Beam damage and dynamics modelled with equivariant neural networks

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Background incl. aims

Molecular Dynamics (MD) simulations require a model for the interatomic interactions that is both accurate and computationally inexpensive. Density Functional Theory (DFT) offers good accuracy and decent computational performance for small systems, and ab-initio molecular dynamics (AIMD) is now possible based on DFT. However, many simulations require system sizes and/or simulation times that makes AIMD impossible.

Machine learning potentials can be used to generate inter-atomic potentials for specific systems, with accuracy close to that of DFT and computational efficiency close to that of classical interatomic potentials [1]. While such machine learning potentials have been used to extend the size and time scale of MD simulations to obtain adequate sampling of structures and thermodynamical properties, they suffer from the need of very large training sets and depend on the quality of the rotationally invariant descriptors used as input to the neural networks [2]. Equivariant Neural Networks change this dramatically [3], cutting down the amount of training data by several orders of magnitude while giving better accuracy. In an equivariant network, vector quantities can be used directly as input to the network and are then processed in a rotationally equivariant way.

We apply these methods to two different scientific problems. The first is heating of supported metallic nanoparticles by the electron beam in HR-TEM. The second is thermal vibrations in supported 2D nanoparticles, where recent HR-TEM studies show significantly increased vibrations near the edges of the nanoparticles [4].

Methods

We develop a simulation protocol for training Equivariant Neural Network Potentials (ENNPs) on a limited number of DFT simulations, based on the NequIP package [3]. We gradually expand the training set based on Molecular Dynamics simulations, where we use an ensemble of ENNPs to identify configurations where the uncertainty is large. DFT calculations are then added to the training data, gradually improving the ENNPs. After a few generations, we have a stable ENNP, but can still use the ensemble method to monitor the accuracy, also when simulating systems beyond the size limitations of DFT.

We simulate the heat transport between gold nanoparticles and TiO_2 support. From this we make a simple model for the temperature of the nanoparticle based on the heat deposited by the electron beam and the heat transport into the substrate. Parameters for the energy deposition by the beam are extracted from electron energy-loss spectroscopy (EELS), whereas the heat transport parameters come from the MD simulation.

Results

We simulate the heat transport between gold nanoparticles and TiO_2 support with molecular dynamics and ENNP potentials. We see that thermal transport is limited by the interface between nanoparticle and support, due to the mismatch in phonon frequencies. From this we make a simple model for the temperature of the nanoparticle based on the heat deposited by the electron beam and the heat transport into the substrate. Parameters for the energy deposition by the beam are extracted from EELS data, whereas the heat transport parameters come from the MD simulation [5].

We also simulate thermal vibrations in nanoparticles of the two-dimensional material MoS_2, supported on graphite. We use the simulated vibrational amplitudes as input to HR-TEM image simulations using the multislice algorithm and the frozen phonon formalism for vibrations. We find that edge phonon states significantly increase the vibrational amplitude not just at the edge of the nanoparticle, but several lattice constants away from the edge. This matches well what has been found experimentally by Chen et al. [4], where exit wave reconstructions were used to gauge vibrational amplitudes in HR-TEM image series of supported MoS_2 nanoparticles.

Conclusion

Equivariant Neural Network Potentials make it possible to use molecular dynamics simulations on systems with tens to hundreds of thousands of atoms, with accuracy close to that of DFT, but without the computational cost of DFT, where even a thousand atoms are a challenge. The accuracy of the potentials can be monitored with ensemble methods, and problematic configurations can be used to retrain the ENNP. The method can be directly applied to study effects of the electron beam in HR-TEM, and the role of realistic atomic vibrations in imaging.

Keywords

Equivariant Neural Network Potentials, HR-TEM, image simulation, molecular dynamics, nanoparticles.

Keywords:

Molecular Dynamics, HR-TEM, Machine Learning

Reference:

[1] J. Behler and M. Parrinello, Generalized Neural-Network Representation of High-Dimensional Potential-Energy Surfaces, Phys. Rev. Lett. 98, 146401 (2007).

[2] A. E. G. Mikkelsen et al. Is the water/Pt(111) interface ordered at room temperature? J Chem Phys 155, 224701 (2021).

[3] S. Batzner et al. E(3)-equivariant graph neural networks for data-efficient and accurate interatomic potentials. Nat. Commun. 13, 2453 (2022).

[4] F.-R. Chen et al. Probing atom dynamics of excited Co-Mo-S nanocrystals in 3D. Nat. Commun. 12, 5007 (2021).

[5] C. Nuñez Valencia et al. Beam induced heating in electron microscopy modeled with machine learning interatomic potentials. Nanoscale, 16, 5750 (2024).