# Accurate modeling of zeolite (in-)stability using tailored neural network potentials

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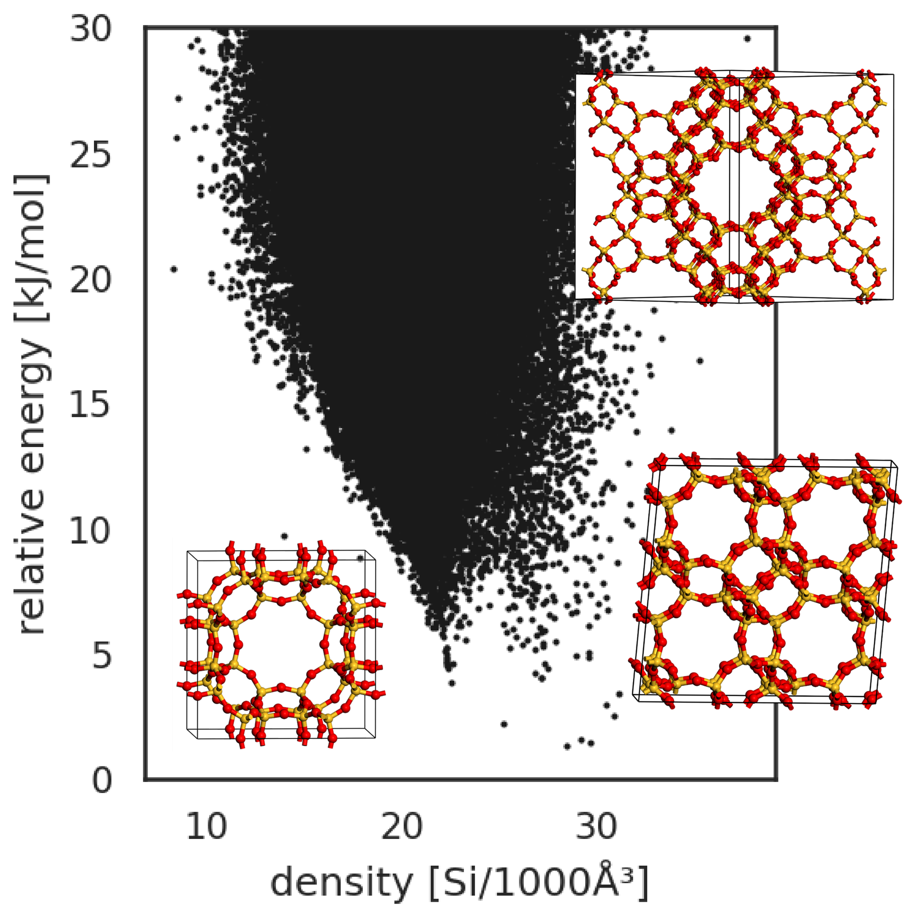
## Introduction

- Zeolites show enormous structural diversity [1]
- Design and discovery of new zeolites requires knowledge of the thermodynamic stability of zeolite frameworks
- Chemoselective hydrolysis of Ge containing zeolites allows targeted synthesis of novel zeolites [2]
- Requirement: accurate and computationally efficient approximation of the PES

Aims:

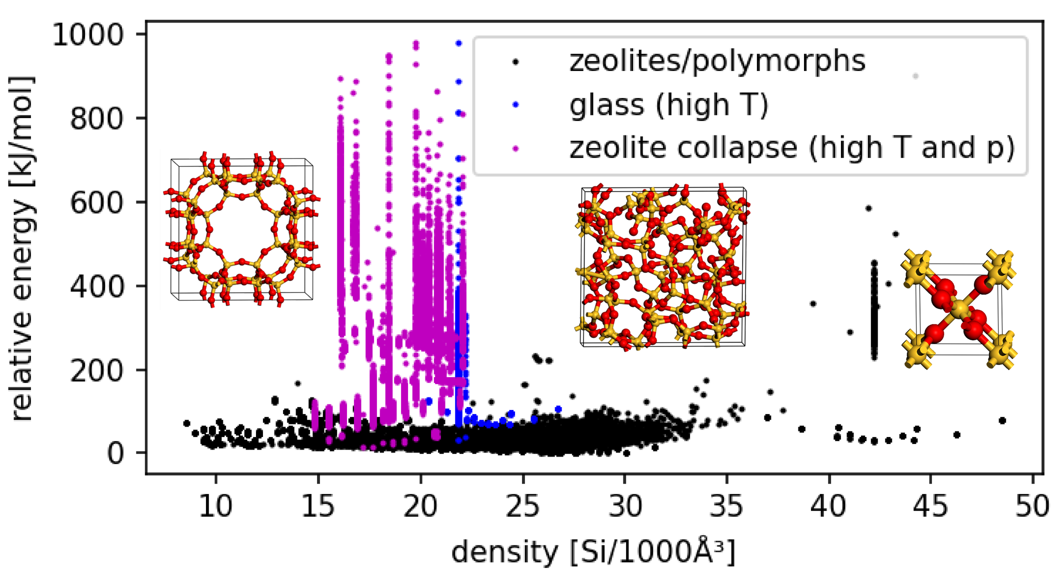
- ❑ Development of (reactive) neural network potentials (NNP) for siliceous zeolites
- ❑ NNP development for large scale simulations of Ge containing zeolites



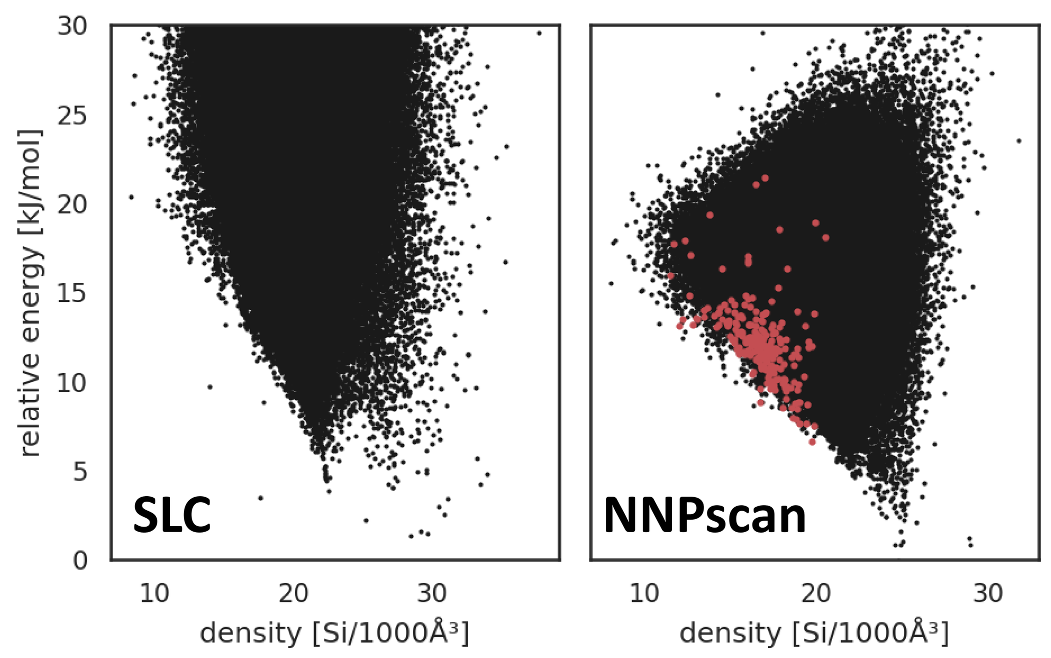

Relative energy (with respect to α-quartz) of more than 330 000 (hypothetical) zeolite frameworks [1]

## NNP training and performance

- Generation of DFT database at the PBE+D3 and SCAN+D3 level by active learning
- NNP model: *SchNet* [3]
- Final database covers low and high energy regions of the PES including low density zeolites and high-pressure silica polymorphs
- Phase transitions at high temperatures (high T) and zeolite amorphization (high T and p)




Relative energy (with respect to α-quartz) and atomic densities of the final DFT (SCAN+D3) dataset (approx. 33 000 datapoints)



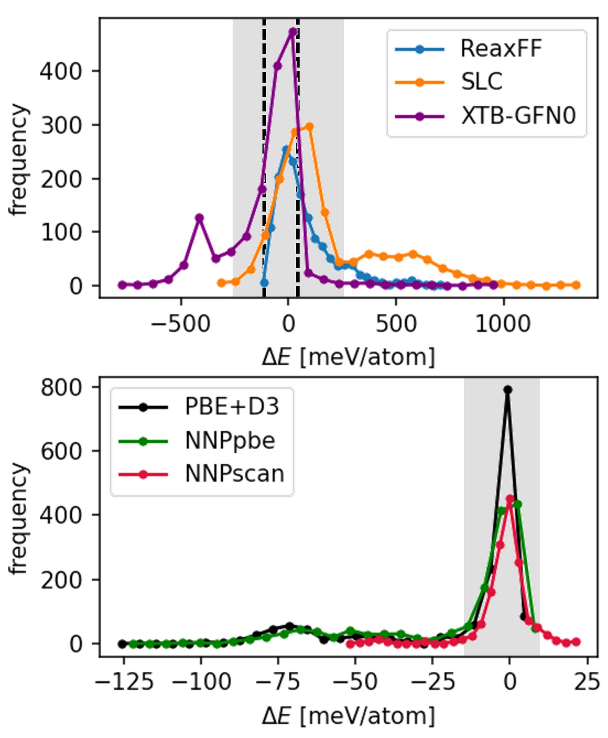

Energy-density plot of hypothetical and existing (red) zeolites calculated using an analytical potential (SLC) and NNPs.

- Analytical force field (SLC) was previously used to generate a database of hypothetical zeolites [1]
- Optimization of the database using the new NNPs parameterized at the SCAN+D3 level
- Database provides vital input for elucidation of structure-property relationships of zeolites using machine learning [4]

- Both NNPs (PBE+D3 and SCAN+D3 level) show virtually same accuracy as DFT
- Energy errors are about one order of magnitude lower compared to analytical potentials (ReaxFF [5], SLC) and TB-DFT (XTB-GFN0 [6])

Root mean square errors (RMSE) with respect to SCAN+D3 of structures not included in NNP training

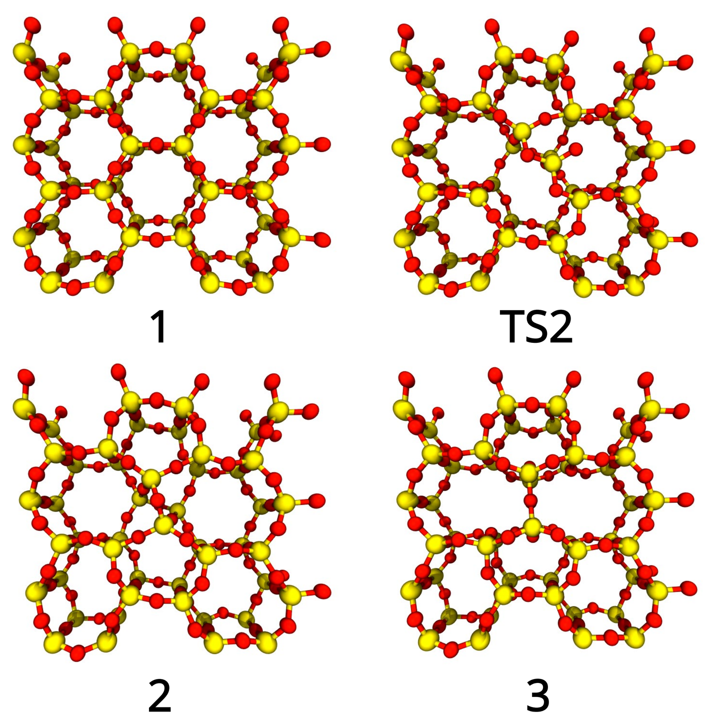
| RMSE | Energy [meV/atom] | Forces [eV/Å] |
|---|---|---|
| PBE+D3 | 31.0 | 0.235 |
| NNPpbe | 29.6 | 0.400 |
| NNPscan | 9.0 | 0.350 |
| XTB-GFN0 | 199.3 | 13.5 |
| ReaxFF | 145.6 | 160.9 |
| SLC | 313.9 | 95.5 |

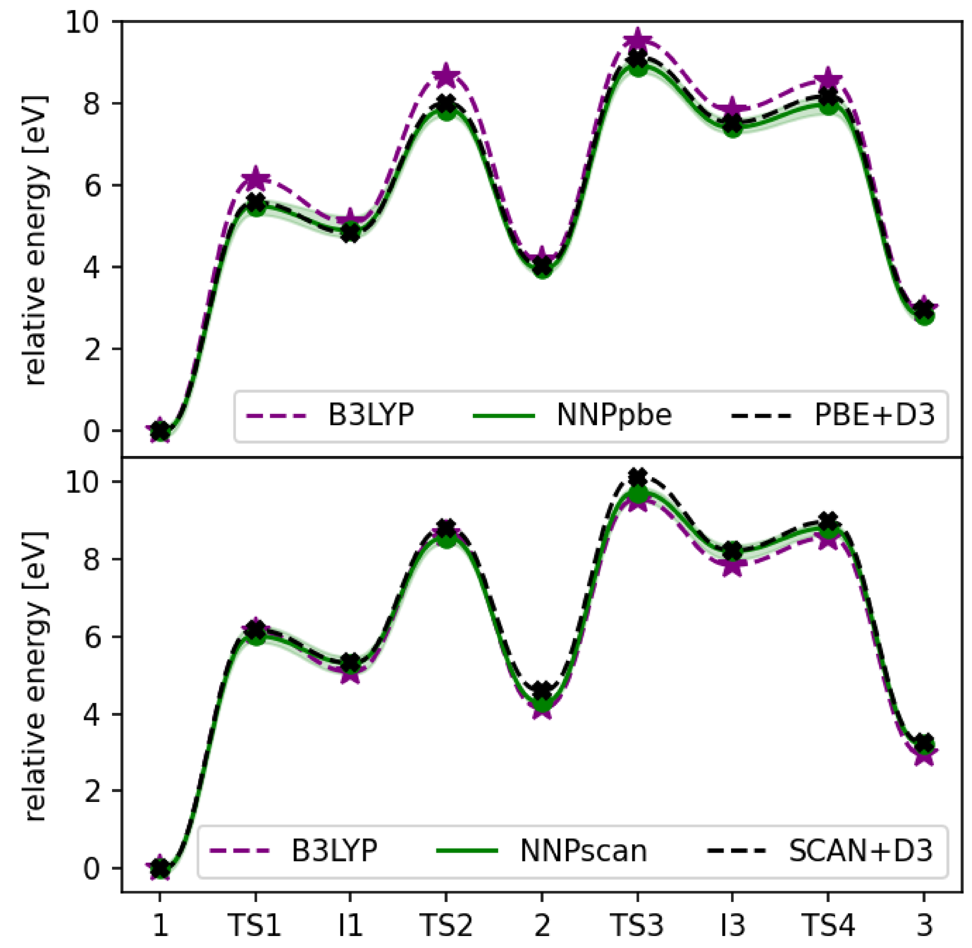



Energy error distribution with respect to SCAN+D3

## Phase transitions of silica

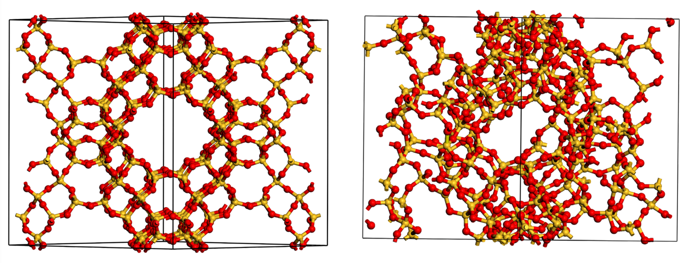
- Stone-Wales defect formation in a silica bilayer *in vacuo* [7]
- NNPs show excellent agreement with DFT results

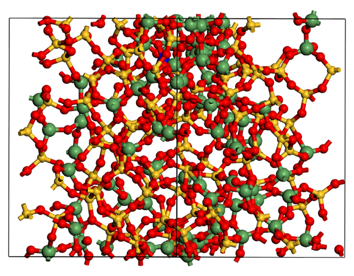



Energies of Stone-Wales defect formation [7]

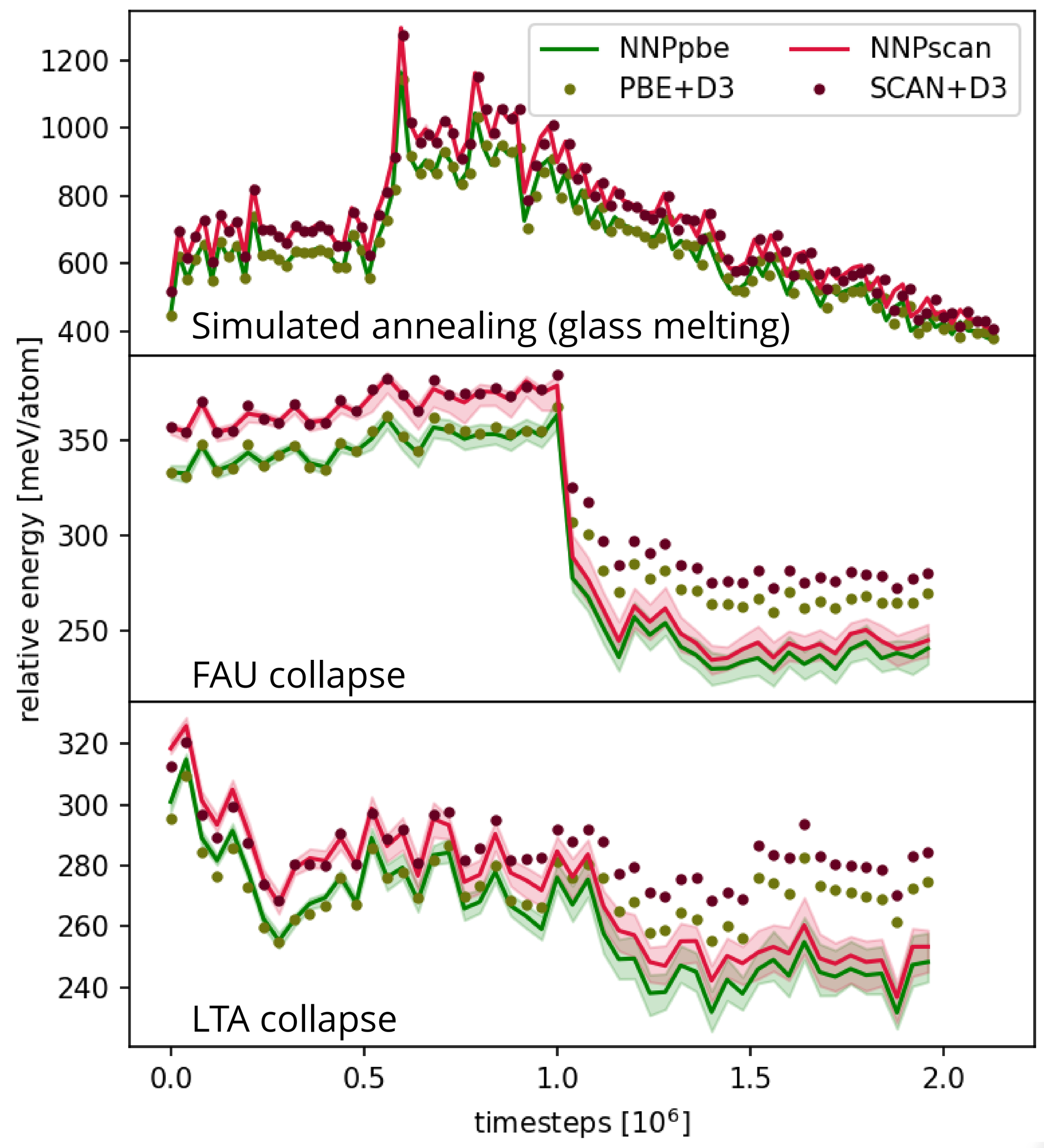
- Melting of high cristobalite at 4800 K and simulated annealing
- Energy difference between PBE and SCAN is considerably larger than NNP error





Collapse of FAU by compression (bond breaking is highlighted in green)

- Zeolite amorphization by MD simulations with stepwise volume reduction at 1200 K
- Reorganization of $SiO_4$ tetrahedra at lower densities
- NNPs systematically underestimate energies (20-40 meV) after bond breaking




Simulated annealing of silica glass and zeolite amorphization by compression at 1200 K

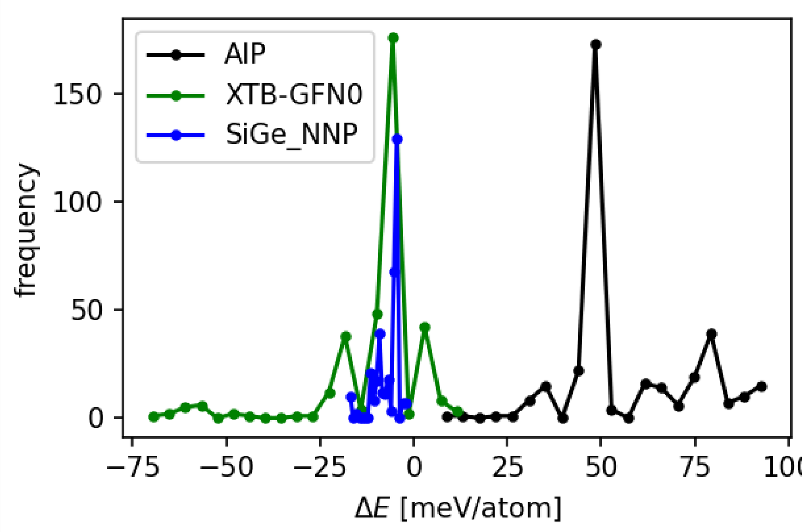
## Summary and outlook

### Tailored silica NNP

- ✓ Robust interpolators of the PES
- ✓ Accuracy close to DFT
- ✓ Fast and accurate simulations of zeolites close to equilibrium as well as at high temperatures and pressures
- ✓ Revised zeolite database as input for future machine learning studies
- ✓ Two DFT databases at the PBE+D3 and SCAN+D3 level for future NNP development

### Next steps

- ⟶ Extension to more complex systems including heteroatoms (Ge, Al, …) and water
- ⟶ NNPs as surrogate model for large scale sampling of the PES (biased dynamics, FEP, …)
- ⟶ Prediction of phase transitions, zeolite hydrolysis, …




Energy error distribution with respect to PBE+D3 of an analytical potential (AIP [8]), TB-DFT (XTB-GFN0 [6]) and tailored NNPs

- Tailored NNP for Ge containing zeolites
- *In silico* screening of the Ge location in zeolites to find promising precursors for the (ADOR [2]) synthesis of new zeolites

Root mean square errors (RMSE) with respect to PBE+D3 of structures not included in NNP training

| RMSE | Energy [meV/atom] | Forces [eV/Å] |
|---|---|---|
| SiGe_NNP | 7.3 | 0.122 |
| XTB-GFN0 | 16.2 | 0.335 |
| AIP | 59.2 | 85.9 |

References

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