Uncertainty Quantification in Atomistic Simulations with Dropout Neural Network Potentials

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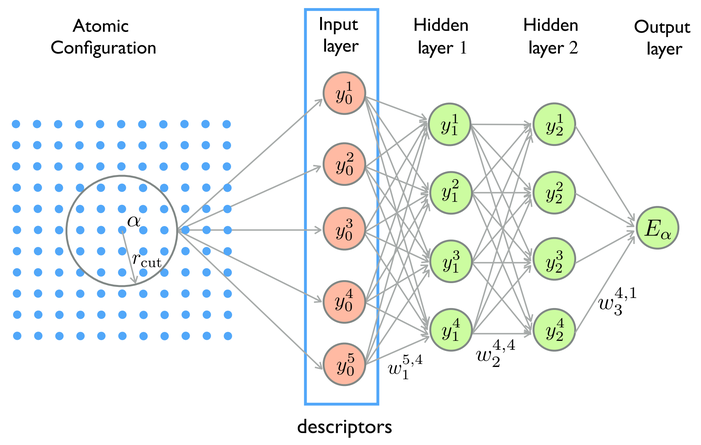
Objective

Create a class of interatomic potentials that can be used easily to

- determine the transferability (i.e. the ability to make appropriate predictions outside the training set) of the potential, i.e. the applicability to new problems of interest
- quantify the uncertainty in potential predictions and propagate the uncertainty to properties of interest obtained from atomistic simulations

Fully connected neural network (NN) potential



An NN potential with two hidden layers

Potential energy:

$$\mathcal{V}(\boldsymbol{r};\boldsymbol{\theta}) = \sum_\alpha E_\alpha$$

$$y_i^j = g\left(\sum_k y_{i-1}^k w_i^{k,j} + b_i^j\right)$$

$$E_\alpha = g(g(\boldsymbol{y}_0\boldsymbol{W}_1 + \boldsymbol{b}_1)\boldsymbol{W}_2 + \boldsymbol{b}_2)\boldsymbol{W}_3 + \boldsymbol{b}_3$$

g: nonlinear function

$\boldsymbol{W}, \boldsymbol{b}$: parameters

Requirement of descriptors: translation, rotation, inversion, and permutation symmetric

Symmetry functions

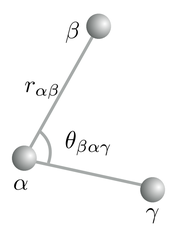
$$y_0^j = \phi^j(\boldsymbol{r})$$

two-body descriptor (bond stretching)

$$\phi_1(\boldsymbol{r}) = \sum_{\beta\neq\alpha} \exp[-\eta(r_{\alpha\beta} - R)^2] \cdot f_c(r_{\alpha\beta})$$

three-body descriptor (bond bending)

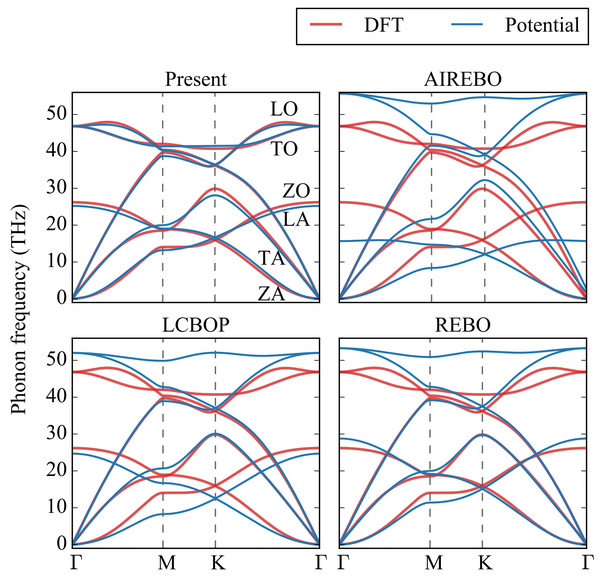
$$\phi_2(\boldsymbol{r}) = 2^{1-\zeta} \sum_{\beta\neq\alpha} \sum_{\gamma\neq\alpha,\beta} (1 + \lambda\cos\theta_{\beta\alpha\gamma})^\zeta \exp[-\eta(r_{\alpha\beta}^2 + r_{\alpha\gamma}^2 + r_{\beta\gamma}^2)] \cdot f_c(r_{\alpha\beta}) \cdot f_c(r_{\alpha\gamma}) \cdot f_c(r_{\beta\gamma})$$



Behler, *J. Chem. Phys.* 134, 074106, 2011

Phonon dispersions

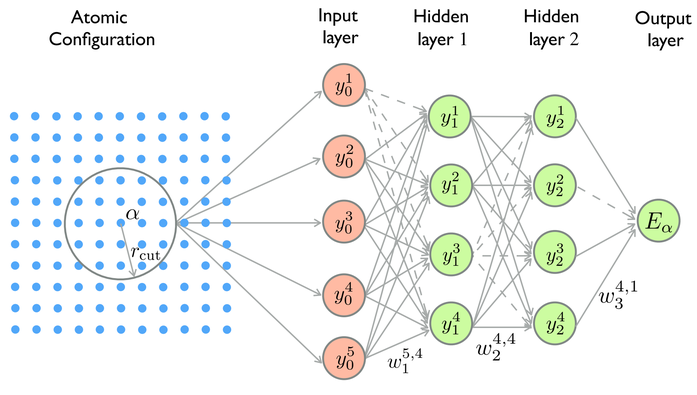
- Phonon dispersions for monolayer graphene
- NN potential training set:
  - (1) monolayer graphene (stretched, compressed, and vacancies)
  - (2) bilayer graphene (different layer spacing, translated, and twisted)
  - (3) graphite (different layer spacing)
- Fully connected NN potential (Present) performs much better than other tested physics-motivated potentials (AIREBO, LCBOP, and REBO), especially for the high-frequency optical modes



Dropout neural network (NN) potential

Problems of fully connected NN potential:

- Low transferability
- Not easy to carry out uncertainty quantification and propagation



A dropout NN potential with two hidden layers (dashed arrows indicate dropped connections)

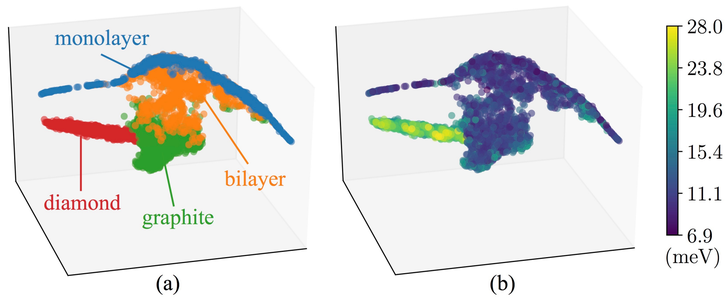
- All the same as a fully connected NN potential except trained with dropout (randomly remove a proportion of connections between adjacent layers at each training step)
- An NN trained with dropout is mathematically equivalent to a Bayesian NN
- At predicting stage, in practice, one only needs to evaluate the dropout NN potential multiple times (each with a different realization of the dropout) and then obtain the predictive mean and uncertainty from these samples

Atomic energy: $E_\alpha = \bar{E}_\alpha = \frac{1}{P}\sum_p E_\alpha^p$ (sample average)

Uncertainty: $\sigma_{E_\alpha} = \sqrt{\frac{\sum_p (E_\alpha^p - \bar{E}_\alpha)^2}{P-1}}$ (sample standard deviation)

Wen and Tadmor, in preparation; Gal, Ph.D. Thesis, Cambridge University, 2016

Transferability determination

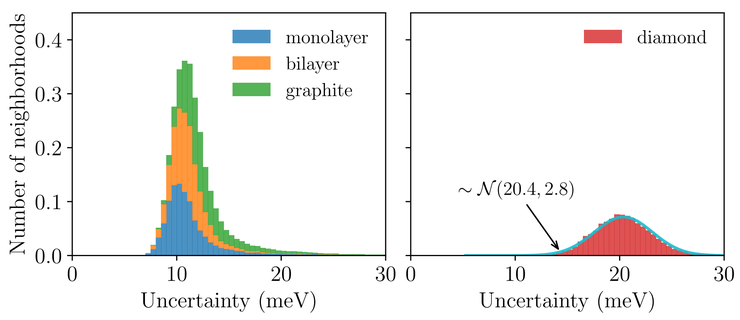


Representations of the carbon local atomic neighborhoods by UMAP

Uncertainty in atomic energy

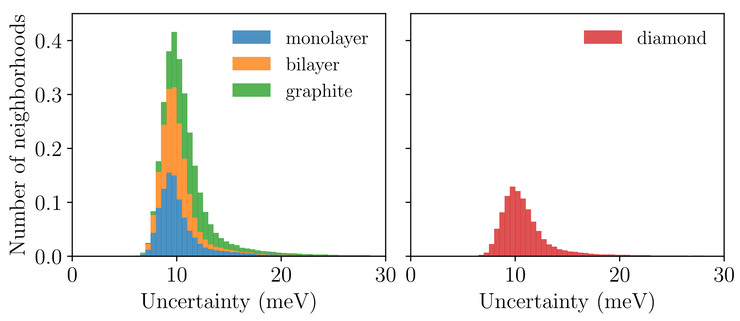
- UMAP (uniform manifold approximation projection) on local neighborhoods of individual atoms (i.e. descriptor values of atoms)
- Monolayer, bilayer, and graphite in the training set; diamond NOT in the training set
- Configurations not in the training set (diamond) have much higher uncertainty

A representation of the above uncertainty in atomic energy (diamond has larger uncertainty since it is not in the training set)



Histogram of uncertainty (diamond not in training set)

Uncertainty in atomic energy of diamond decreases once it is added to the training set



Histogram of uncertainty (diamond in training set)

Compare the uncertainty in configurations that characterize the problem of interest and the uncertainty in the training set to determine the transferability.

Uncertainty in stress

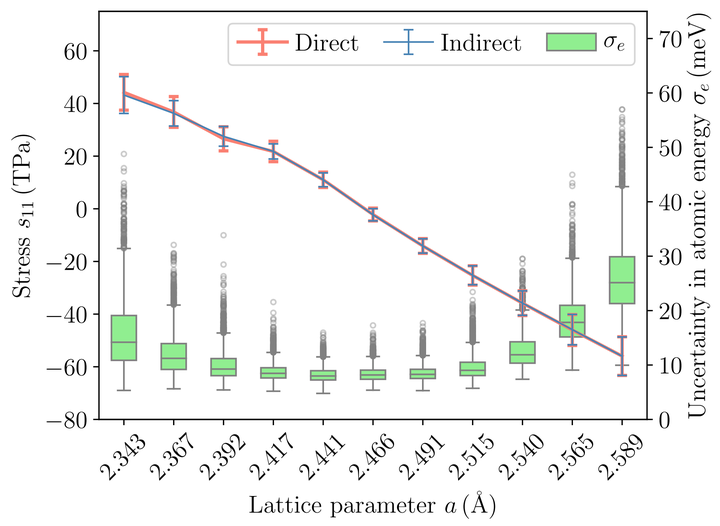
Potential part of the virial stress: $s_{ij} = \frac{1}{VT}\sum_{t=1}^{T}\sum_{\alpha=1}^{N} r_{i,t}^\alpha f_{j,t}^\alpha$

V: volume T: # MD steps N: # atoms
r: coordinates f: forces

Direct method: compute multiple samples of the stress (each with a different but fixed dropout), and then obtain the mean and uncertainty

Indirect method: compute mean and uncertainty in the forces, and then propagate the uncertainty to the stress

Training set lattice parameter range: [2.40, 2.52] Å



Potential part of the 11 component of the virial stress in a monolayer graphene (red and blue error bar plot), and uncertainty in atomic energy (green box plot) at various lattice parameters

Uncertainty in stress correlates with the uncertainty in atomic energy

Uncertainty in a property can be obtained from both the direct method and indirect method. The former works for any property, while the latter works when there exists a "simple" relationship between the property and energy (or forces). However, the latter is computationally much cheaper.

Uncertainty in phonon dispersions

- Phonon dispersions for monolayer graphene
- The dashed lines denote predictive mean, and the red, green, and blue bands denote uncertainty in the phonon frequency; obtained using the direct method
- The results by the dropout NN potential are slightly worth than the fully connected NN potential, but are still better than REBO; the dropout NN potential provides uncertainty in the predictions