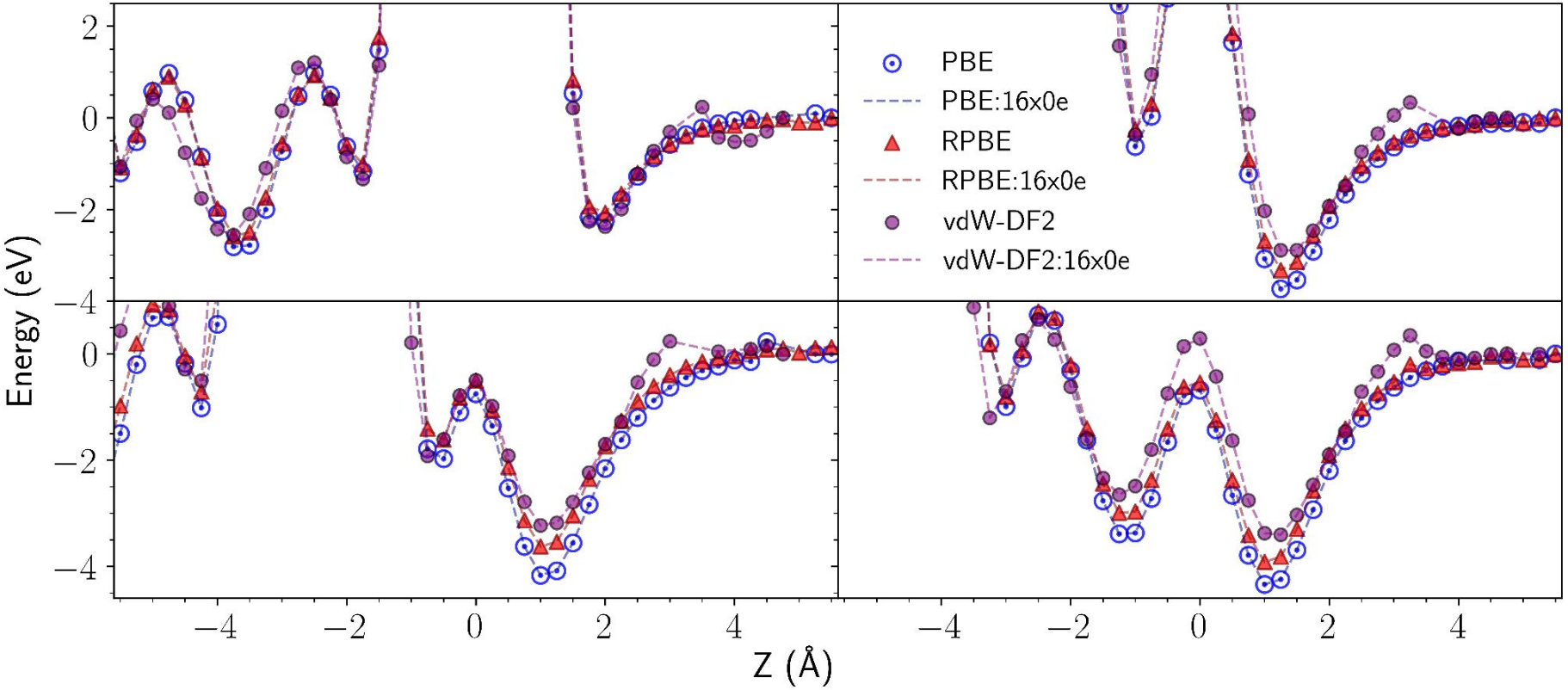
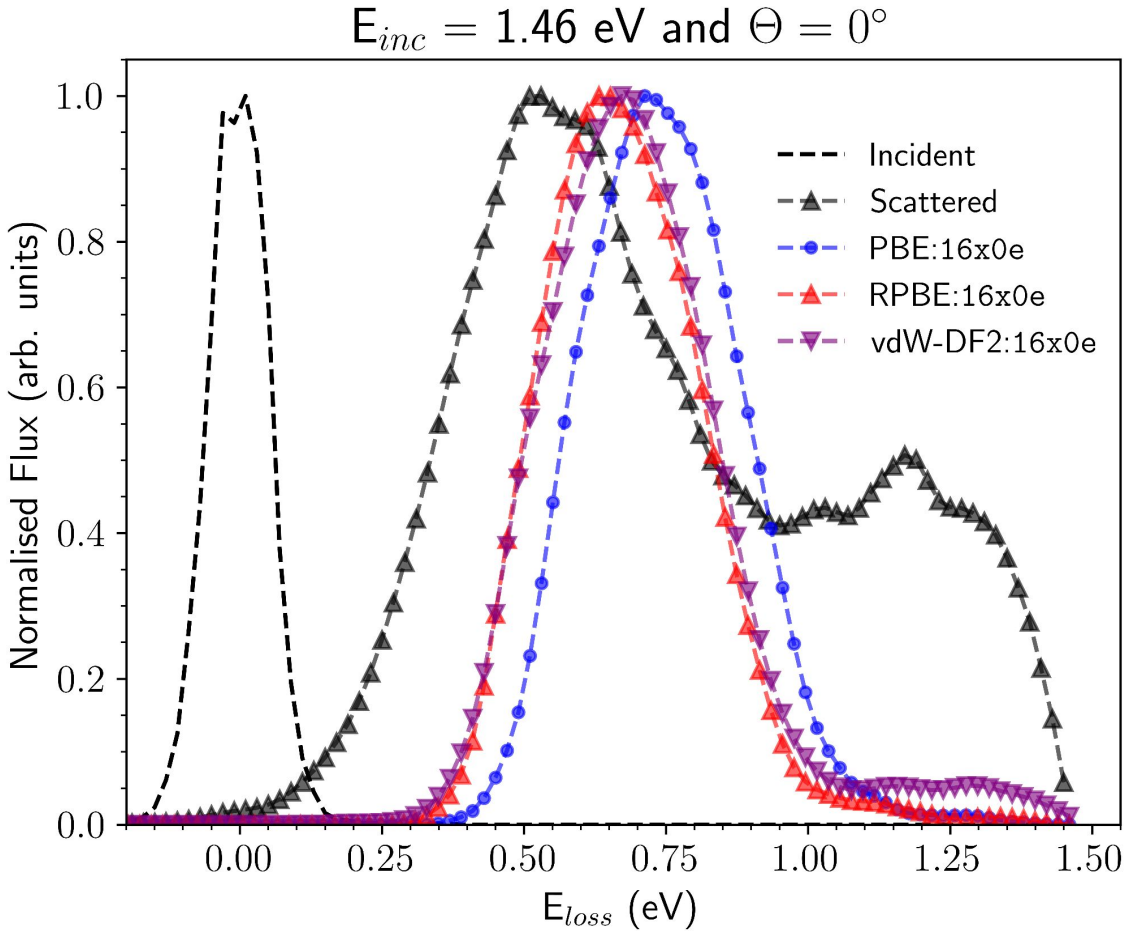
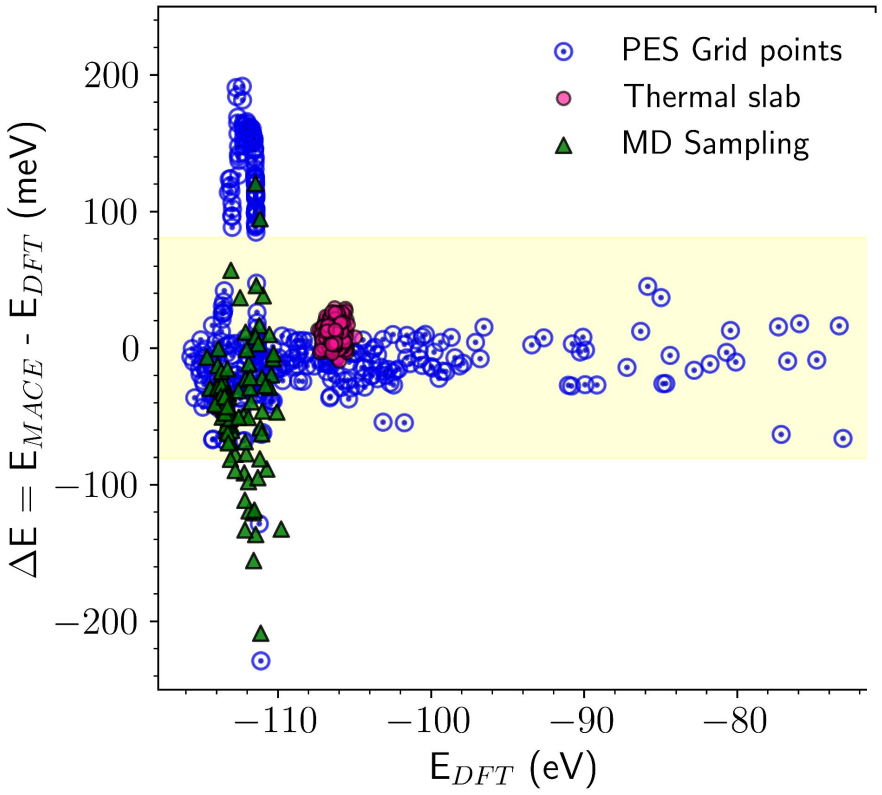
C Atom Scattering

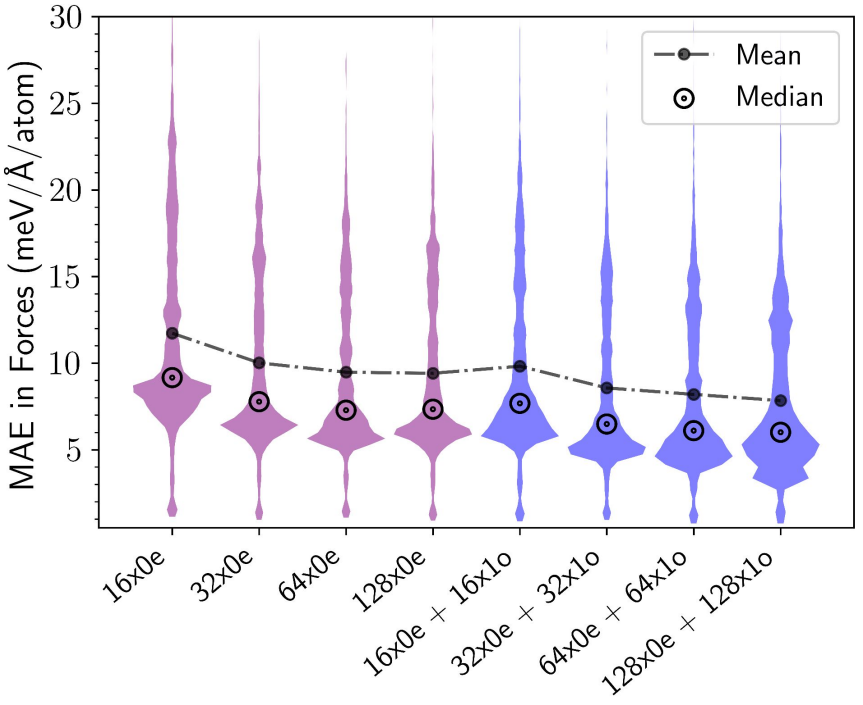
PES Grids for high symmetry sites



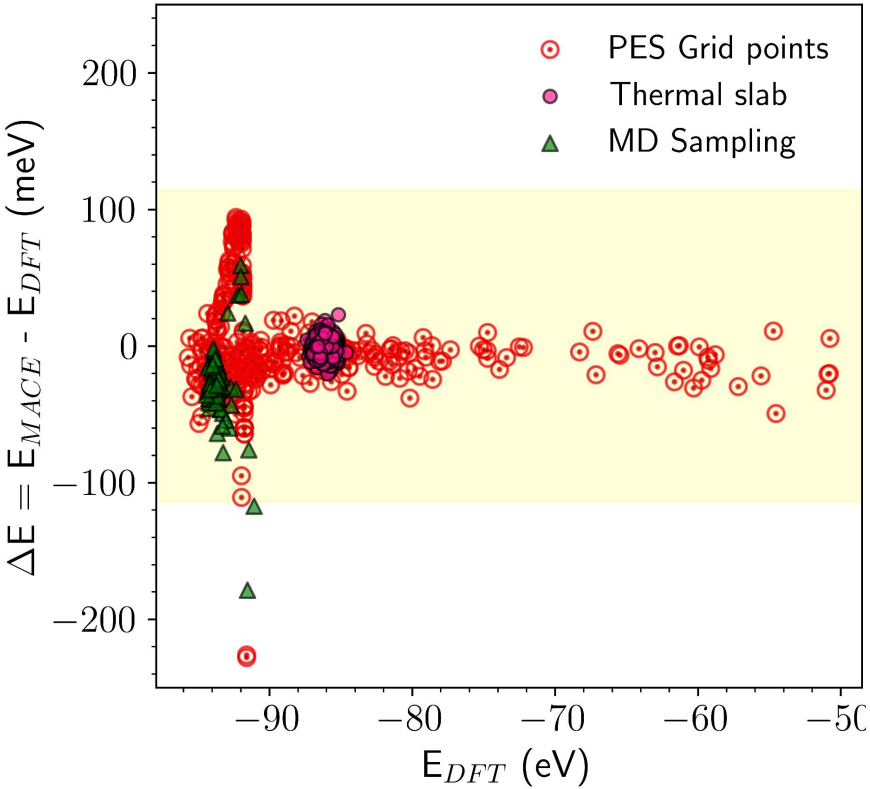

E_{inc} = 1.46 eV and $\Theta = 0°$
Normalised Flux (arb. units)
E_{loss} (eV)
0.0
0.2
0.4
0.6
0.8
1.0
0.00
0.25
0.50
0.75
1.00
1.25
1.50
Incident
Scattered
PBE:16x0e
RPBE:16x0e
vdW-DF2:16x0e

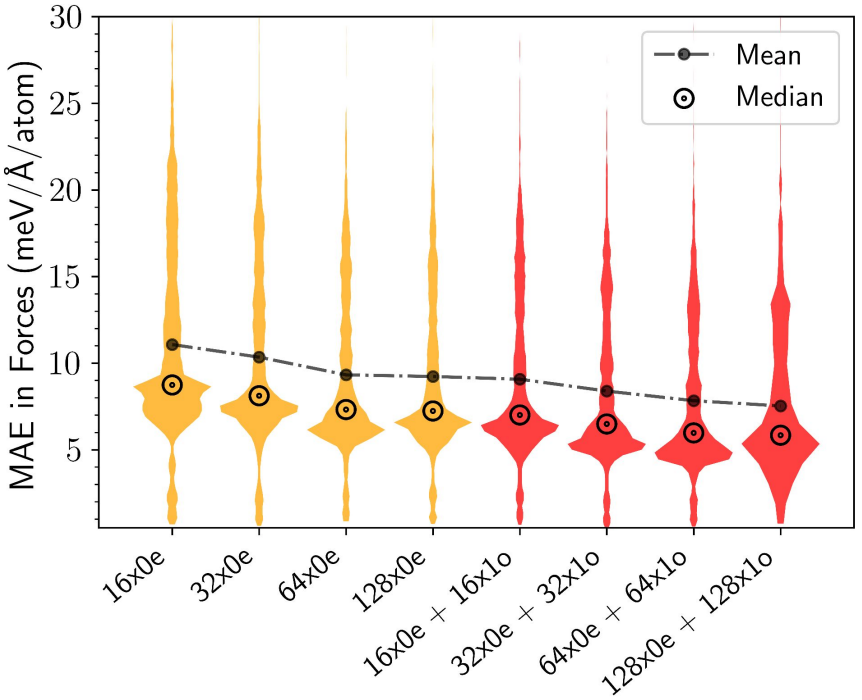
PBE: 16x0e


PES Grid points
Thermal slab
MD Sampling
200
100
0
−100
−200
−110
−100
−90
−80
$\Delta E = E_{MACE} - E_{DFT}$ (meV)
E_{DFT} (eV)

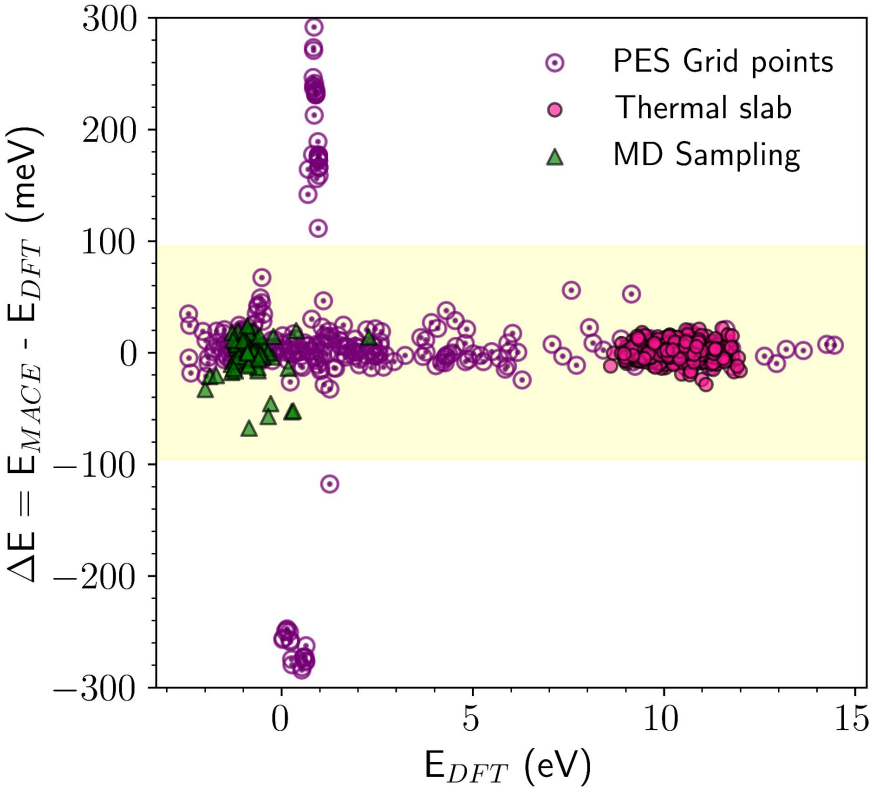

Mean
Median
30
25
20
15
10
5
MAE in Forces (meV/Å/atom)
16x0e
32x0e
64x0e
128x0e
16x0e + 16x1o
32x0e + 32x1o
64x0e + 64x1o
128x0e + 128x1o

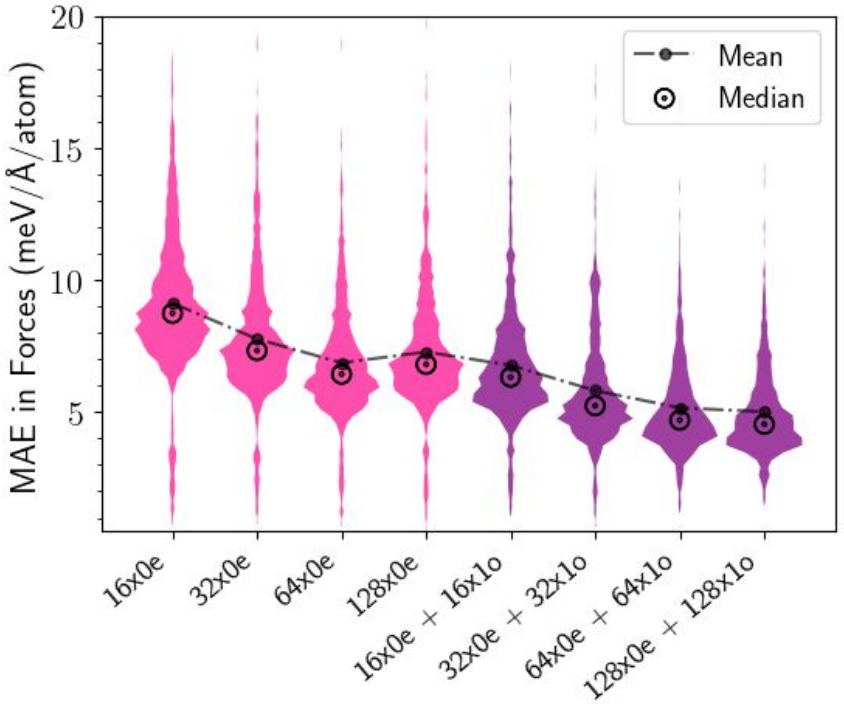
RPBE: 16x0e


PES Grid points
Thermal slab
MD Sampling
200
100
0
−100
−200
−90
−80
−70
−60
−50
$\Delta E = E_{MACE} - E_{DFT}$ (meV)
E_{DFT} (eV)

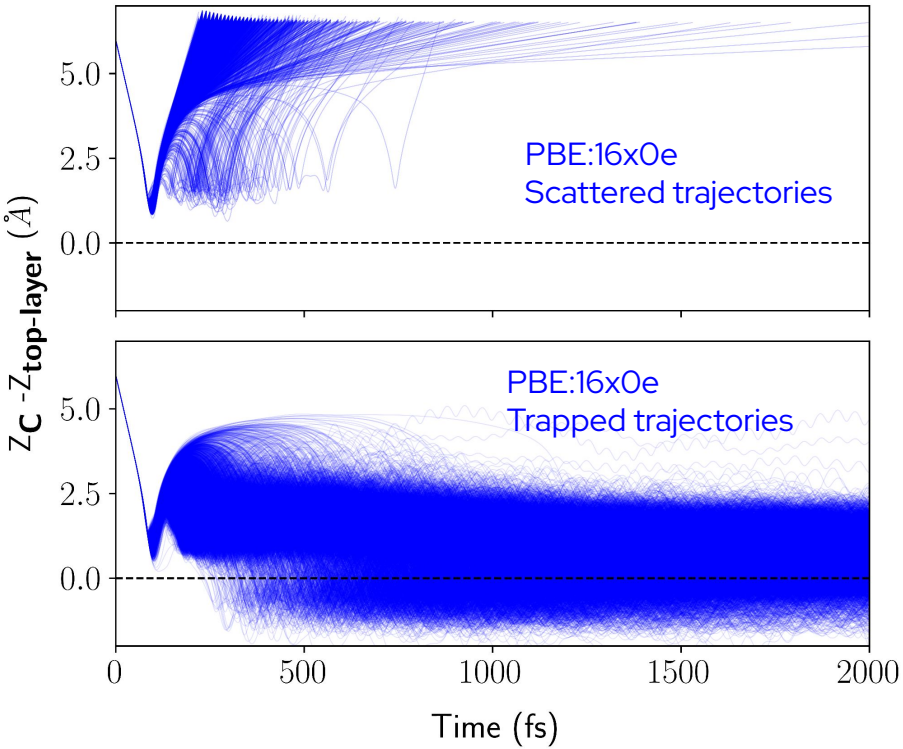

Mean
Median
30
25
20
15
10
5
MAE in Forces (meV/Å/atom)
16x0e
32x0e
64x0e
128x0e
16x0e + 16x1o
32x0e + 32x1o
64x0e + 64x1o
128x0e + 128x1o

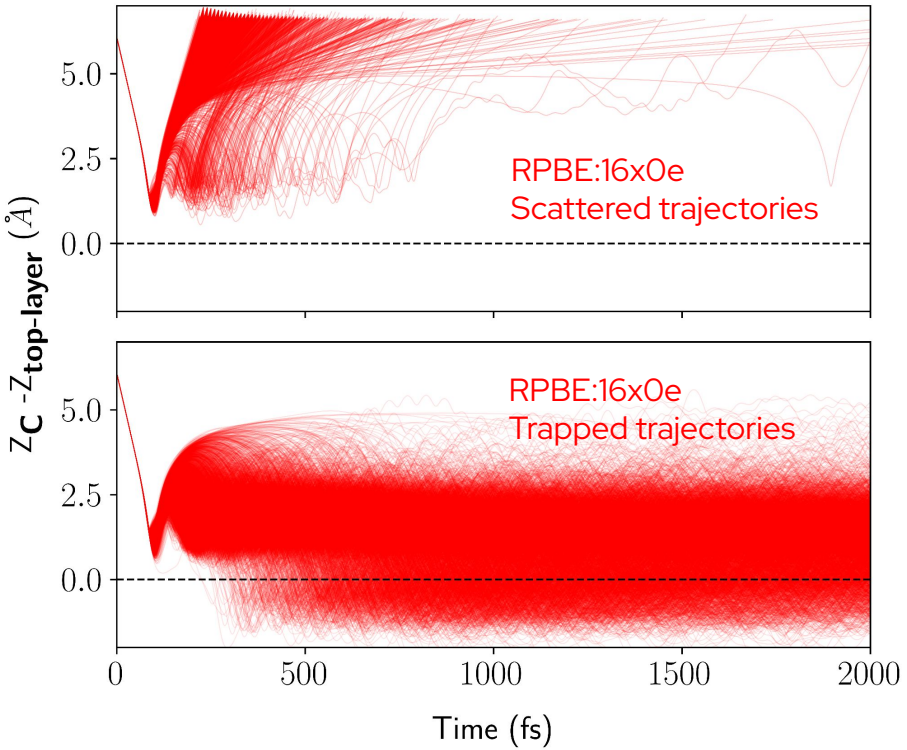
vdW-DF2: 16x0e



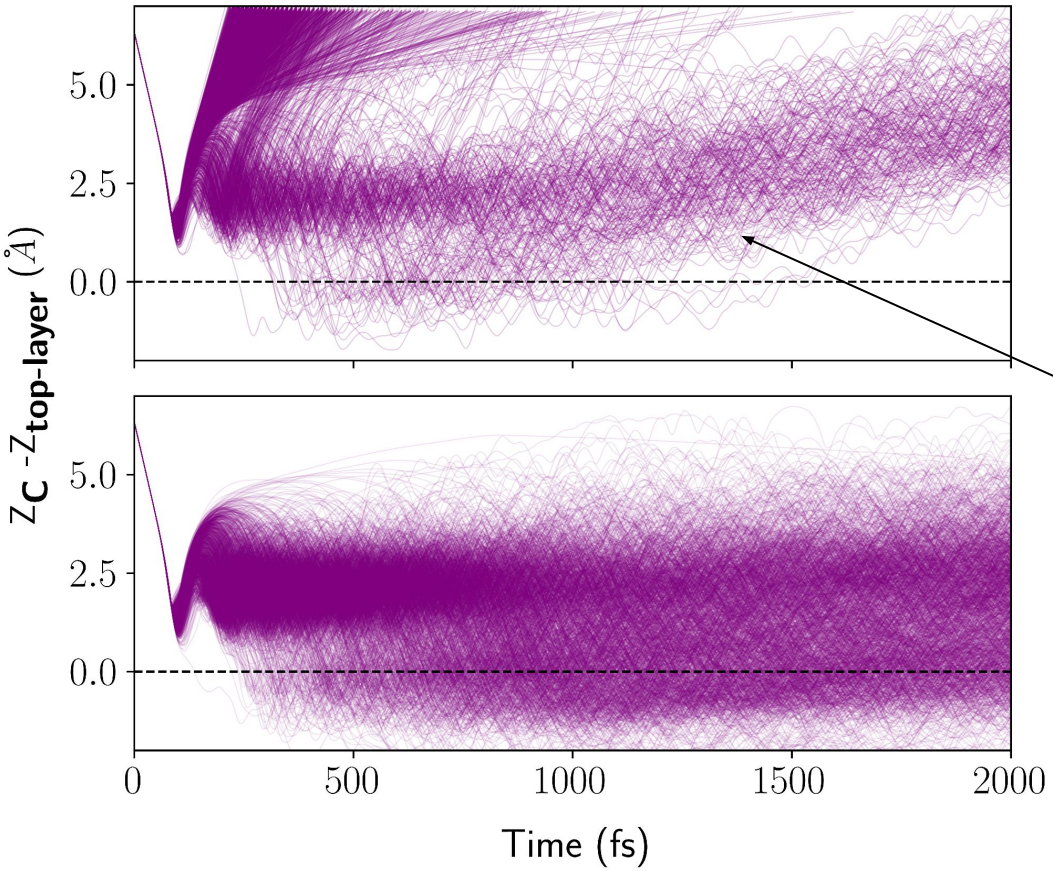


10k Scattering Trajectories in a snapshot





First generation MACE models with the new vdW-DF2 functional



vdW-DF2: 16x0e
Scattered trajectories

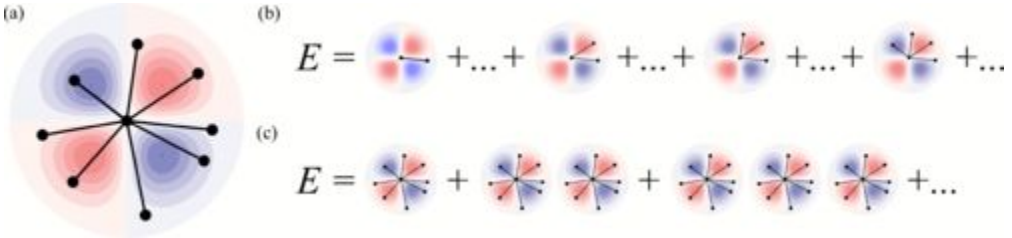
Unphysical turning points with lifting of the top surface atoms

vdW-DF2: 16x0e
Trapped trajectories

Machine-Learned Interatomic Potentials (MLIPs)

MACE is equivariant message passing neural network (MPNN) model that utilizes the atomic-cluster expansion (ACE) framework to pass higher body-order messages. In our models, we have only used upto 3-body features.

$$E = V_0 + \sum_i V^{(1)}(\boldsymbol{r}_i) + \frac{1}{2}\sum_{ij} V^{(2)}(\boldsymbol{r}_i, \boldsymbol{r}_j) + \frac{1}{3!}\sum_{ijk} V^{(3)}(\boldsymbol{r}_i, \boldsymbol{r}_j, \boldsymbol{r}_k) + \frac{1}{4!}\sum_{ijkl} V^{(4)}(\boldsymbol{r}_i, \boldsymbol{r}_j, \boldsymbol{r}_k, \boldsymbol{r}_l) + \cdots,$$



To generate the DFT dataset to train MLIP models, convergence tests were performed for a 3x3x4 slab of Au(111) and, a k-mesh of 15x15x1 with sigma=0.2 eV was used for all the calculations.

DFT	Total # of configurations	Symmetry sites	Randomly Sampled	Thermalised Slab
PBE	1225	455	483	500
RPBE	1236	475	495	495

Models (Hidden irreps)	Total number of parameters
16x0e	31808
16x0e + 16x1o	41856
32x0e	51440
32x0e + 32x1o	78576

Almost all the models that were trained had :

RMSE in Energy: ~1.8 meV / atom

RMSE in Forces: ~ 15 meV / Å / atom

Drautz, Ralf. Physical Review B 99.1 (2019): 014104.

Batatia, Ilyes, et al. Advances in neural information processing systems 35 (2022): 11423-11436.

Stark, Wojciech G., et al. Machine Learning: Science and Technology 5.3 (2024): 030501.