Can machine learning potentials give DFT a run for its money?

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Outline

- Introduction: Overview of three (four) different MLPs
- Relaxations: Tests of different ASE optimizers
- Phonons: Parameter screening for ASE and Phonopy
- Outlook: How I plan to use MLPs in future projects

Introduction: Machine learning potentials (MLPs)[1]

- Potentials that have been **trained** using **machine learning**
- Continuously developed since **introduced in 1995**
- Have become practically **useful** during the last **two decades**
- Exists a **vast variety** based on different models
- **Recently** two **universal** MLPs have been developed
- Have been **pre-trained** on the **Materials Project** database
- Cover **all** practically relevant **elements** in the periodic table

[1] E. Kocer et al., "Neural network potentials: a concise overview of methods", Annual Review of Physical Chemistry **73**, 163–186 (2022).

Introduction: Three (four) different MLPs

- M3GNet:
  - Materials graph network with 3-body interactions
  - New (Pytorch) version available via MatGL
  - C. Chen and S. P. Ong, "A universal graph deep learning interatomic potential for the periodic table", Nature Computational Science **2**, 718–728 (2022)
- CHGNet:
  - Crystal Hamiltonian Graph neural Network
  - B. Deng et al., "Chgnet as a pretrained universal neural network potential for charge-informed atomistic modelling", Nature Machine Intelligence **5**, 1031–1041 (2023)
- NEP:
  - Neuroevolution potential
  - Z. Fan et al., "Neuroevolution machine learning potentials: combining high accuracy and low cost in atomistic simulations and application to heat transport", Phys. Rev. B **104**, 104309 (2021)

Introduction: MLP comparison

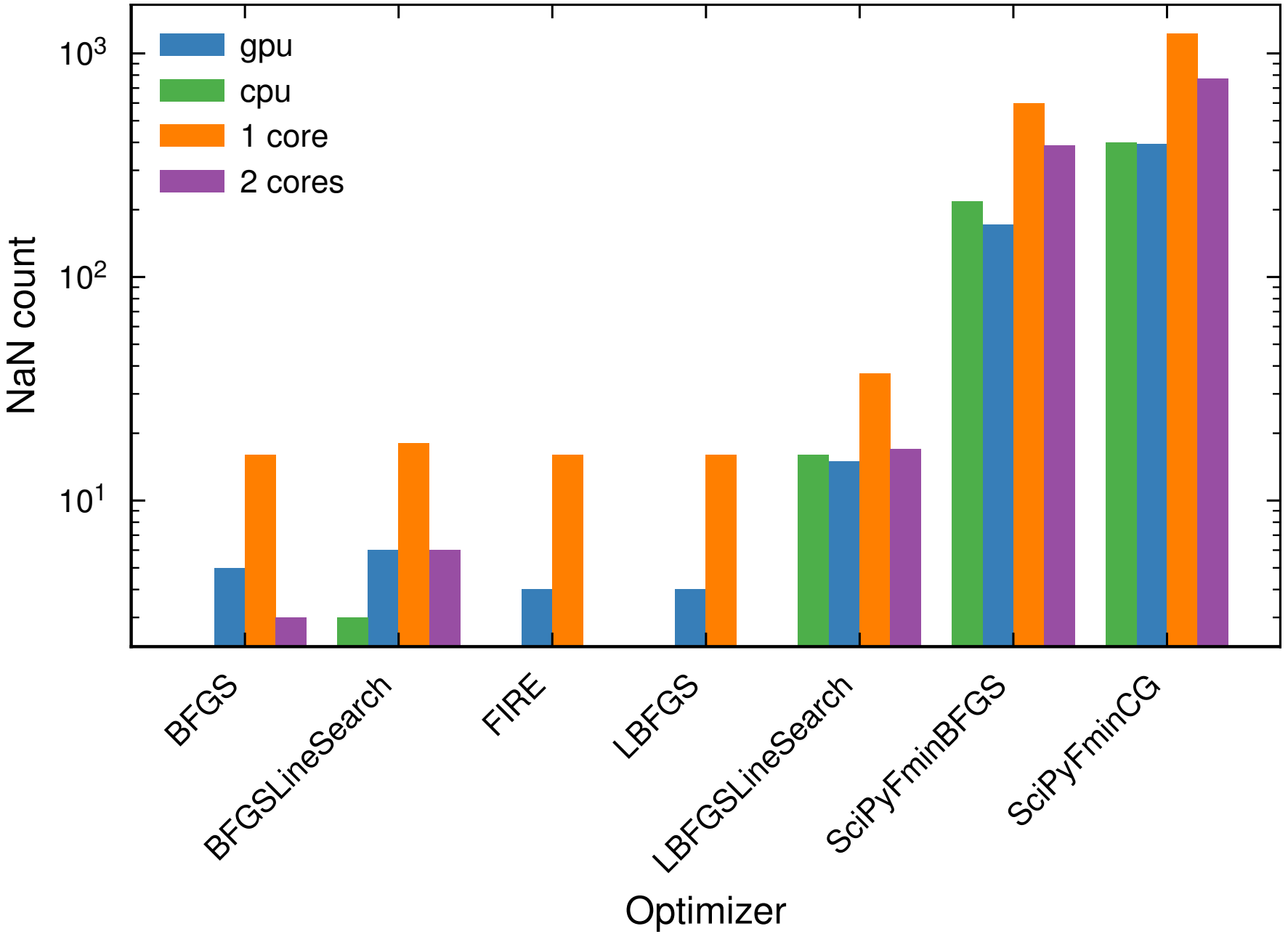
MLP	M3GNet	MatGL†	CHGNet	NEP
Universal Potential?	✓	✓	✓	✗
Archived Version?	✓	✗	✗	✗
Corrected Energies?	✗	✗	✓	?
Pytorch Based?	✗	✓	✓	✗
ASE Calculator?	✓	✓	✓	✓
GPUMD Support?	✗	✗	✗	✓
ROCM Compatible?	✓	✗	✓	?
MPS Compatible?	✓	✗	✓	?

† MatGL stands for the Pytorch version of M3GNet

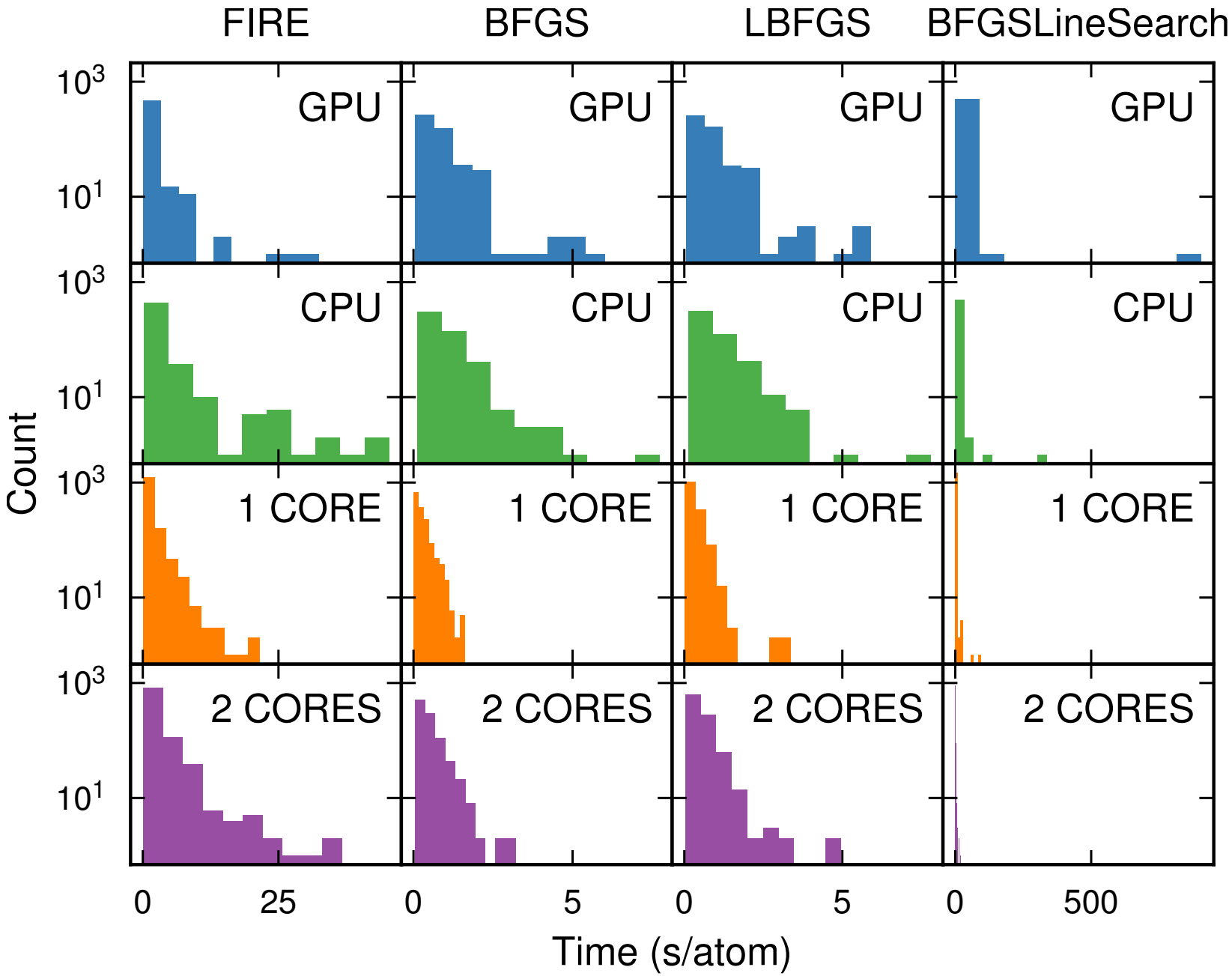
Relaxations: Overview of M3GNet tests

- Performed with archived Tensorflow version on Tetralith
- Structure relaxations using different ASE optimizers
- Considered range of convergence criteria ($F \leq 0.1, 0.01, 0.001\,\mathrm{eV\,Å^{-1}}$)
- Compared various processing schemes:
  - 32 cores/task (1 task/node)
  - 1 core/task (32 tasks/node)
  - 2 cores/task (16 tasks/node)
  - 1 GPU/task (1 task/node)

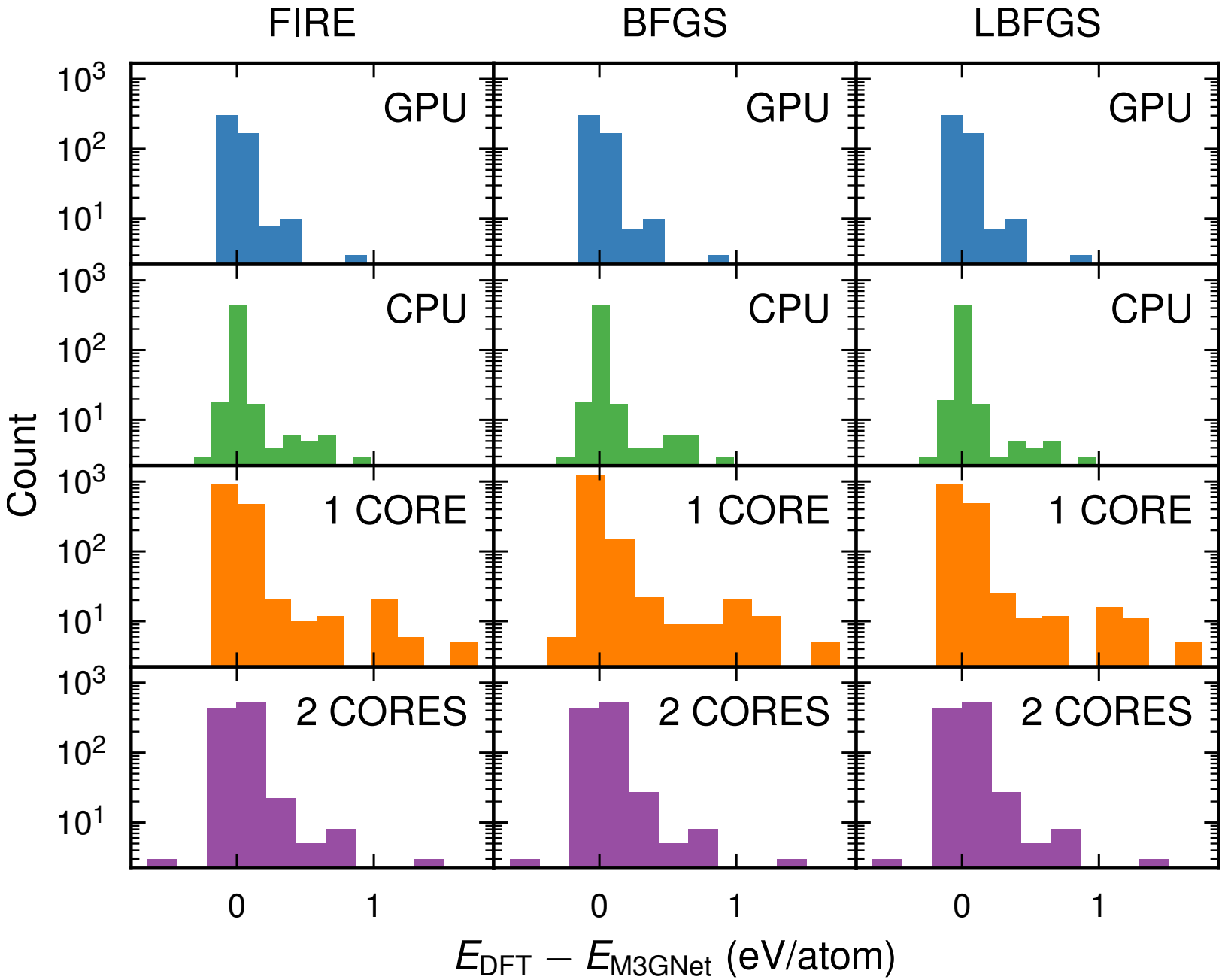
Relaxations: Number of failed calculations



Relaxations: Average time required



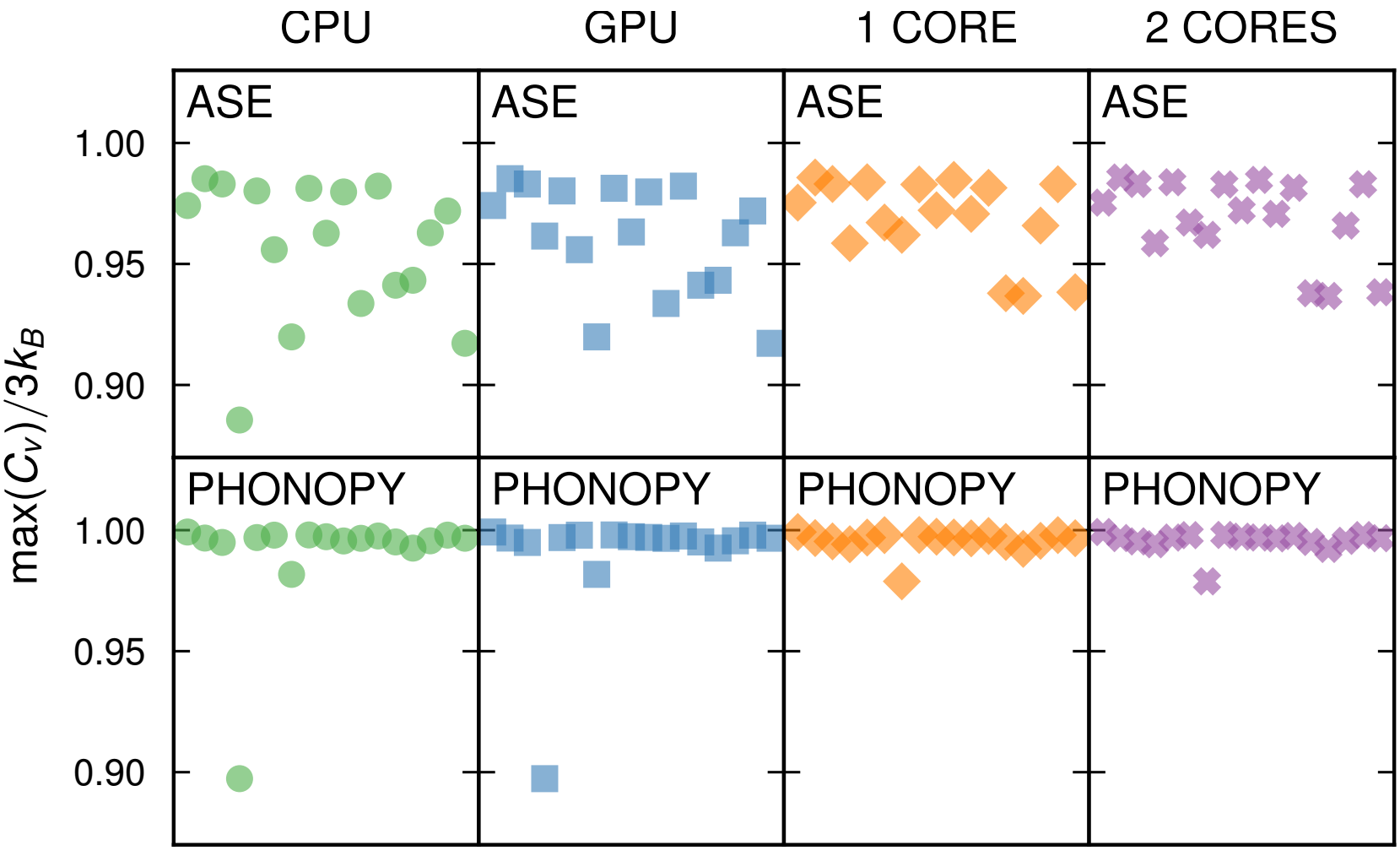
Relaxations: Difference in relaxed energy



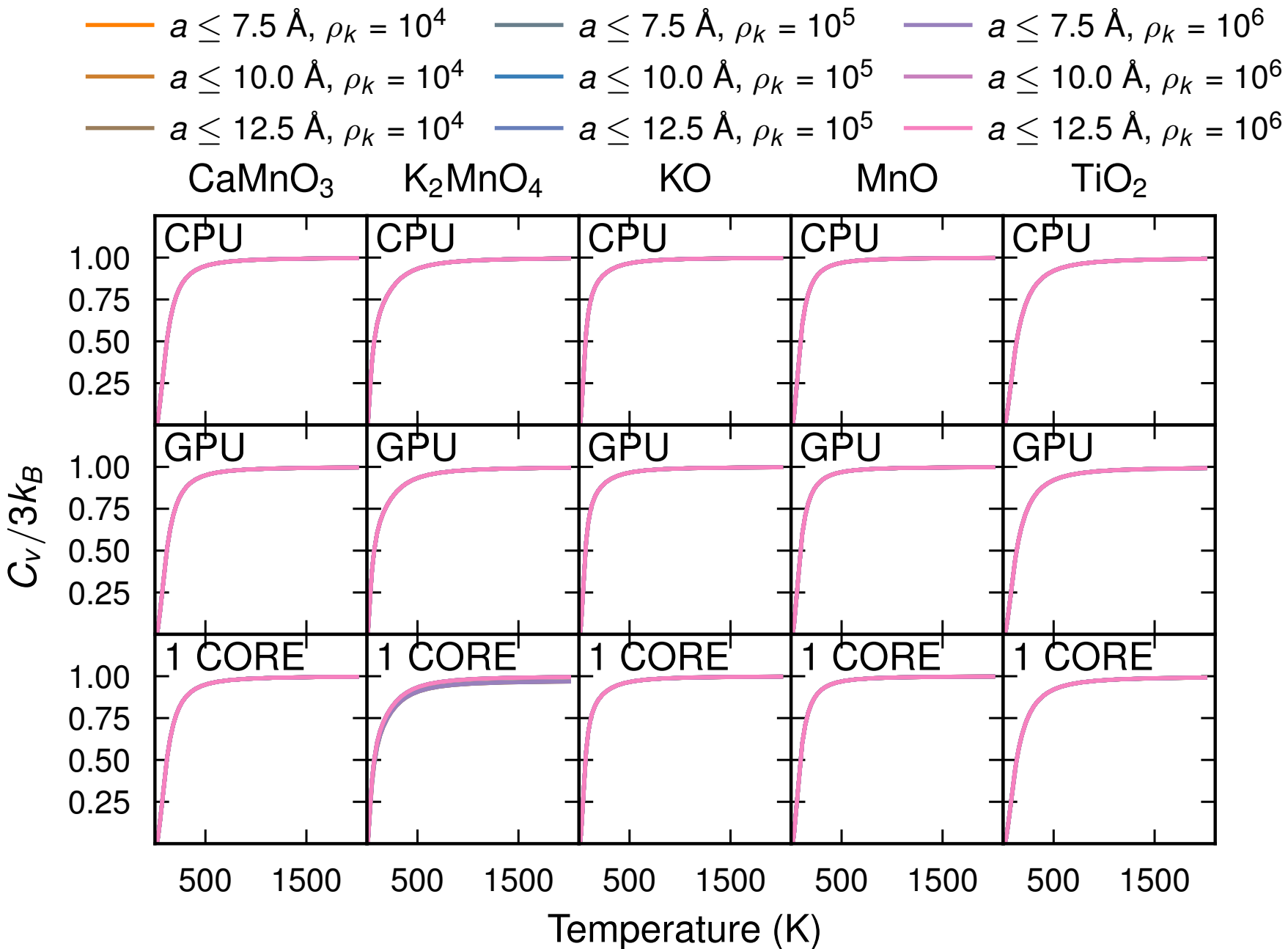
Phonons: Overview of M3GNet tests

- Parameter screening on Tetralith:
  - Harmonic phonon calculations using ASE and Phonopy
  - Quasi-harmonic (QHA) calculations using Phonopy
- Compared additional processing schemes on Dardel:
  - 128 cores/task (1 task/node)
  - 1 core/task (112 tasks/node)
  - 2 cores/task (64 tasks/node)
  - 1 GPU/task (1 task/node)
  - 1 GPU/task (8 tasks/node)

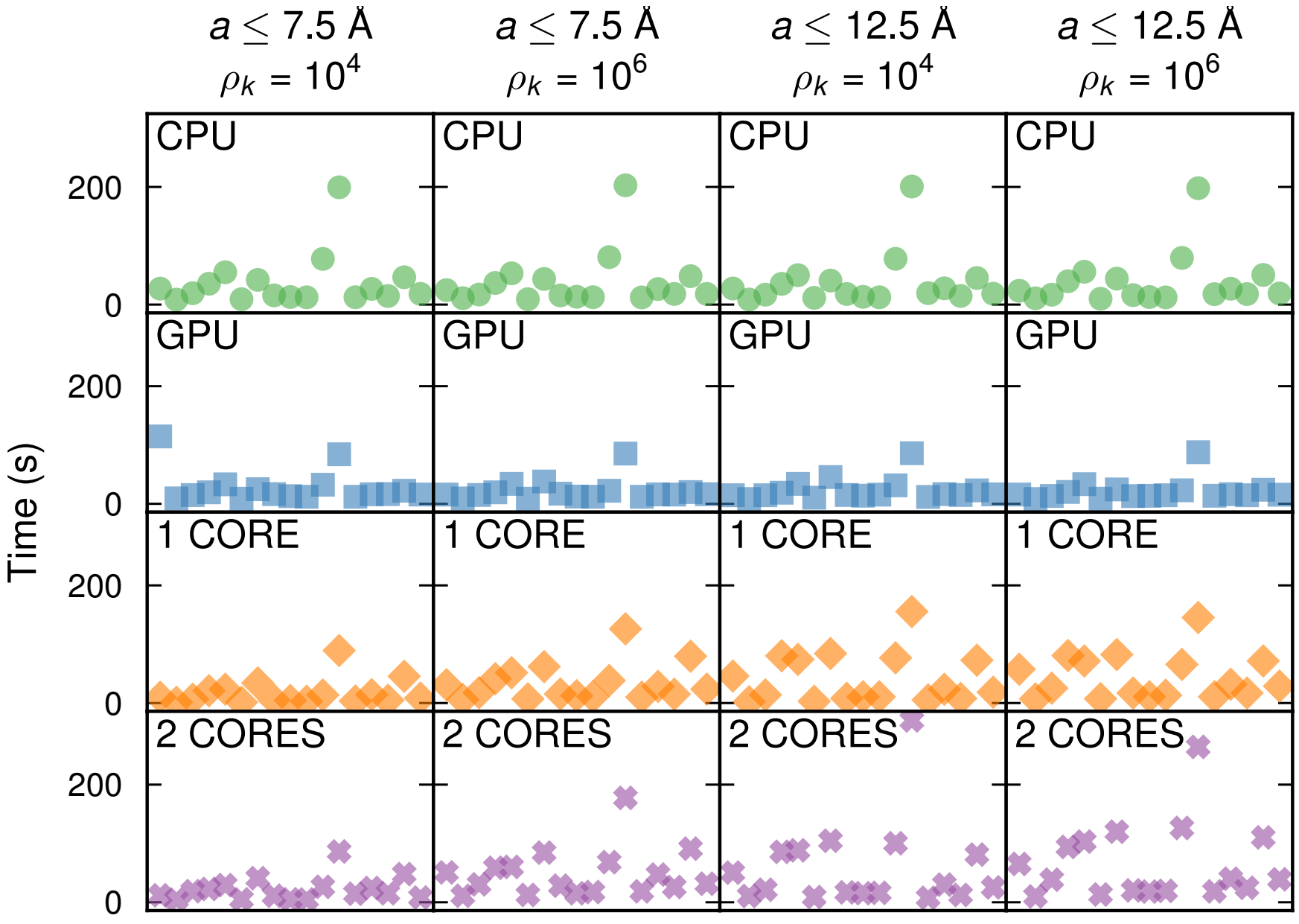
Phonons: ASE is inferior to Phonopy



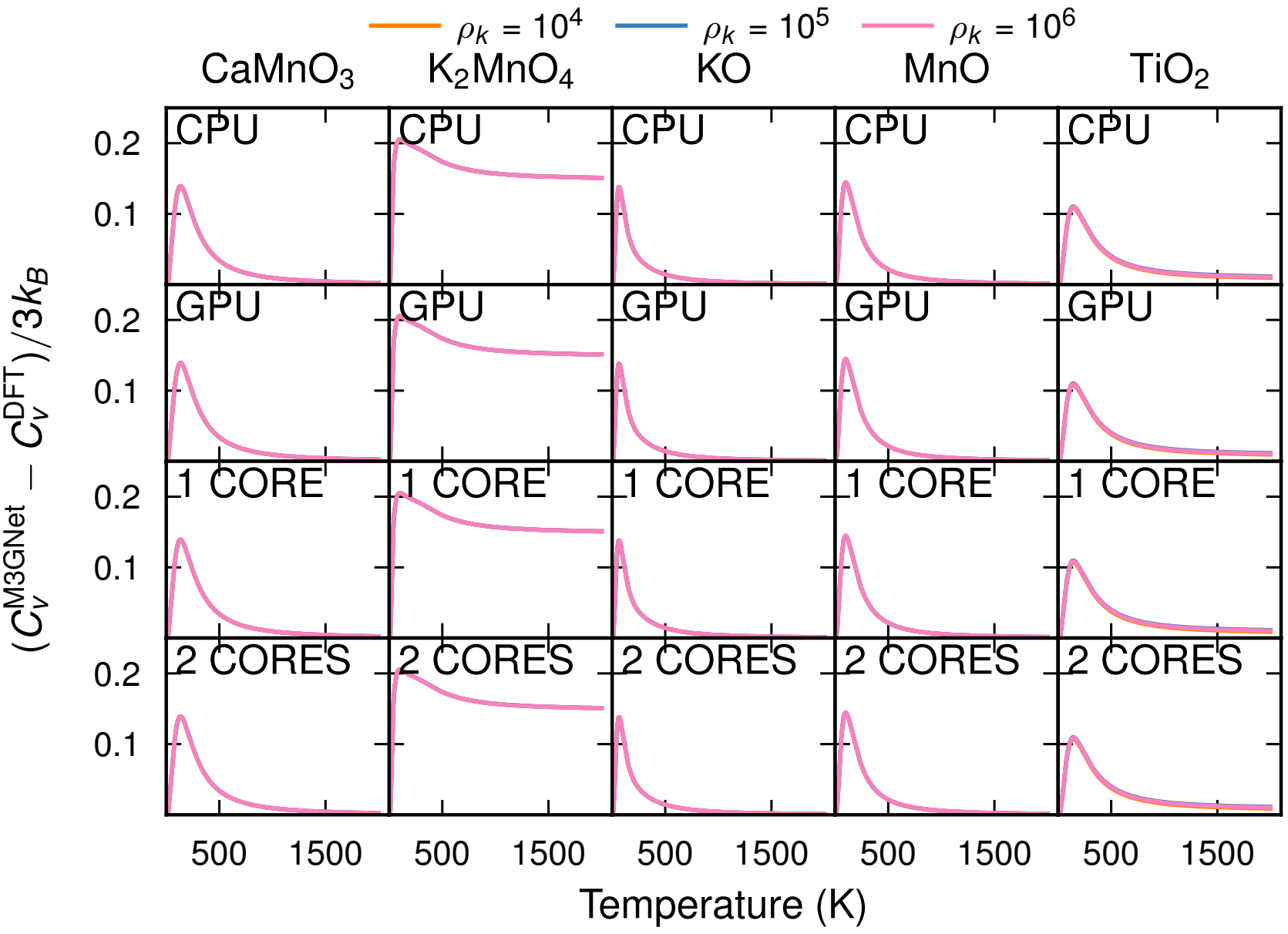
Phonons: Parameter dependence of heat capacity



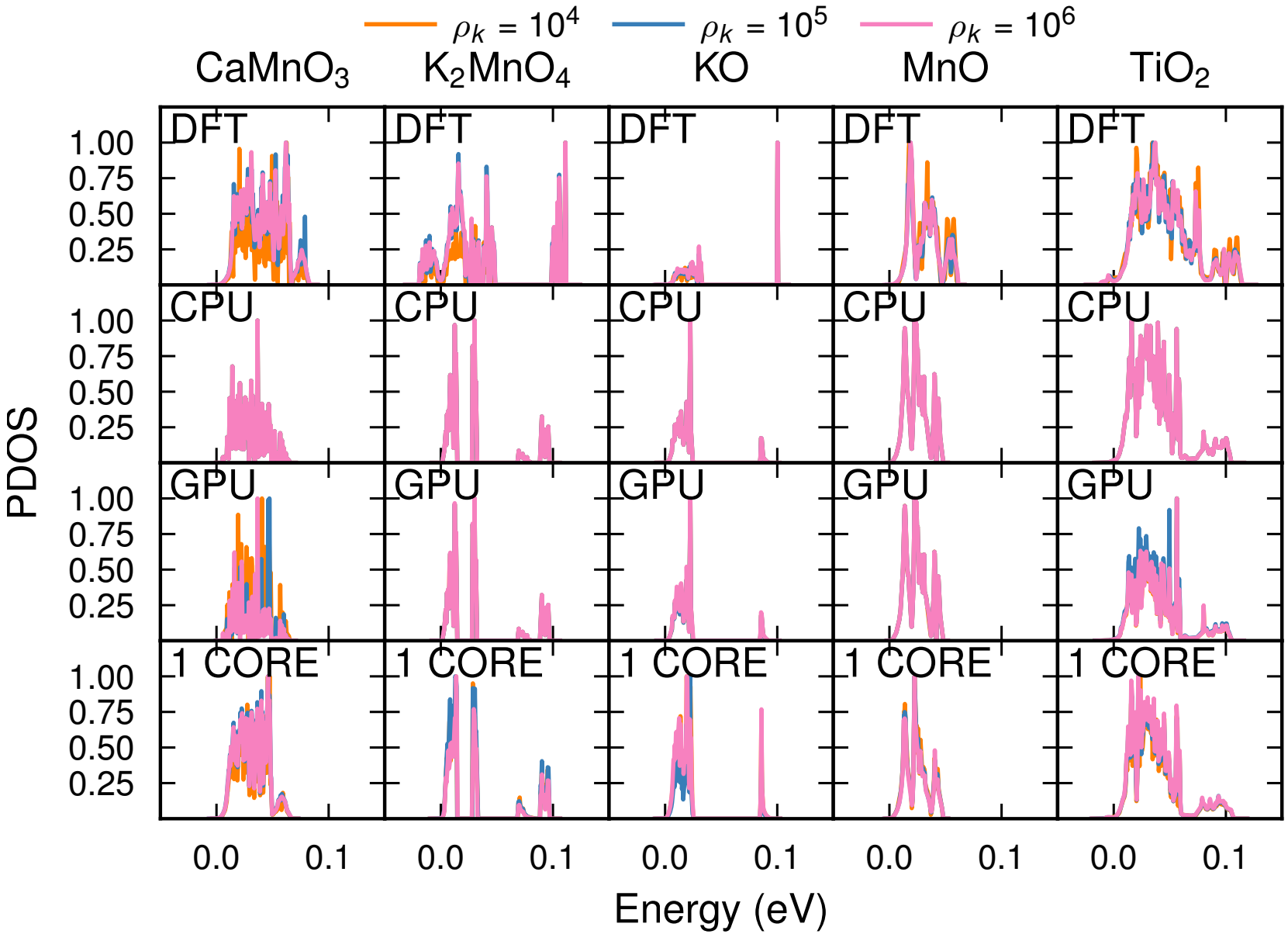
Phonons: Parameter dependence of computational time



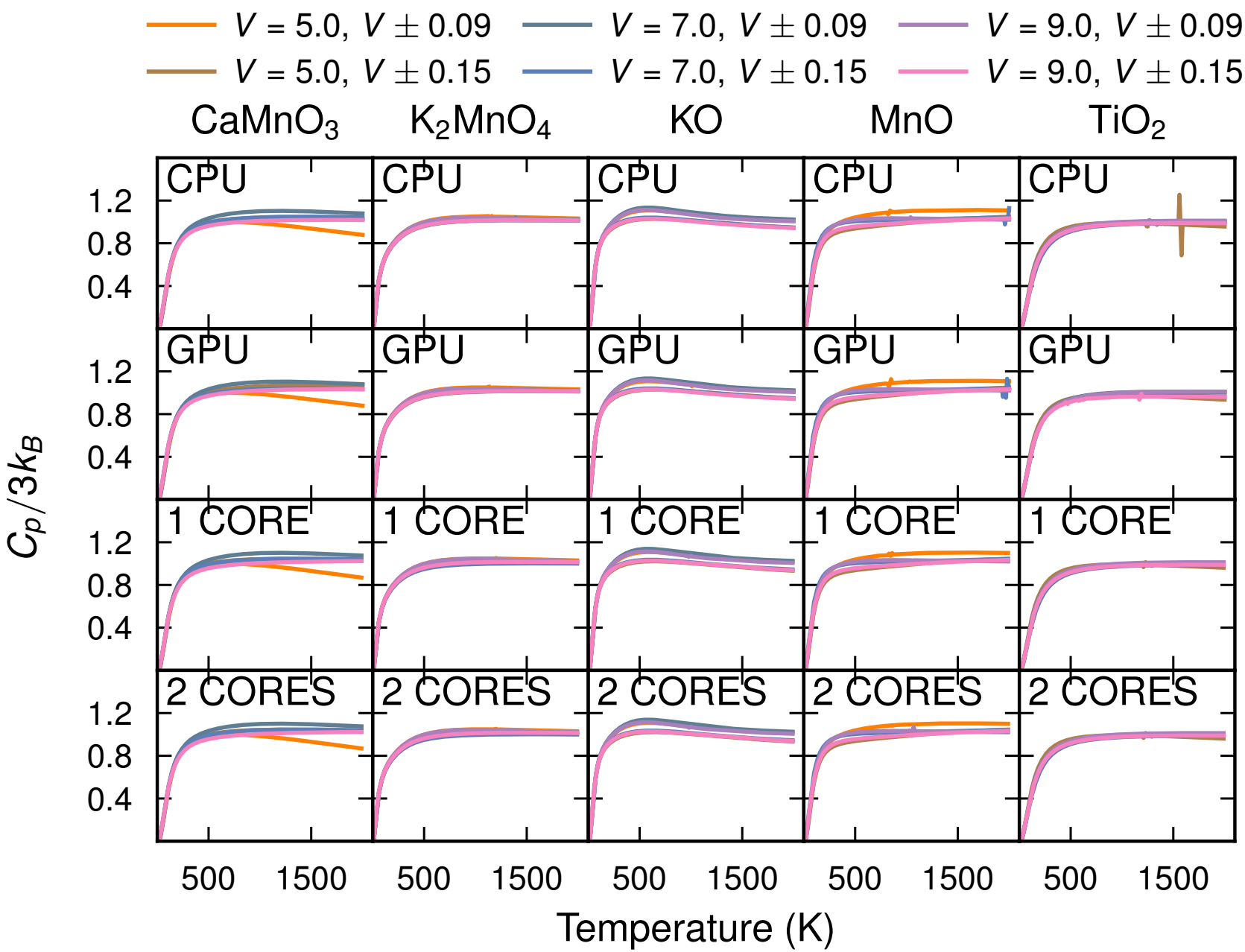
Phonons: Comparison with C_V from DFT



Phonons: Comparison with PDOS from DFT



Phonons: QHA calculations on Tetralith



Phonons: QHA calculations on Dardel

tasks/node	cores/task	GPUs/task	time (h)	time/task (min)	comment
1	128	0	22.74	6.821	
112	1	0	4.084	1.225	memory issues
64	2	0	3.864	1.159	
1	128	1	10.73	3.218	
8	16	1	2.886	0.8659	Tensorflow 1.X
8	16	1	2.955	0.8864	Tensorflow 2.X
8	16	1	5.611	1.683	semi-parallel

Results: Main conclusions & insights

- MLPs are relatively fast and reliable
- Parallelise the calculations if posssible
- Use > 1 core/task to avoid memory issues
- GPUs are faster but leads to more failures
- Phonopy is superior to ASE for harmonic phonon calculations
- QHA calculations are more costly ($10\times$) and trickier
- M3GNet is softer than DFT and overestimates C_V rise

Outlook: Examples of projects involving MLPs

Project 1:

Goal: Find candidate oxygen carriers for energy storage

MLPs: Calculate C_V for compounds retrieved from DFT database

Gain: More accurate estimates of formation energies

Project 2:

Goal: Estimate thermodynamic properties of $CaMnO_{3-\delta}$

MLPs: Perform calculations with CHGNet and NEP as well as DFT

Gain: Makes it possible to consider smaller steps in δ

Project 3:

Goal: Find high entropy oxygen carriers via active learning

MLPs: Estimate oxygen release capacity in chemical looping

Gain: Significantly speed up the learning cycle

Outlook: Available resources

- Jupyter notebook with M3GNet tests
- Gitlab repository for METAL code
  - Code for Material Exploration Through Active Learning
  - Submodule for relaxations and phonon calculations
- Gitlab repository for METAL examples
  - Advanced examples of how to use the METAL code
  - Simple scripts for testing CHGNet, M3GNet, and MatGL
- Matbench Discovery website
  - Platform for benchmarking MLPs based on the Materials Project
  - Compares CHGNet and M3GNet with other MLPs
  - E.g., MACE (not yet implemented in METAL)

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