Neural-network-backed evolutionary search for $SrTiO_3$ surface reconstructions

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TAMING COMPLEXITY TOGETHER

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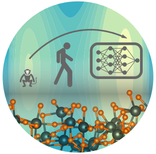
Institute of Materials Chemistry, TU Wien

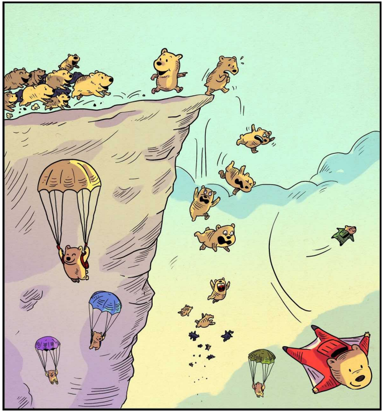


Goal:

Global structure exploration of $SrTiO_3$ surfaces by combining

- evolutionary search strategies and
- machine learning techniques.

Wikimedia Commons.

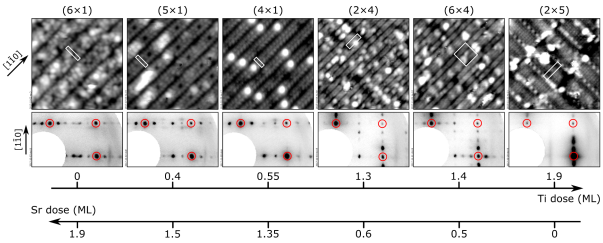


David Ha, blog.otoro.net (2017).

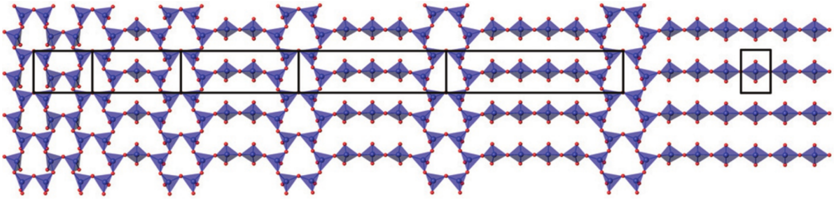
$SrTiO_3$

Strontium titanate

- Cubic perovskite oxide
- n-type semiconductor
- very *practical* material



Riva et al., Phys. Rev. Mater. 3, 043802 (2019).

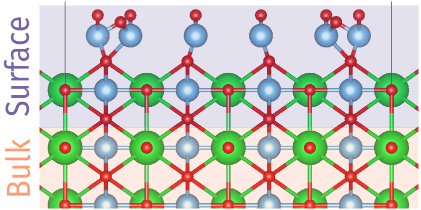


Enterkin. Nat. Mater. 9 (2010).

Objectives

Adapt CMA-ES to surface structures

- Redefine parameters in contrast to bulk systems.
- Compatibility with GPAW (parallelization, call structure).



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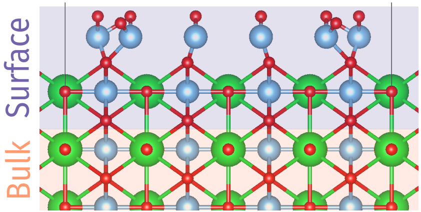
Train a surrogate neural-network model

- Generate data (training, validation, test).
- Adapt the NN parameters and loss function.



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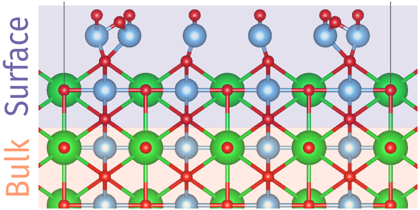

Perform thorough evolutionary searches

- Utilize the CPU cluster and GPUs.
- Sets of CMA-ES runs with NN backend.

Covariance Matrix Adaptation Evolution Strategy (CMA-ES)

- Optimization technique based on ideas of evolution.
- Works for rough energy landscapes.
- Structures are sampled from a multivariate normal distribution.
- Adaptation: Repeat recent successful steps.

$$x_k^{(g+1)} \sim \mathcal{N}\left(m^{(g)}, (\sigma^{(g)})^2 C^{(g)}\right)$$

Hansen, N. (2016). arXiv:1604.00772 [cs.LG]

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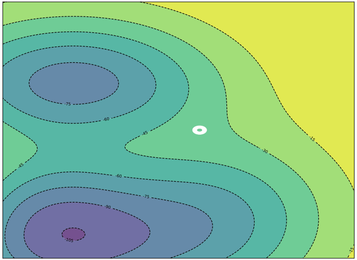

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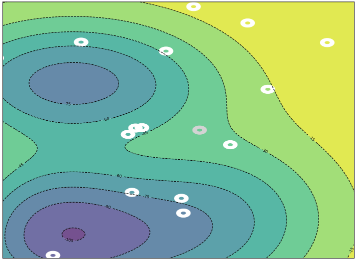

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Explore & Exploit

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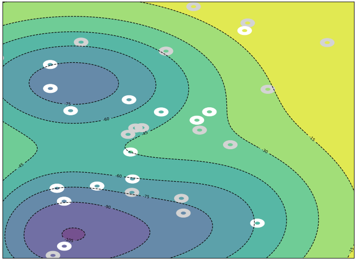

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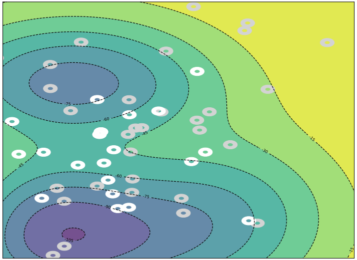

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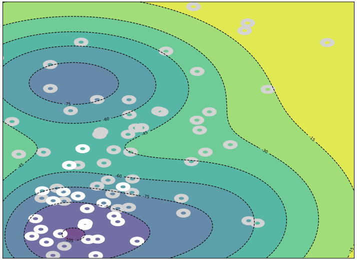

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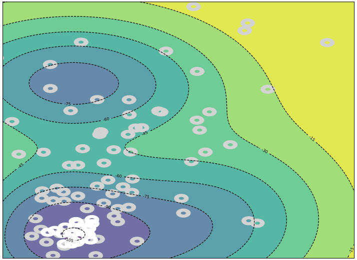

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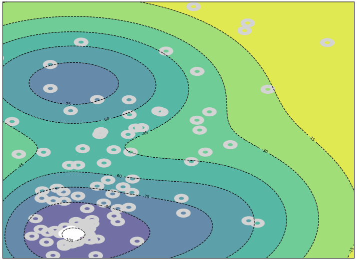

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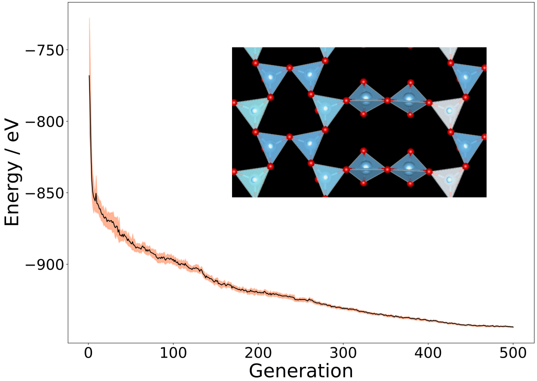
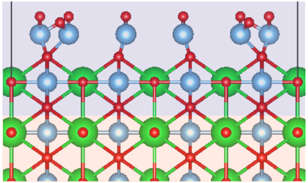
GPAW-backed evolution

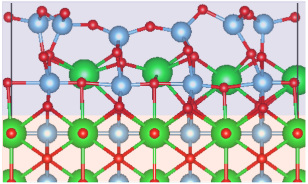
CMA-ES evolution on 4x1 structure

- 500 generations, 11000 individual structures.
- Subsequent local optimization.





Founder

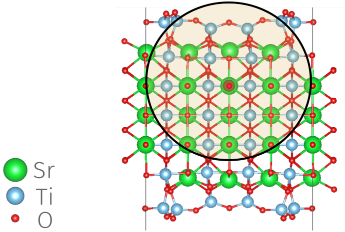


Result

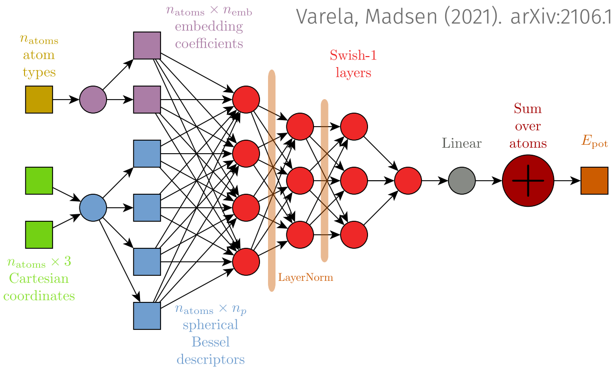
Neural-Network Force Field

Differentiable neural-network force field

- Trained on ab-initio **forces**.
- Atom-centered, **local descriptors**.
- **Transferable** to different system sizes.
- Built on Google JAX.



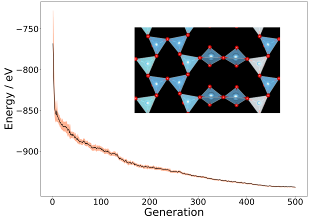
Montes-Campos, Carrete, Bichelmaier, Varela, Madsen (2021). arXiv:2106.16220.

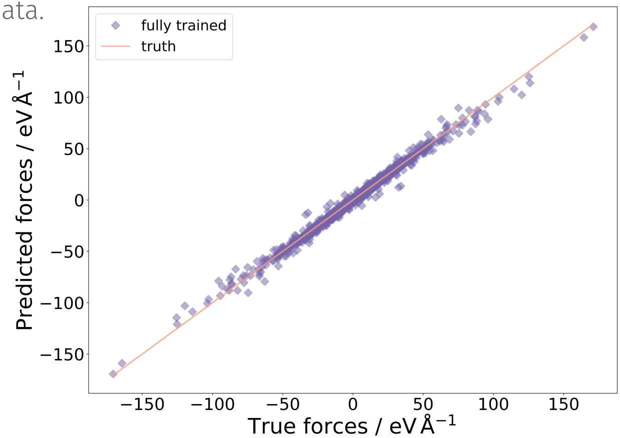


Training the NN potential

Fully trained model (4x1 structures)

- Weakly correlated structures.
- 3000 training data, 500 validation data.
- MAE f = 75.3 meV/Å





Training the NN potential

Fully trained model (4x1 structures)

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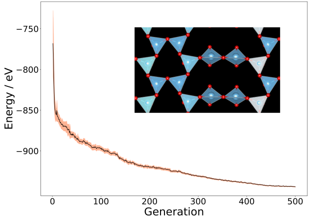
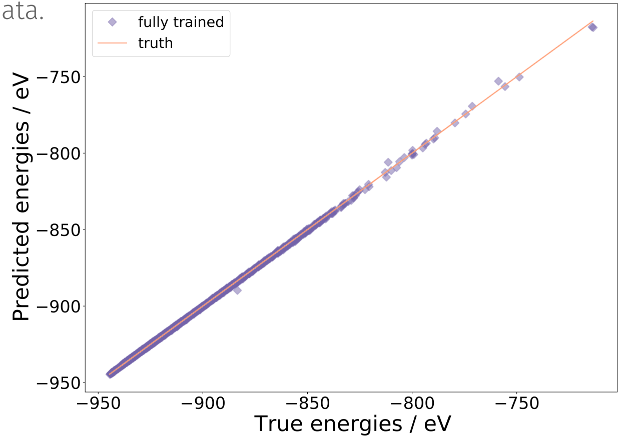
- MAE f = 75.3 meV/Å
- MAE E_{pot} = 2.2 meV/atom

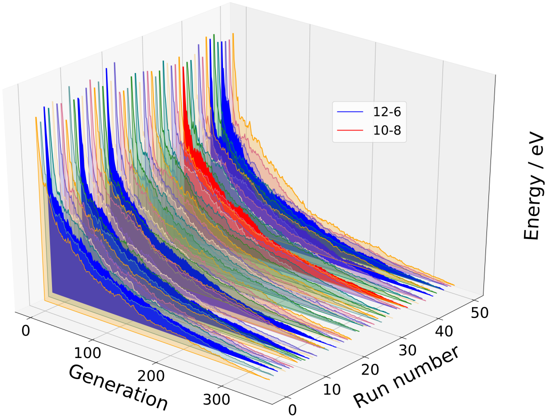




Structure search

Investigate the 5x1 reconstruction

- Use the NN for energy evaluation.
- Run a set of 50 CMA calculations.
- Optimize each CMA result.
- Identified two distinct minima.



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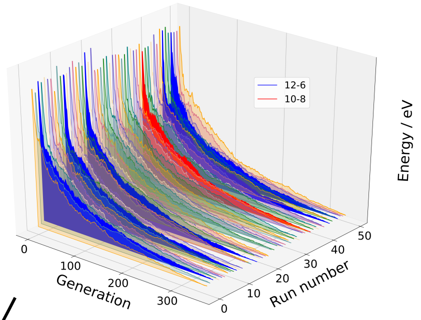
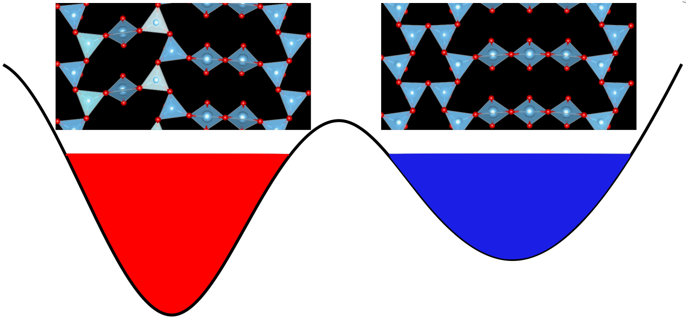
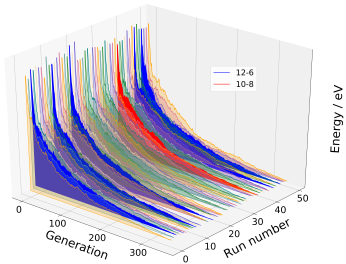


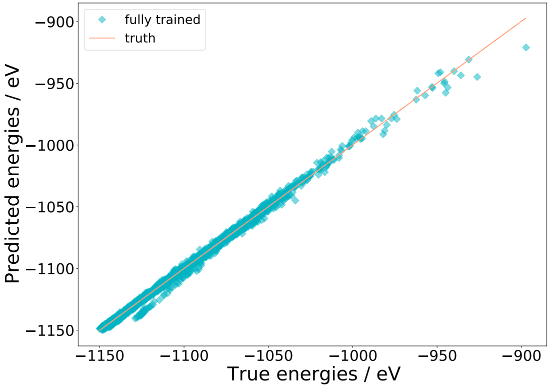


NN performance on 5x1 structures

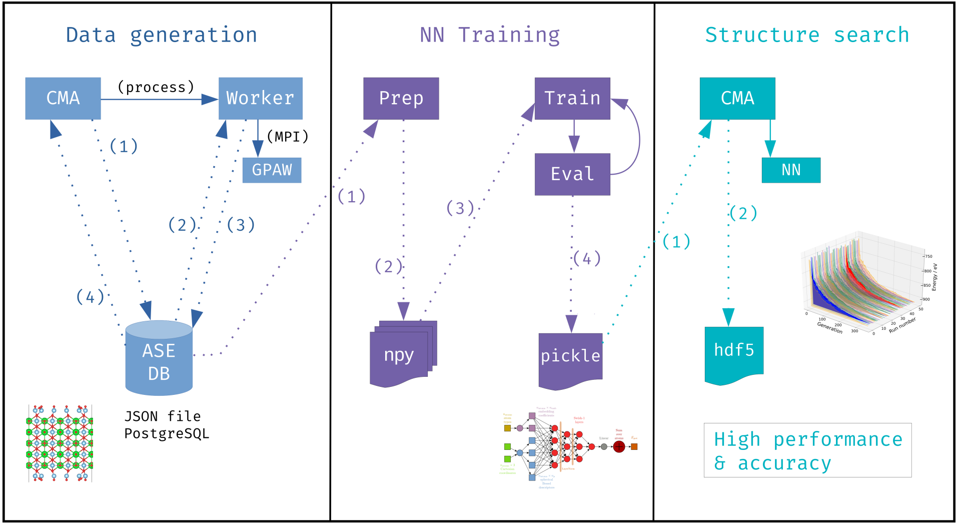
Test the 4x1 model

- 3500 test structures.
- Compare to GPAW results.





Workflow


Data generation
CMA
(process)
Worker
(MPI)
GPAW
(1)
(2)
(3)
(4)
ASE DB
JSON file
PostgreSQL
NN Training
Prep
Train
Eval
(1)
(2)
(3)
(4)
npy
pickle
Structure search
CMA
NN
(1)
(2)
hdf5
Energy / eV
Generation
Run number
High performance
& accuracy

Acknowledgements

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FWF
Der Wissenschaftsfonds.

Bonus - NN performance on 5x1 structures

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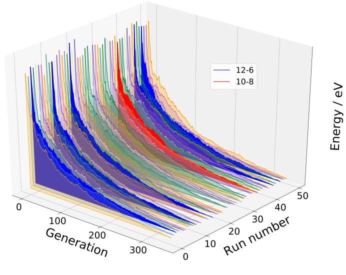

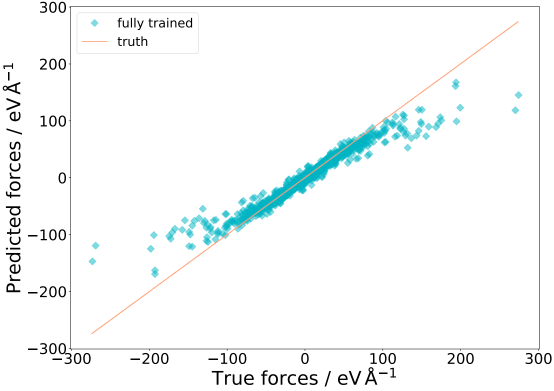

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