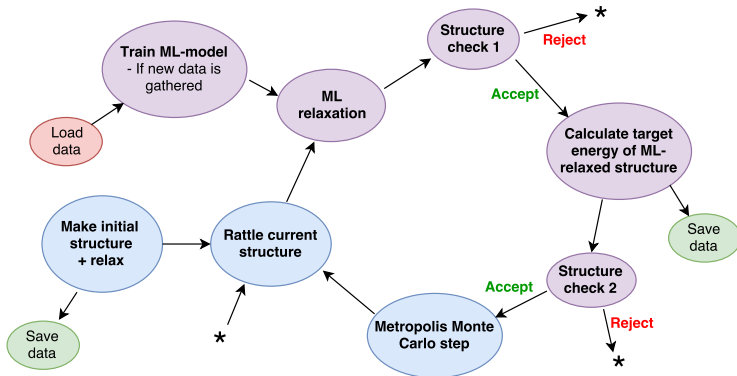


# Structure relaxation using Kernel Ridge Regression

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

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

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

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

## 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 gaussian-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

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

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

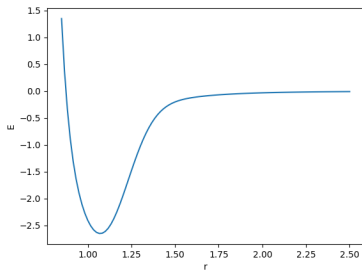
## Prediction error

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

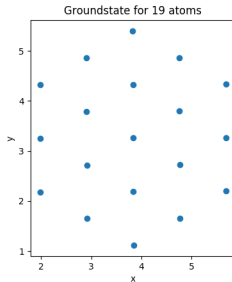
where

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

# Model system - "Double" Lennard Jones



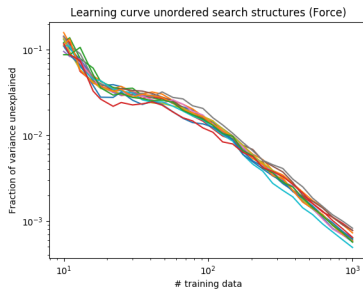
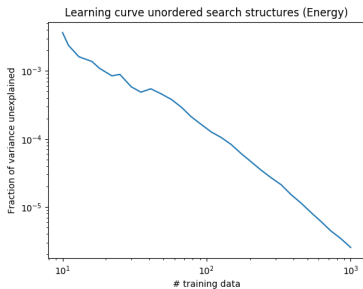
Interaction potential



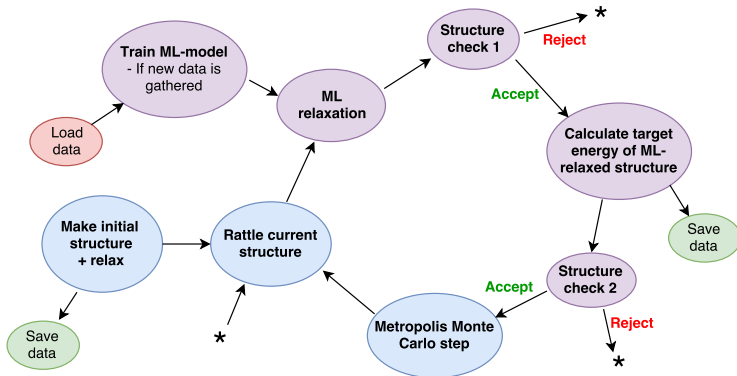
Ground state for 19 atoms

# Learning Curves

## Structures with 7 atoms



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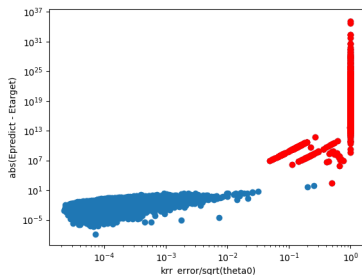
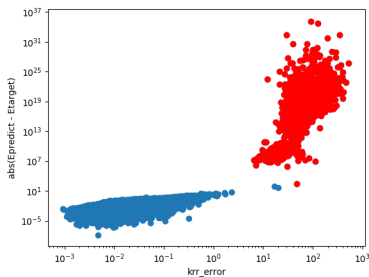
# Structure check - prediction error

Filtering unreasonable ML-relaxed structures

Rejection criteria:

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

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



# Search results

## Structures with 19 atoms

