Machine Learning Assisted Calculation Of Phonon Properties In Layered Systems

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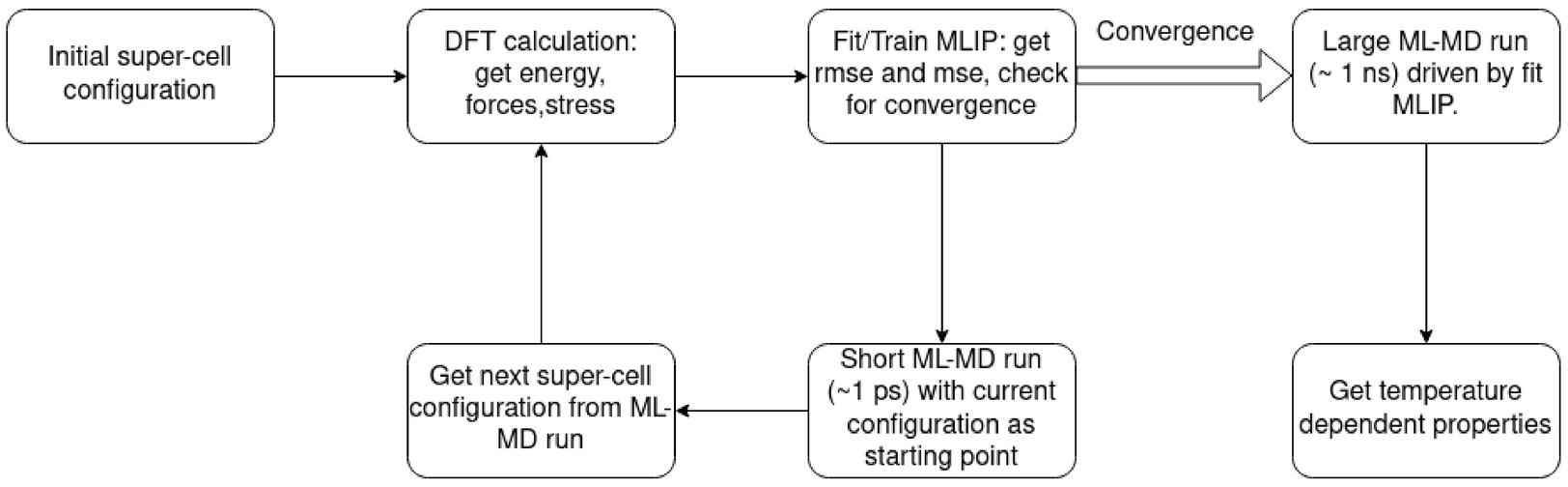
1. Introduction

The usage of Machine Learning (ML) potentials has recently proliferated in the field of materials science. In particular, its usage to train interatomic potentials (MLIPs) and generate canonical sampling (MLACS) allows for great acceleration in the prediction of temperature dependent (TD) properties of materials [1]. Training an MLIP which properly describes the system allows us to drive Molecular Dynamics simulations (ML-MD) for propagation times of the order of the ns, something that is prohibitively time consuming using *ab initio* MD. In this work we show a particular example of such a workflow applied to mono and bilayer hBN, widely popular in 2D applications and a common target for these kinds of MLIPs [2, 3]. We then show that by performing ML-MD simulations, in conjunction with the Temperature Dependent Effective Potential (TDEP) (https://ollehellman.github.io)method [4], we can obtain accurate TD properties like the phonon DOS and thermal conductivity at otherwise inaccessible precision and speed.

2. The MLACS/MLIP workflow

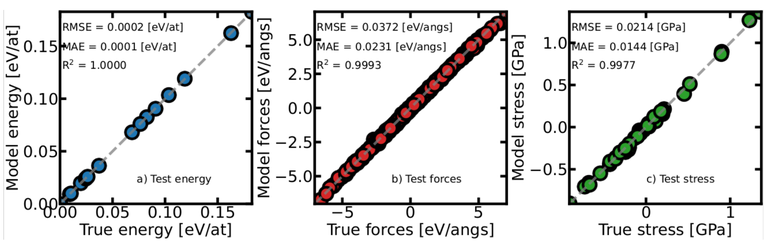


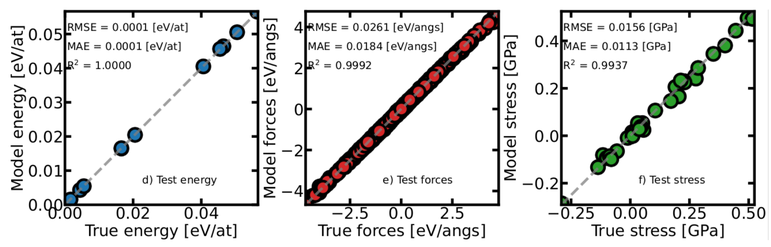
The self-consistent cycle has been shown to minimize the difference between the DFT Gibbs free energy (or, equivalently, the canonical equilibrium distribution) and the MLIP one [1].

3. Calculation Details

- Method applied to mono and bilayer (AA' stacking) hBN.
- DFT done with ABINIT:
  - energy-cutoff: 40 Ha.
  - k-point mesh: 8×8×1.
  - s-cell multiplicity matrix: [[6,0,0], [3,6,0], [0,0,1].
- ML-MD uses LAMMPS (0.5 ns prop. time). MLIP fit linearly to snap potential.
- ABINIT/LAMMPS interface via home-built Python package.
- TD properties obtained with TDEP.

4. Results and Discussion





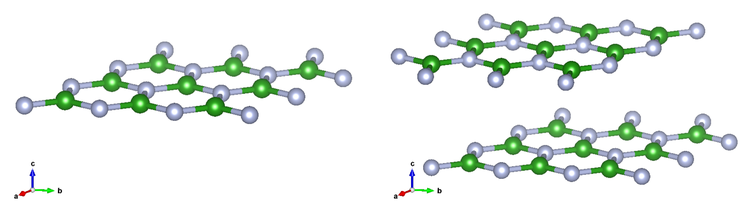


Figure 1: Monolayer and bilayer hBN

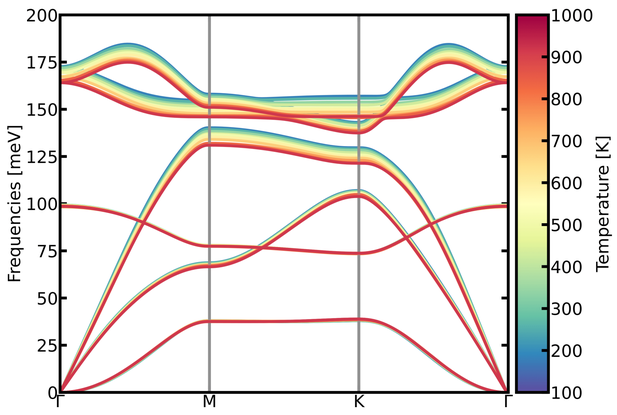


Figure 2: Monolayer hBN ML-MD TDEP phonon band structure

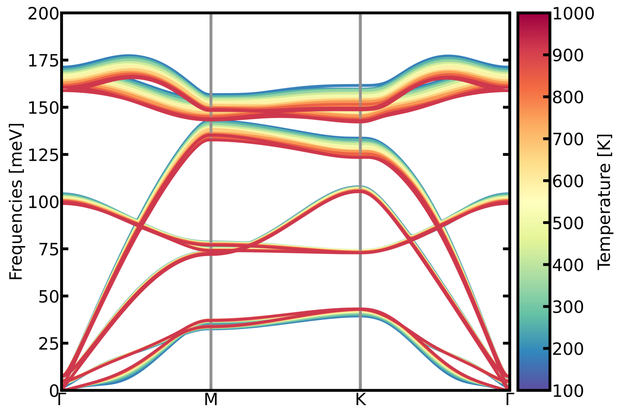


Figure 3: Bilayer hBN ML-MD TDEP phonon band structure

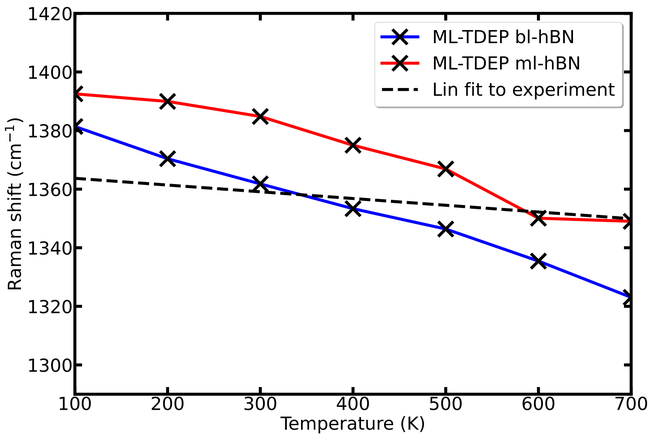


Figure 4: Mono and bilayer hBN Raman shift vs lin. fit of exp. data

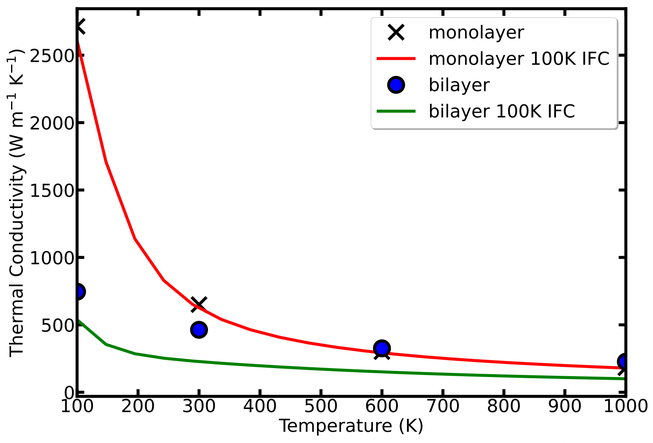


Figure 5: κ for T dependent and fixed IFC phonons

MLIP trained on 65 DFT configurations:

- Test set very well described by the MLIP.
- Phonon band structure in agreement with literature [5, 6].
- Decrease in the Raman shift of the E_{2g} mode with T approximately follows bulk trend as expected for both mono and bilayer [5, 7].
- Low frequency interlayer modes appear to harden with T.

Thermal conductivity calculated with TDEP (iterative Boltzmann):

- Monolayer agrees with [8] for both calculations in order of magnitude.
- For T dependent IFCs the bilayer's κ surpasses the monolayer's in the high T limit.

5. Conclusions And Outlook

- Phonon band structure, Raman shift and thermal conductivity agree with literature for all calculated observables for at least order of magnitude.
- Calculation is substantially faster: $\sim$ 10 days for full workflow vs $\sim$ 20 years of aiMD propagation.
- Other observables easily available (thermal expansion, Gruneisen parameters, etc.).
- Workflow also applicable to magnetic materials! (See poster by Aloïs Castellano)

References

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[7] I Stenger et al. *2D Materials*, 4(3):031003, jun 2017.

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