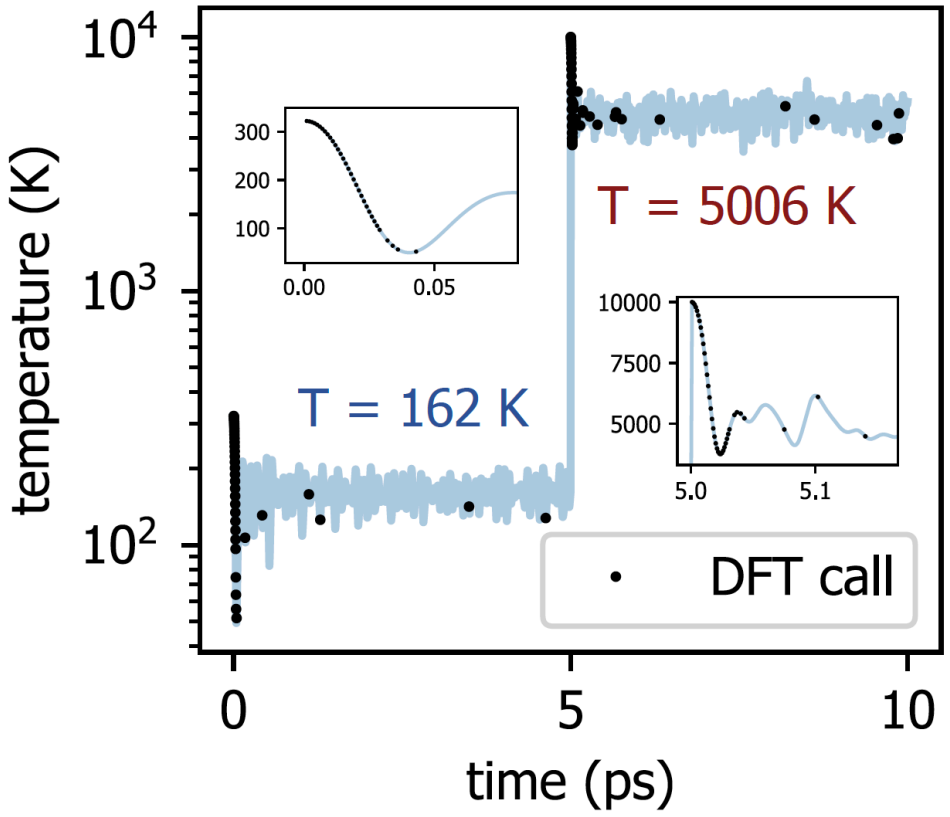
automatic learning of solid/liquid phases



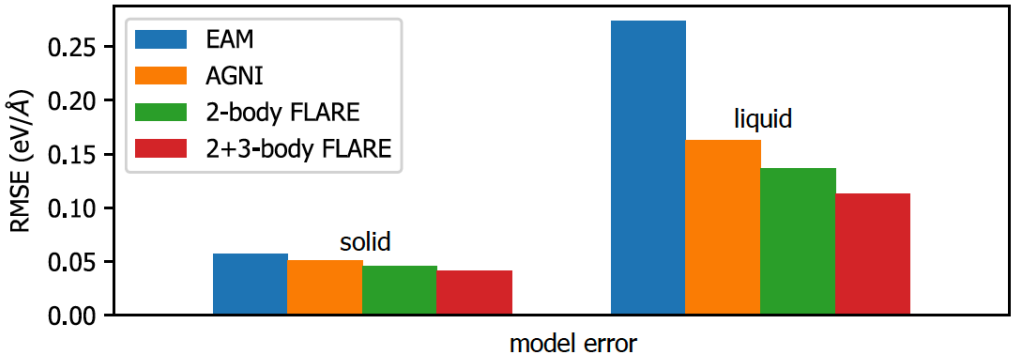


On-the-fly learning: aluminum melt

automatic learning of solid/liquid phases



accuracy

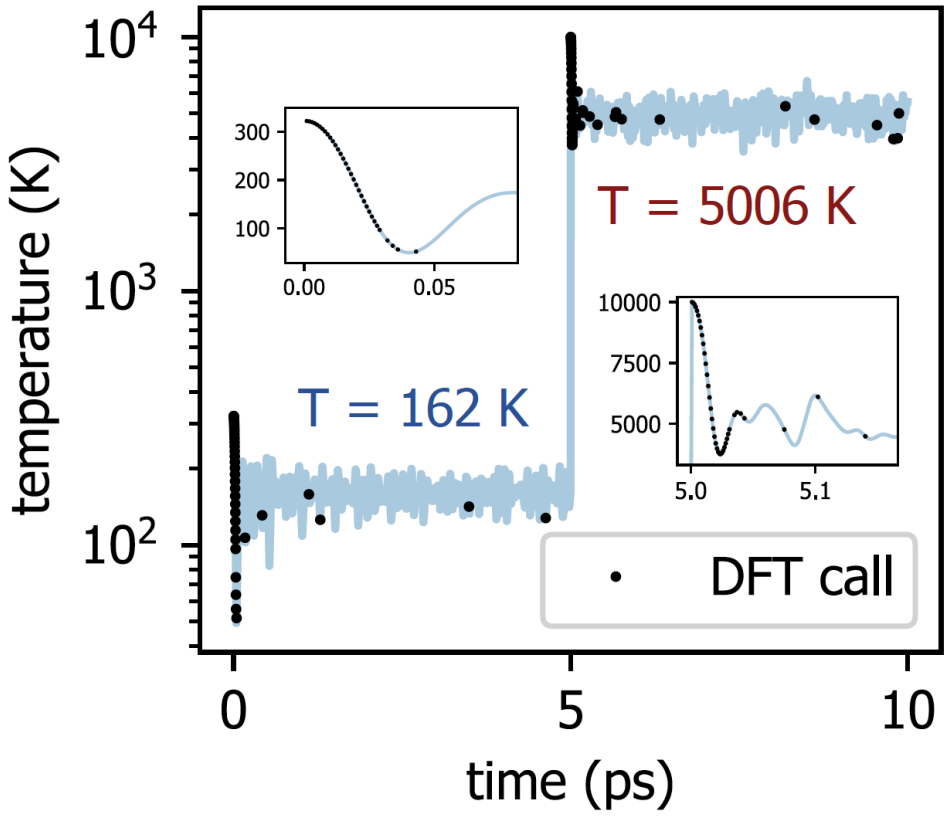


Materials Intelligence Research

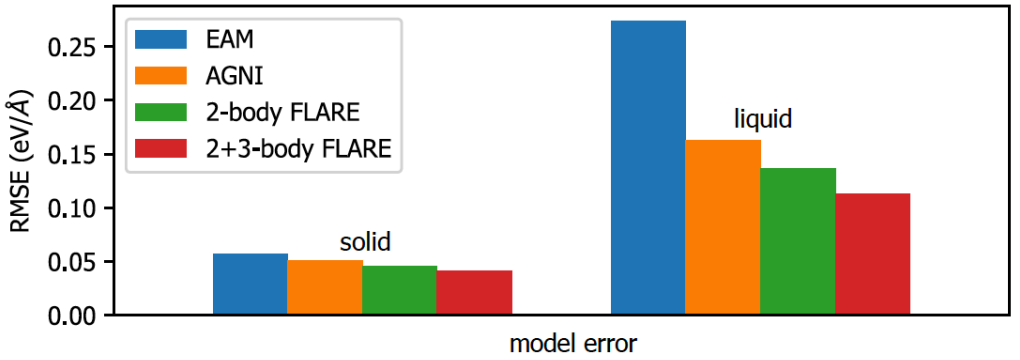
HARVARD School of Engineering and Applied Sciences

On-the-fly learning: aluminum melt

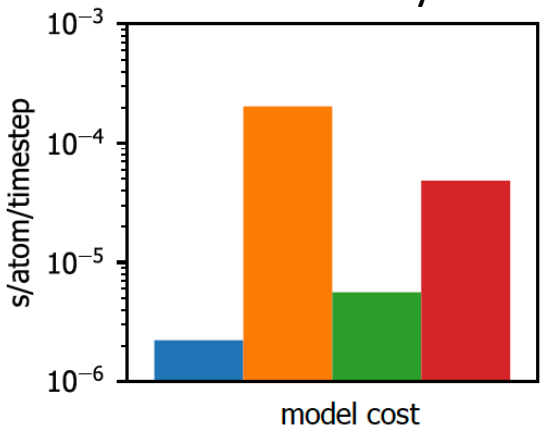
automatic learning of solid/liquid phases



accuracy



efficiency



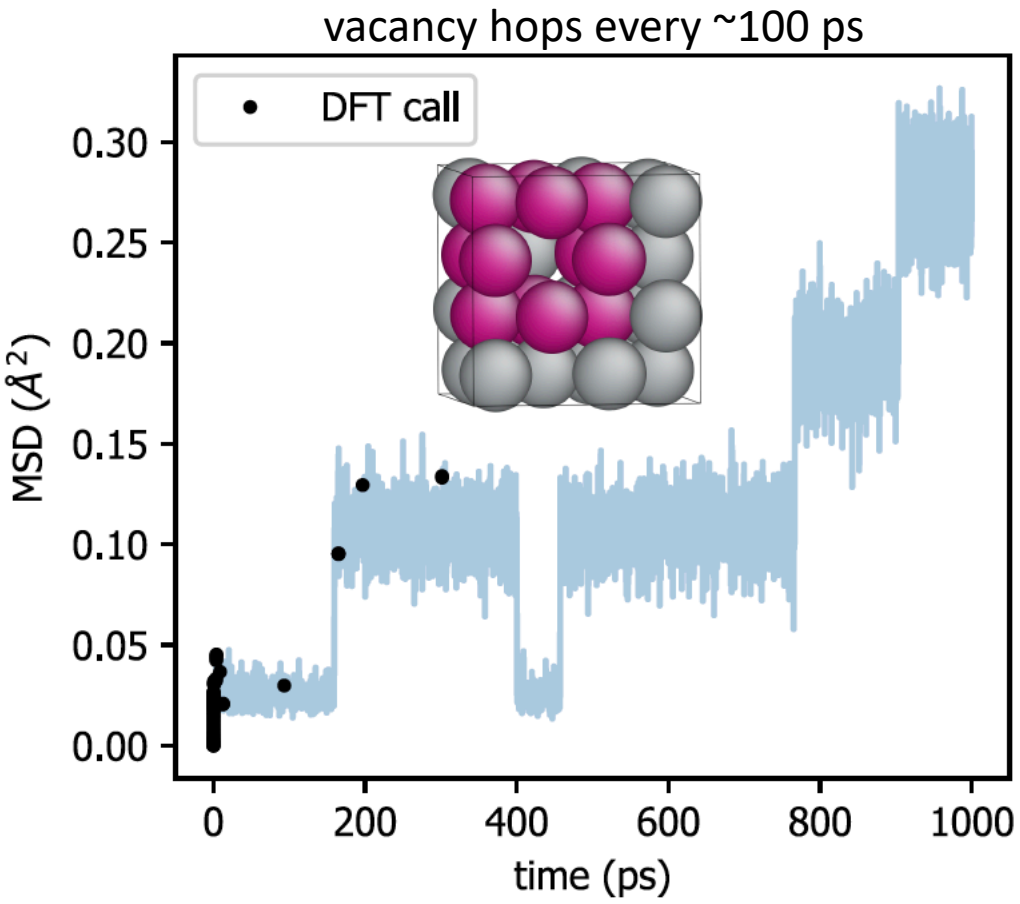
2-body:
5.6×10^{-6} s/atom/step

2+3-body:
4.9×10^{-5} s/atom/step

Materials Intelligence Research

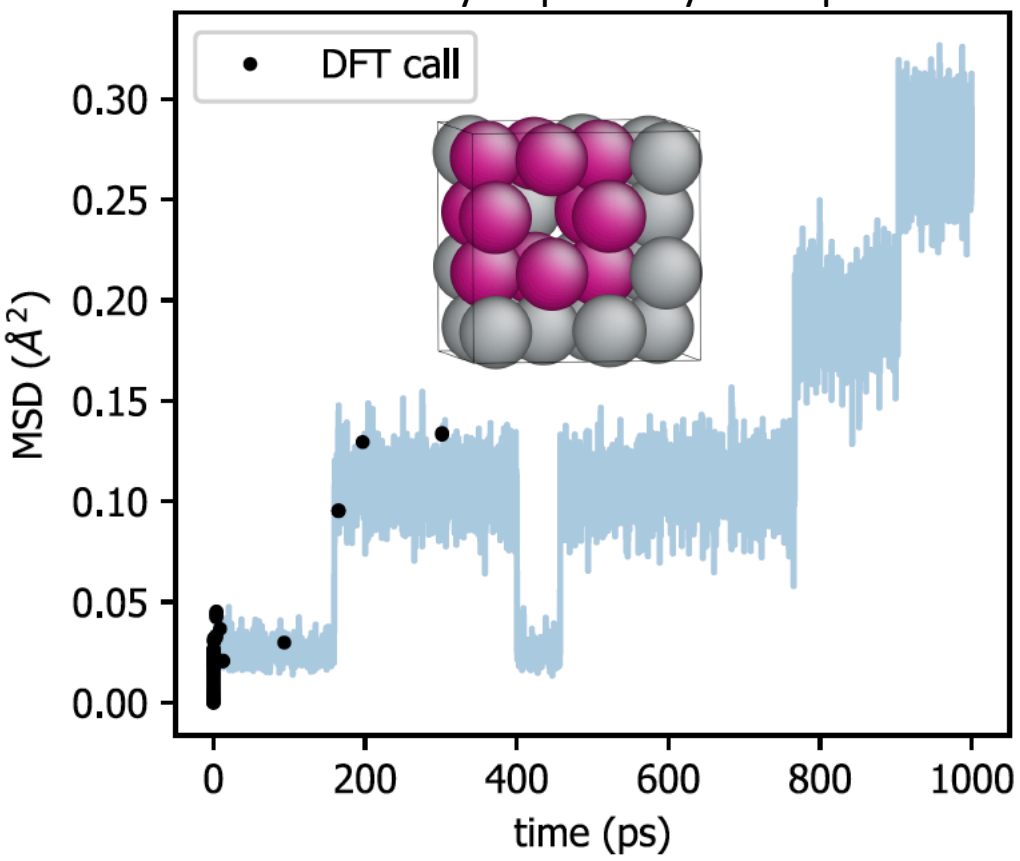
HARVARD School of Engineering and Applied Sciences

On-the-fly learning: Al vacancy diffusion

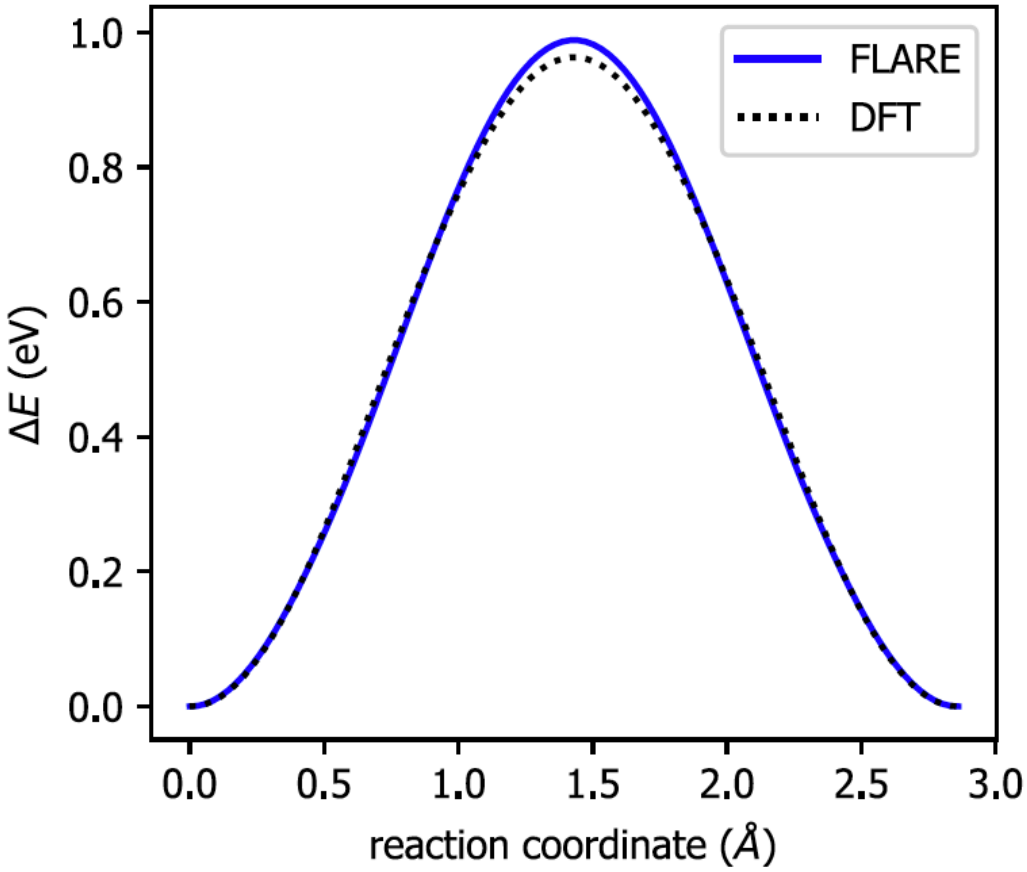


On-the-fly learning: Al vacancy diffusion

vacancy hops every ~100 ps



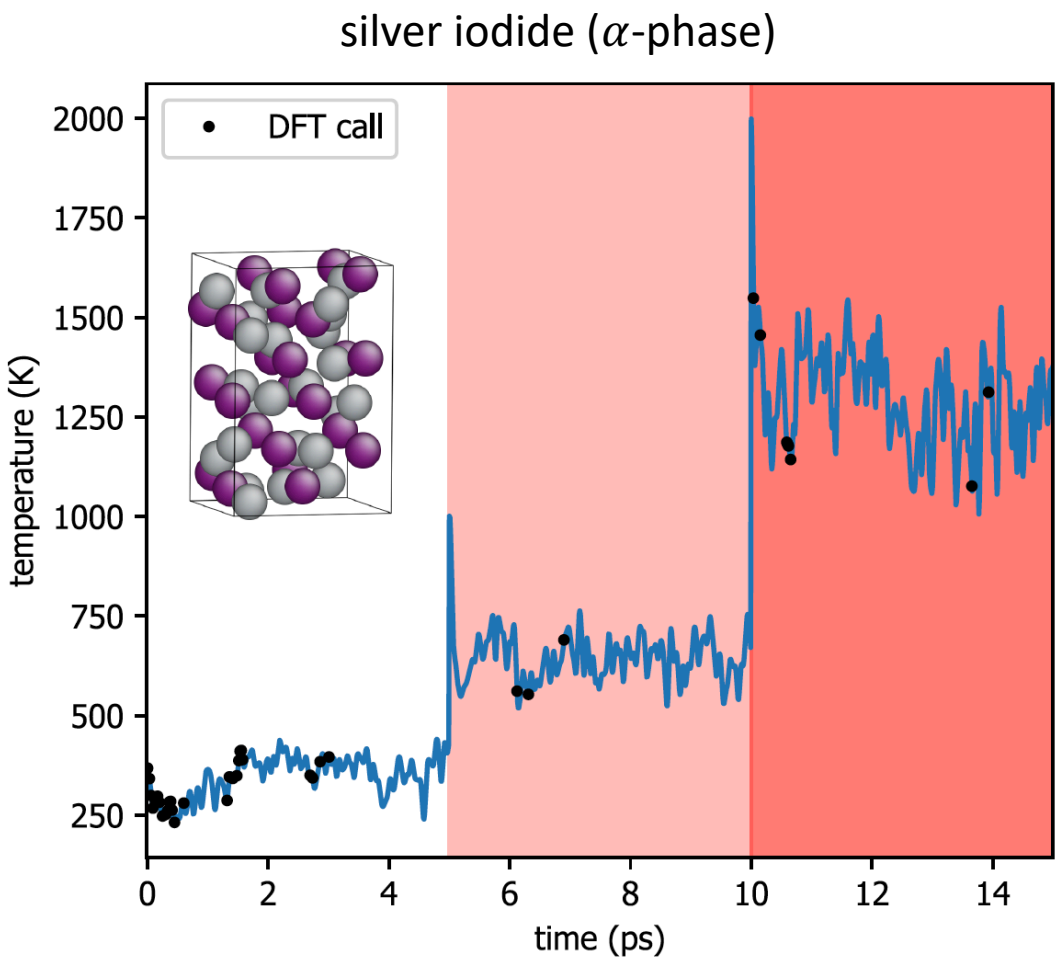
GP barrier matches DFT



Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

On-the-fly learning: fast-ion diffusion

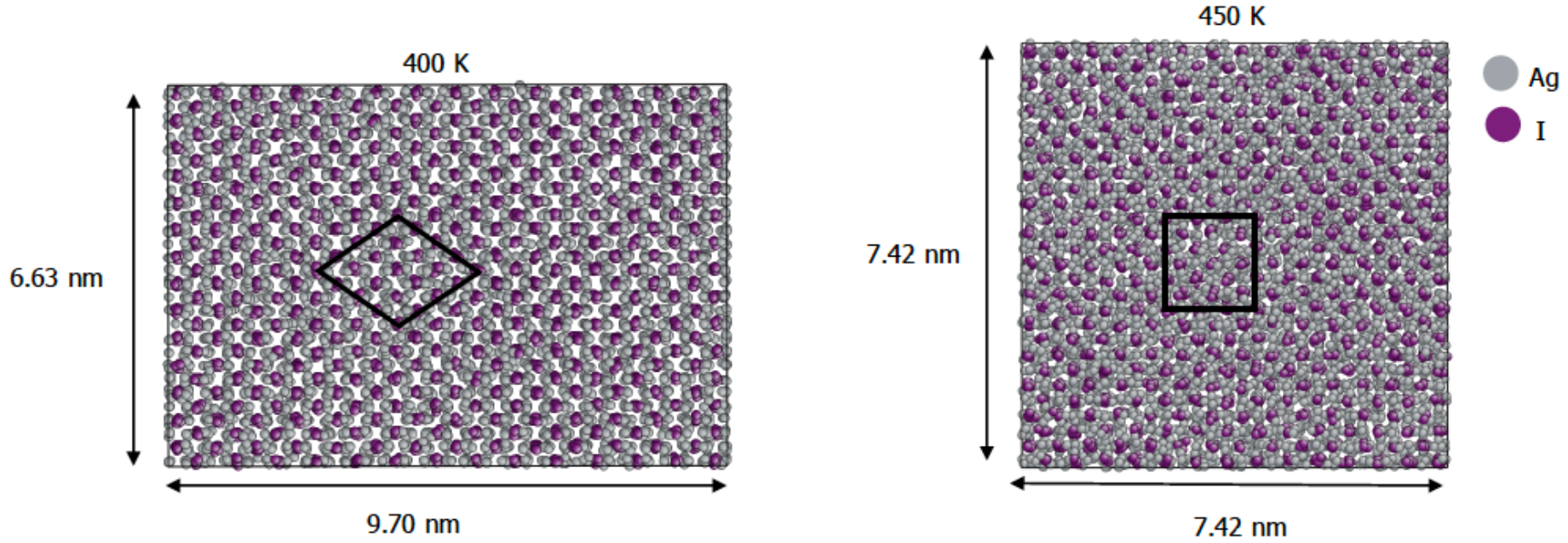


Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

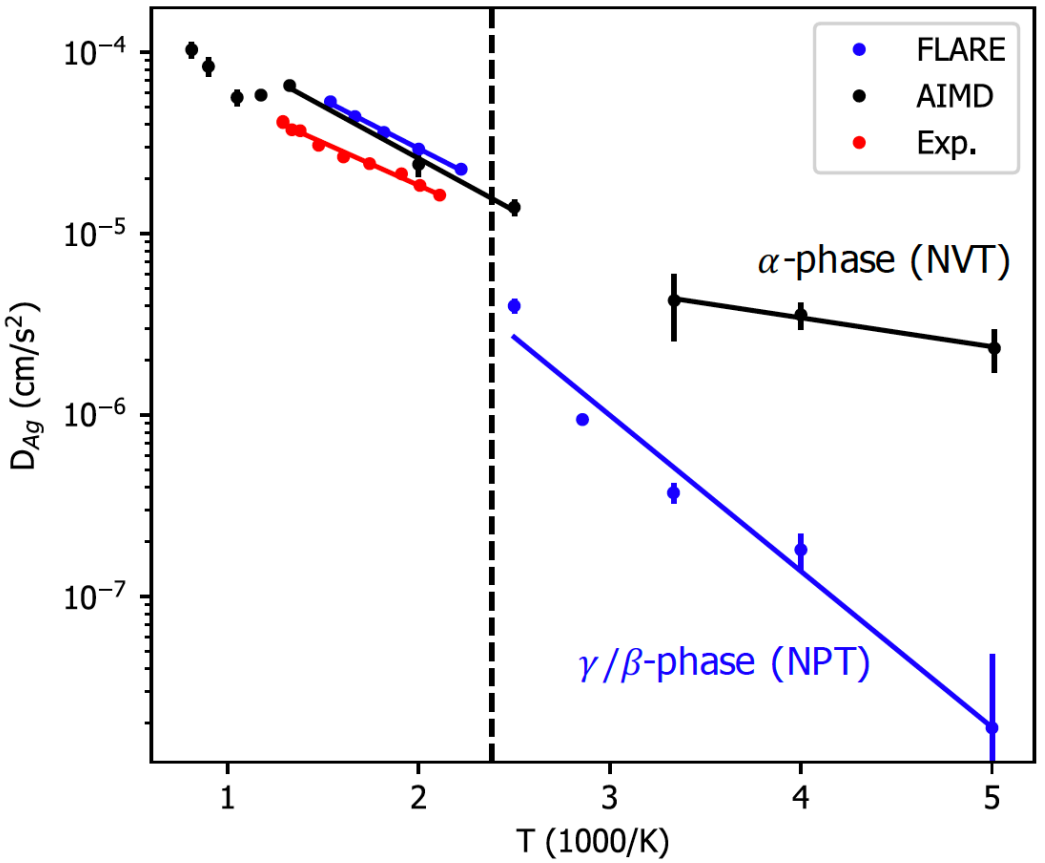
On-the-fly learning: fast-ion diffusion

structural phase transition correctly predicted

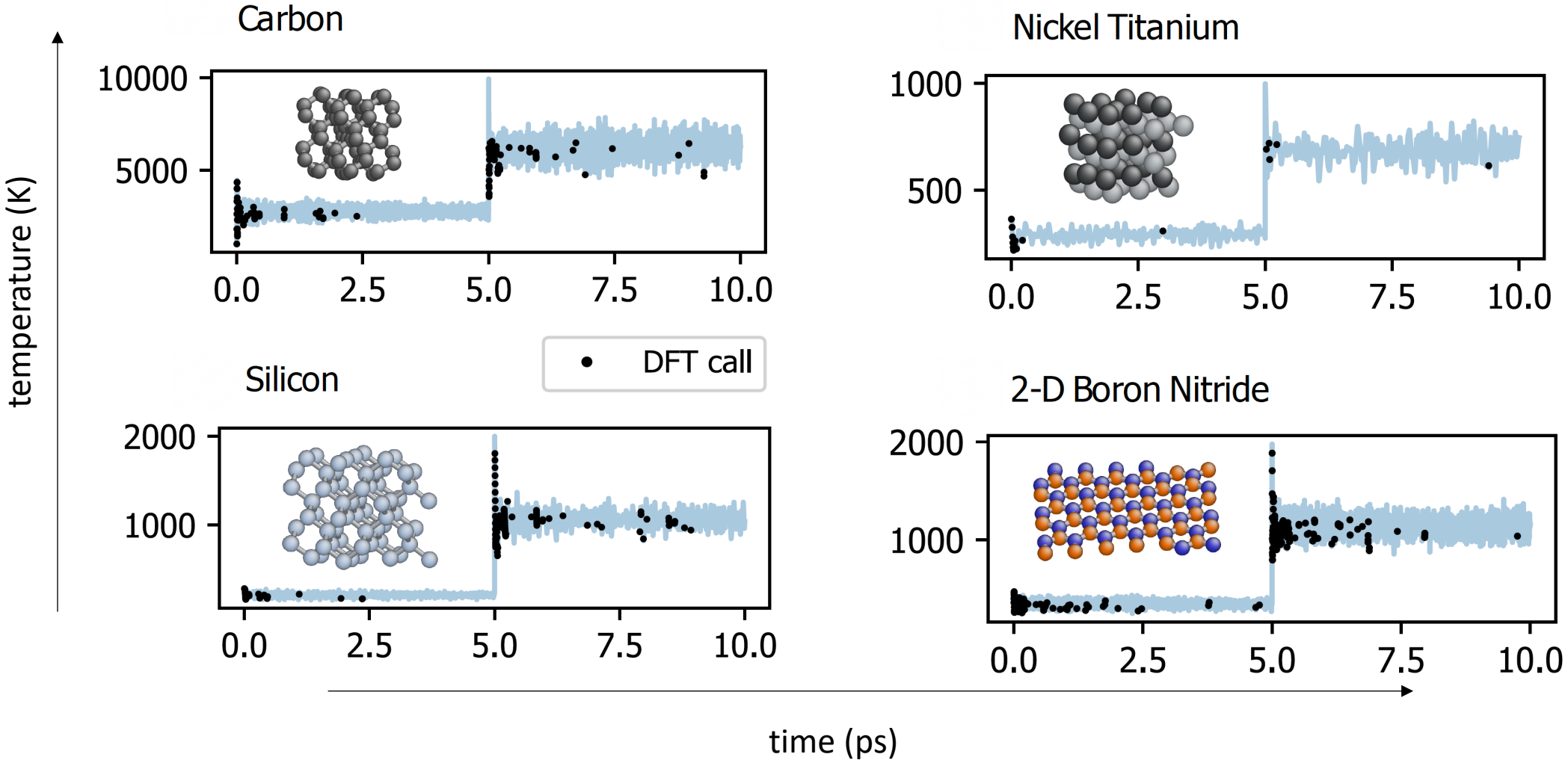


On-the-fly learning: fast-ion diffusion

diffusion coefficients match AIMD



On-the-fly learning: general applicability

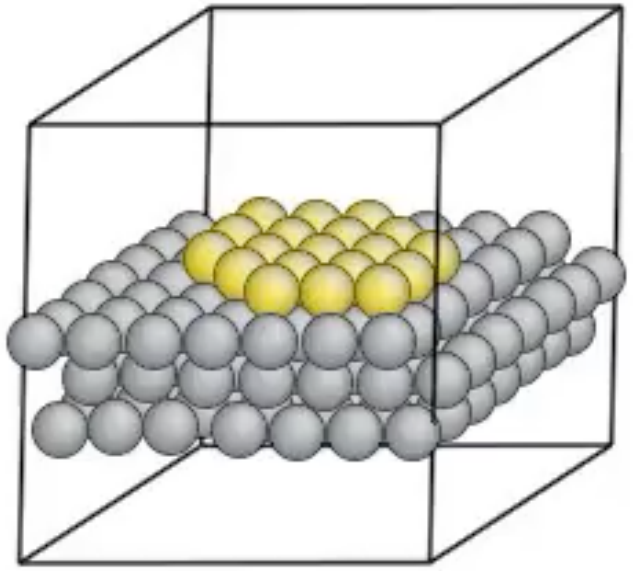


David Lim

Toward large scale applications: Pd/Ag

- How does palladium dissolve into a silver surface?
- Relevant to the selectivity/reactivity tradeoff in bimetallic catalysts

Training simulation



Training environments selected with automated, uncertainty driven active learning

# of Environments	# of CPUs	Wall Time
91	25	12.3 hours

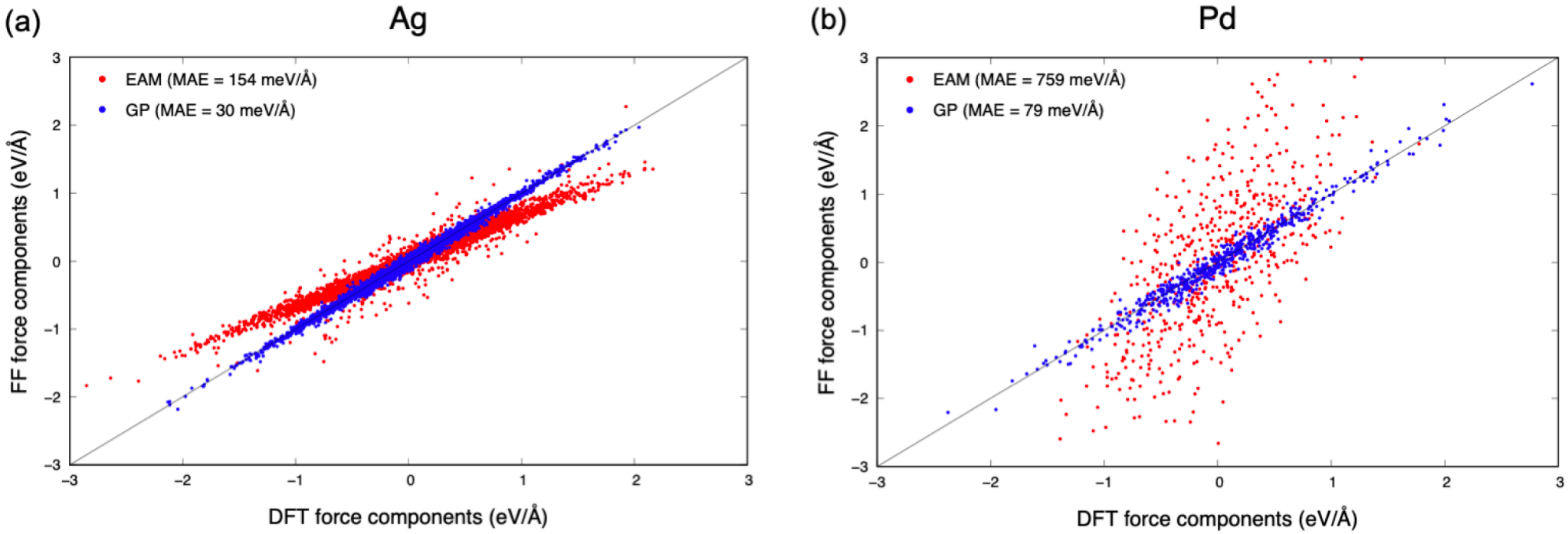
J.S. Lim, J. Vandermause, M.A. von Spronsen *et al.* chemRxiv: 11811660

Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences

Toward large scale applications: Pd/Ag

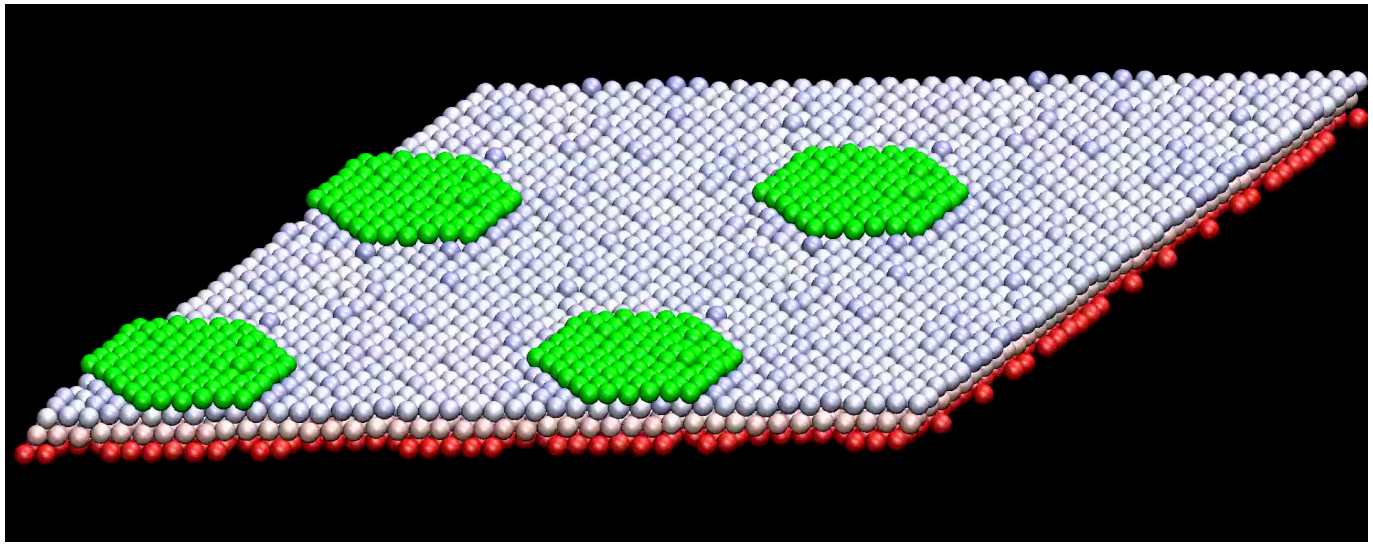
Off-the-shelf classical force fields are inadequate for this problem.



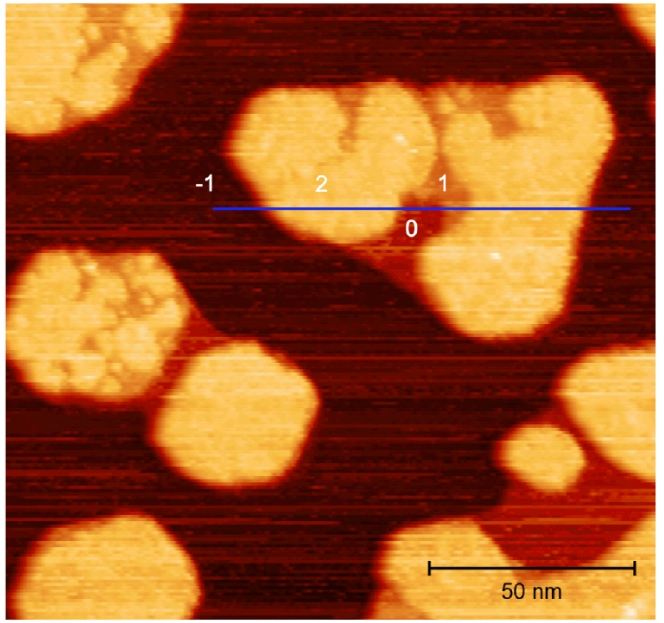
J.S. Lim, J. Vandermause, M.A. von Spronsen *et al.* chemRxiv: 11811660

Toward large scale applications: Pd/Ag

Production MD run T 500 K, 2 microseconds



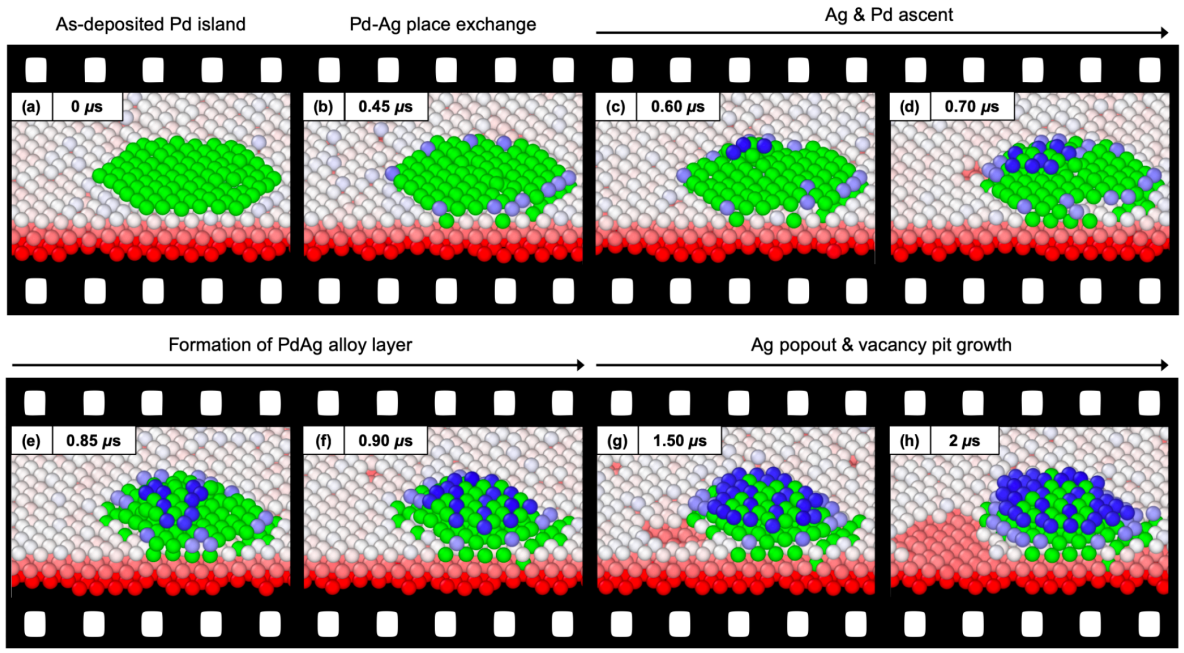
Vacancy pit growth consistent with STM images



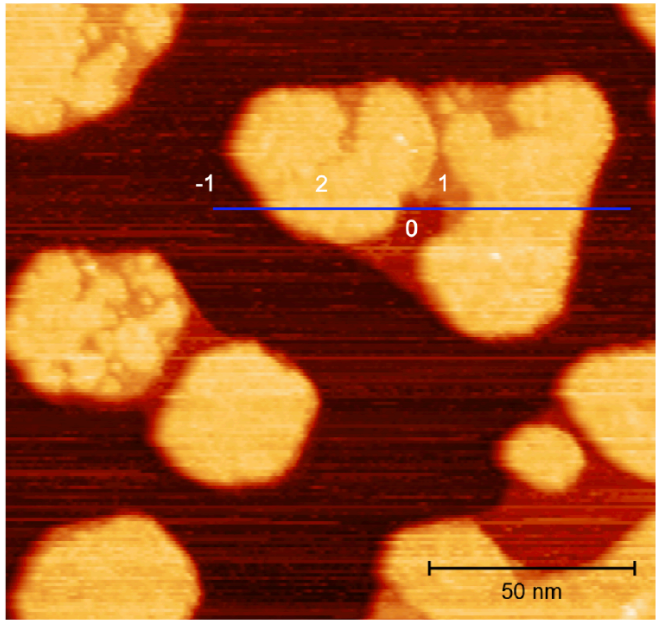
J.S. Lim, J. Vandermause, M.A. von Spronsen *et al.* chemRxiv: 11811660

Toward large scale applications: Pd/Ag

Production MD run T 500 K, 2 microseconds



Vacancy pit growth consistent with STM images

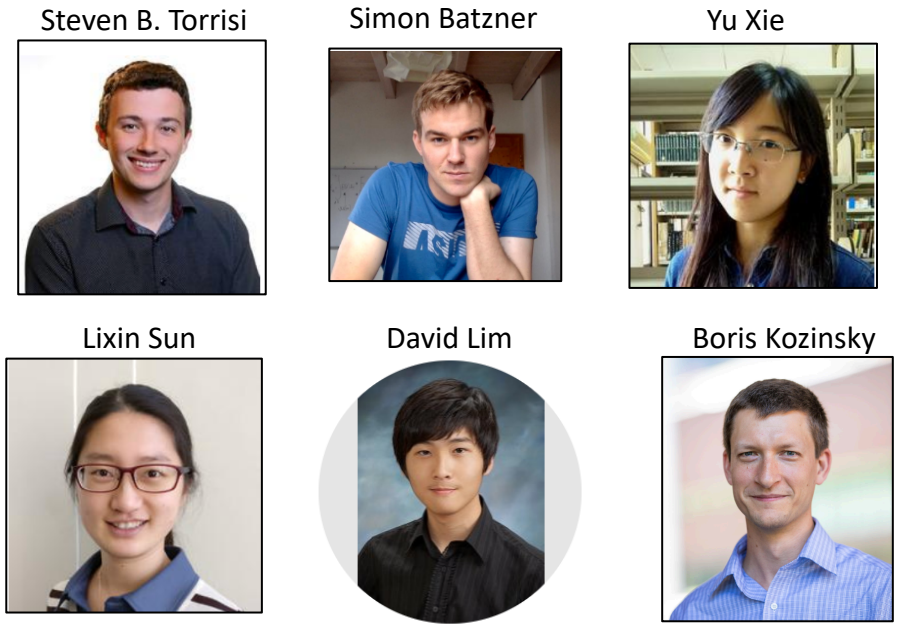


J.S. Lim, J. Vandermause, M.A. von Spronsen *et al.* chemRxiv: 11811660

Thank you for your attention!

Paper: nature.com/articles/s41524-020-0283-z
Github: github.com/mir-group/flare
Documentation: flare.readthedocs.io
Tutorial: tinyurl.com/wrq3kr7

The FLARE team



Vandermause, J., Torrisi, S. B., Batzner, S., Xie, Y., Sun, L., Kolpak, A. M., & Kozinsky, B. (2020). On-the-fly active learning of interpretable Bayesian force fields for atomistic rare events. *npj Computational Materials*, *6*(1), 1-11.

Materials Intelligence Research

HARVARD School of Engineering and Applied Sciences