Structure relaxation using Kernel Ridge Regression

Malthe Kjær Bisbo

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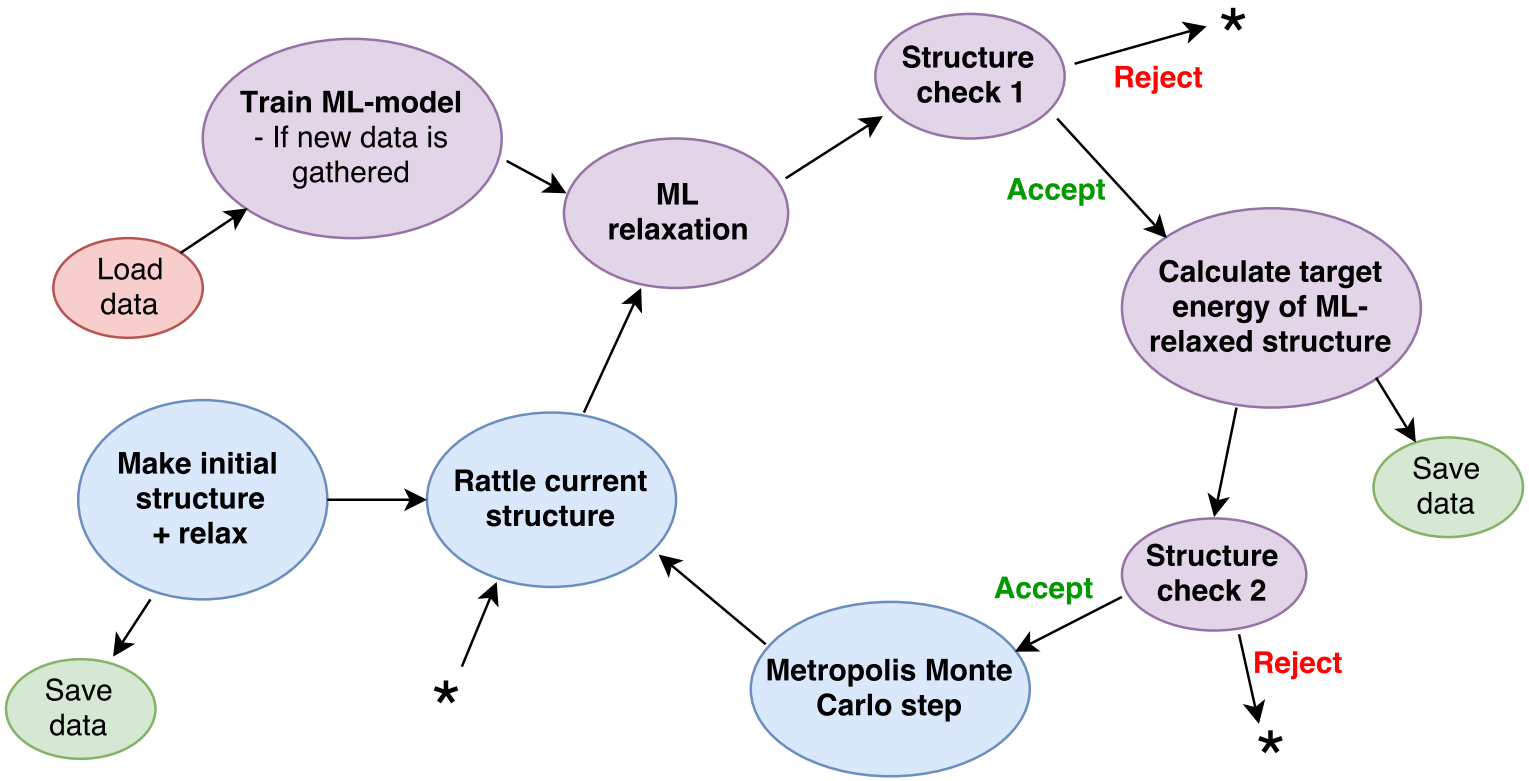
Enhancing global search



Structure check 1: Based on the ML prediction error.

Structure check 2: Based the agreement with the target energy of the relaxed structure

Kernel Ridge Regression

Minimize reguralized least squares error

$$\sum_{i}^{N}(E_i'(\vec{x}_i) - E_i)^2 + \lambda \sum_{i}^{N} w_i^2$$

Defining $\vec{w} = \mathbf{X}^T\vec{\alpha}$, where $\mathbf{X}^T = (\vec{x}_1, \vec{x}_2, ..., \vec{x}_N)$ and using the *kernel trick*, the kernalized solution for the α's is

$$\vec{\alpha} = (\mathbf{K} + \lambda I)^{-1}\vec{E} \qquad \text{with} \quad \mathbf{K}_{ij} = k(\vec{x}_i, \vec{x}_j) \quad (\textit{Training})$$

From which the energy of a new point $\vec{x'}$ is given as

$$E'(\vec{x'}) = \vec{\alpha}^T\vec{\kappa} \qquad \text{with} \quad \kappa_i = k(\vec{x'}, \vec{x}_i) \quad (\textit{Prediction})$$

Kernel

Gaussian kernel

$$k(\vec{x'}, \vec{x}) = \exp\left(-\frac{d(\vec{x'}, \vec{x})^2}{2\sigma^2}\right)$$

with the 2-norm for the dissimilarity d.

Feature

For each atomic type combination, the feature is a sum over the gausian-smoothed radial distributions of each atom.

$$F_{A,B}(R) = \sum_{A_i, B_j} \frac{\delta(R - R_{ij})}{4\pi R_{ij}^2 \Delta\left(\frac{N_A N_B}{V}\right)} - 1$$

The features for each atomic type combination is concatenated.

KRR predictions

Predicting the gradient

$$E'(\vec{x'}) = \vec{\alpha}^T \vec{\kappa} = \sum_i^N \alpha_i k(\vec{x'}, \vec{x_i})$$

$$F'(\vec{x'}) = -\frac{\partial E}{\partial \vec{r'}} = -\sum_i^N \alpha_i \frac{\partial k(\vec{x'}, \vec{x_i})}{\partial \vec{r'}}$$

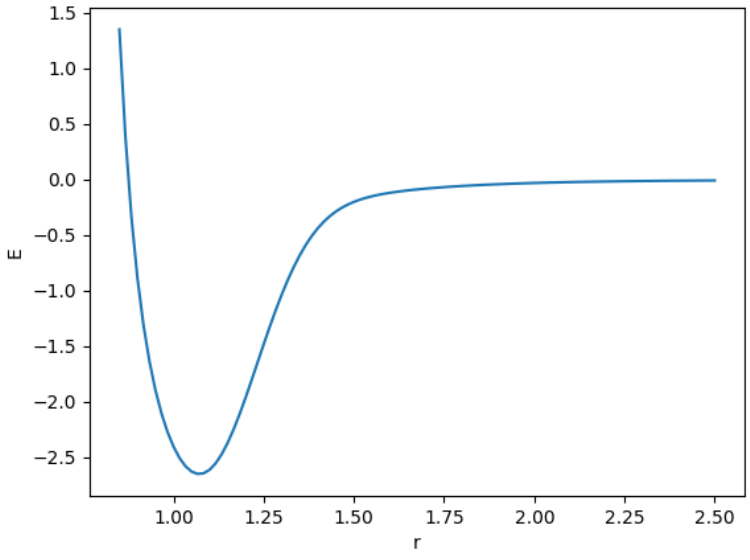
Prediction error

$$\mathrm{err}_{KRR}(\vec{x'}) = \sqrt{\left|\theta_0(1 - \kappa(\vec{x'}) \cdot \alpha_{err}(\vec{x'}))\right|}$$

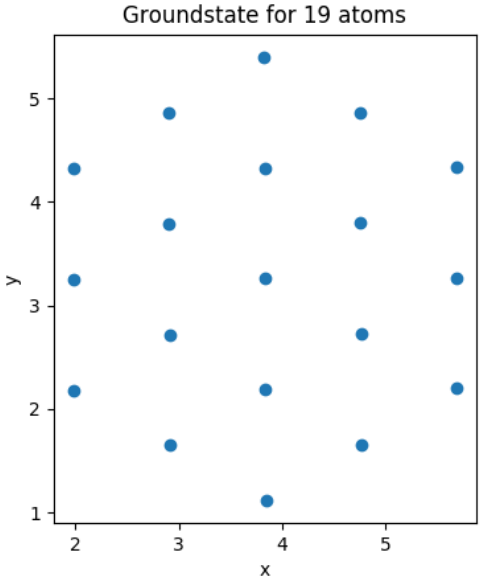
where

$$\alpha_{err}(\vec{x'}) = (\mathbf{K} + \lambda \mathbb{1})^{-1} \kappa(\vec{x'}) \quad \text{and} \quad \theta_0 = \frac{\vec{y} \cdot \vec{\alpha}_1}{N_{train}}$$

Model system - "Double" Lennard Jones



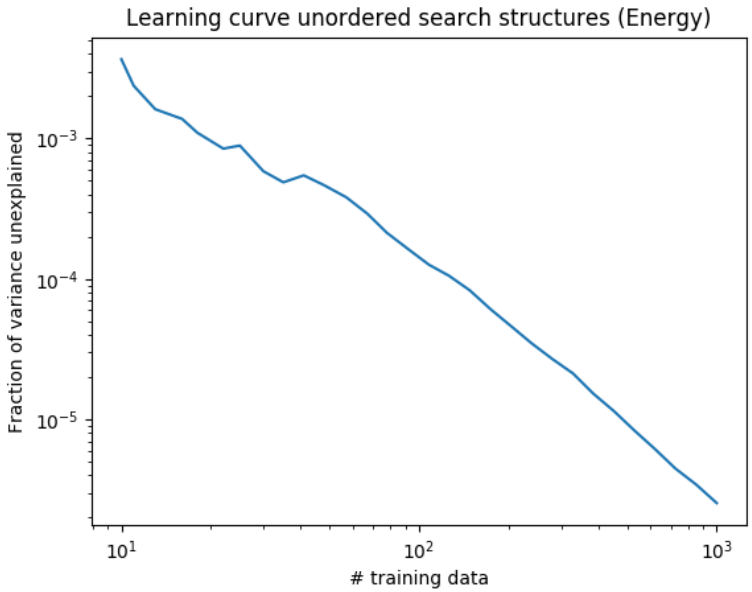
Interaction potential

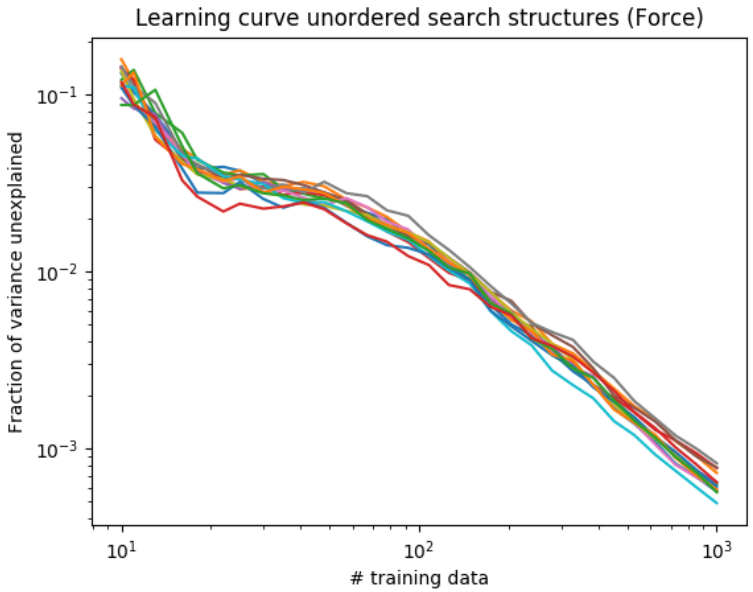


Ground state for 19 atoms

Learning Curves

Structures with 7 atoms





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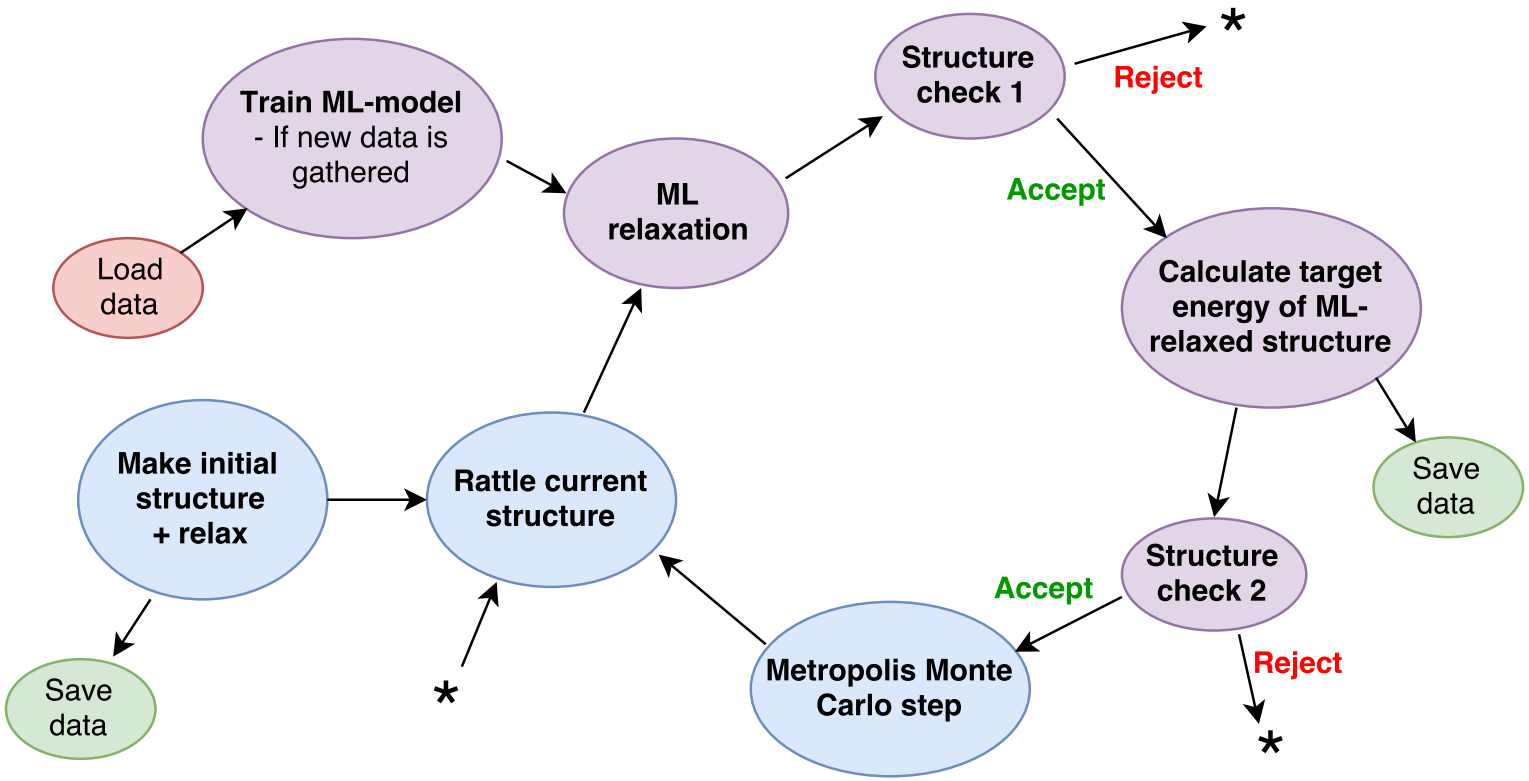

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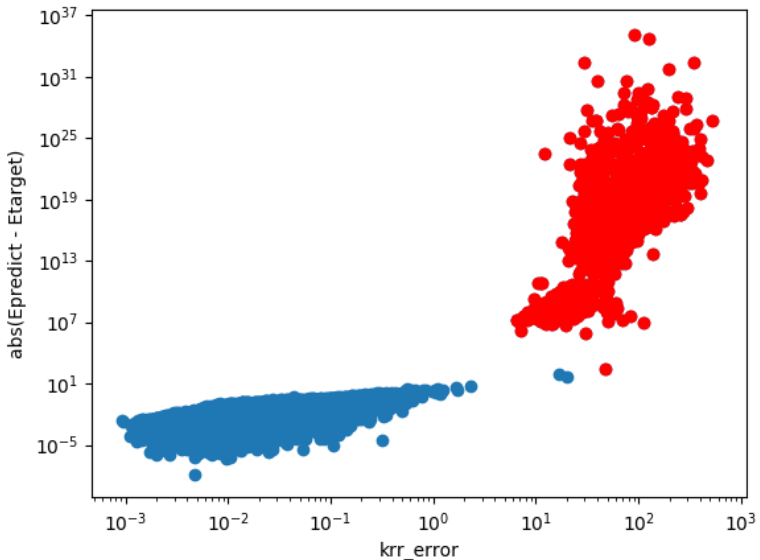
Structure check - prediction error

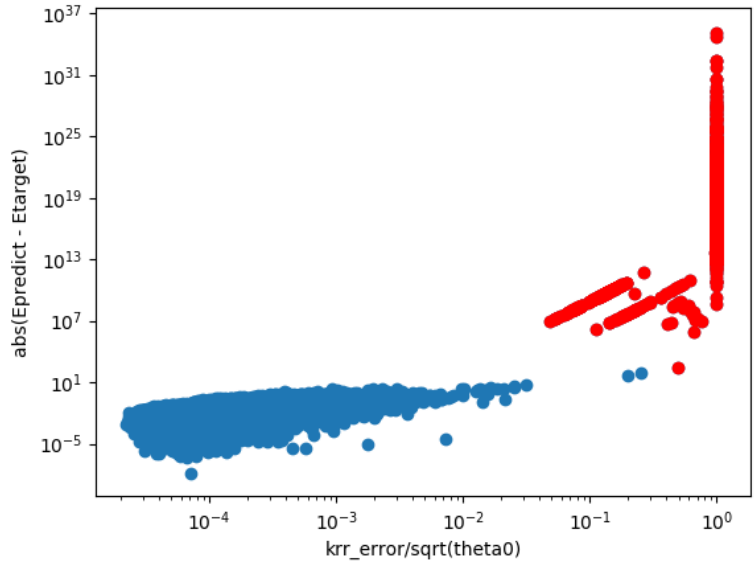
Filtering unresonable ML-relaxed structures

Rejection criteria:

$$\text{err}_{KRR} > ..$$

$$\frac{\text{err}_{KRR}}{\sqrt{\theta_0}} > ..$$





Search results

Structures with 19 atoms