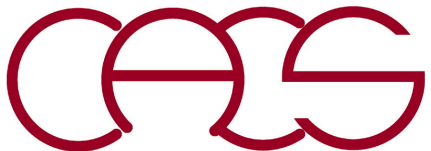


Path-Integral Molecular Dynamics

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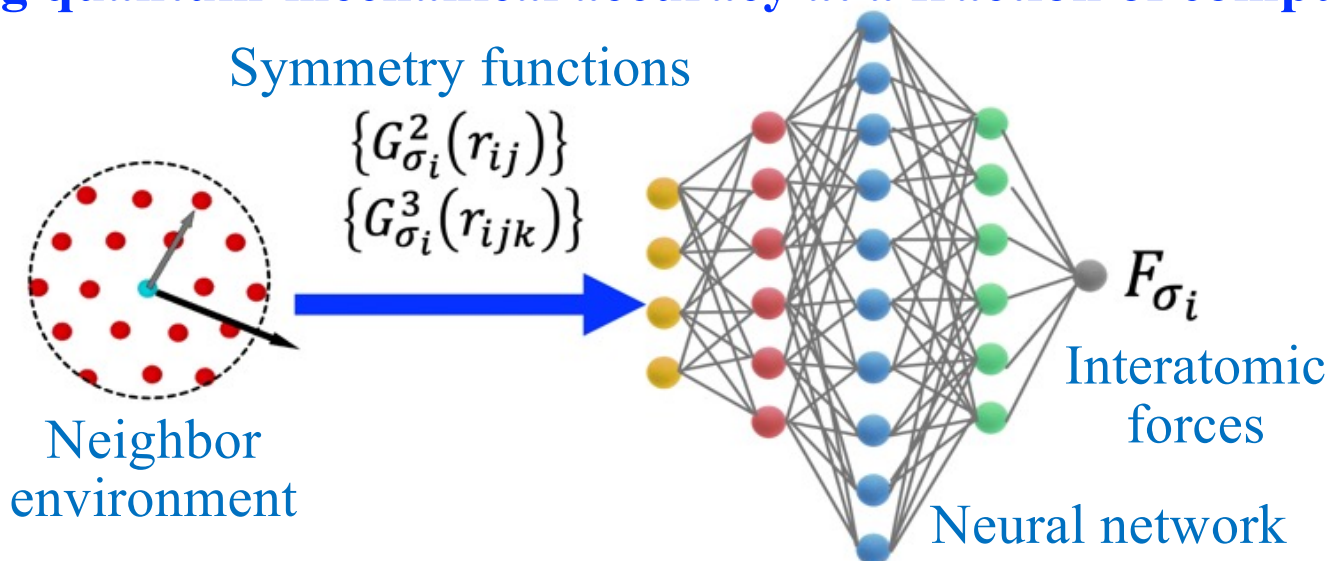


https://wp.nyu.edu/tuckerman_group/movies/

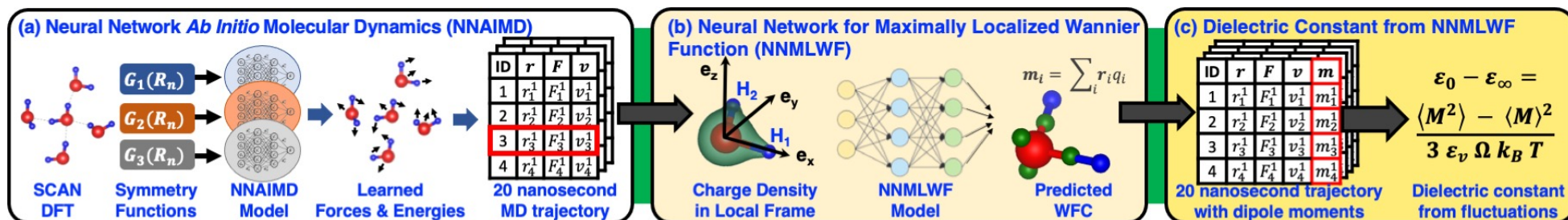


Neural-Network Quantum Molecular Dynamics

- NNQMD@scale could revolutionize atomistic modeling of materials, providing quantum-mechanical accuracy at a fraction of computational cost

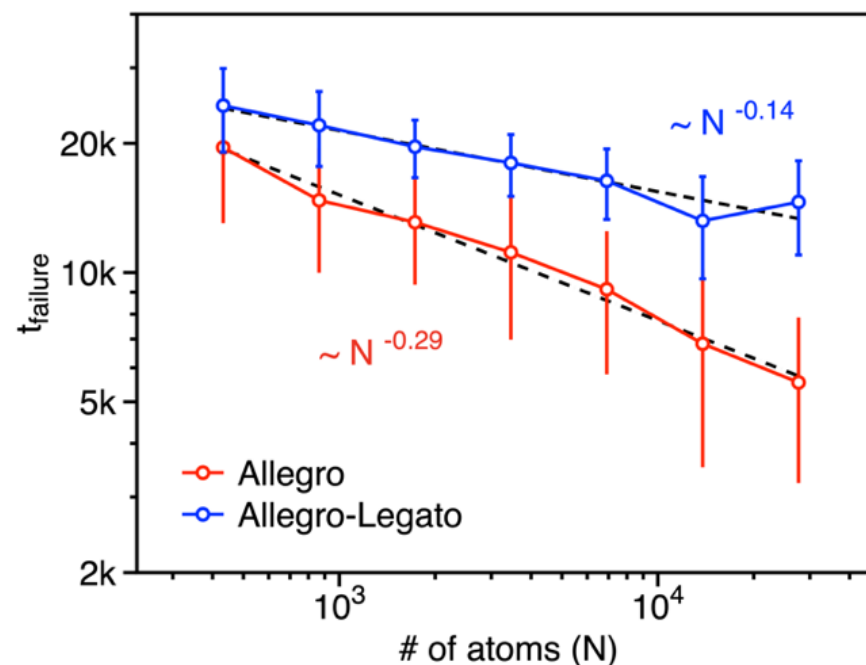
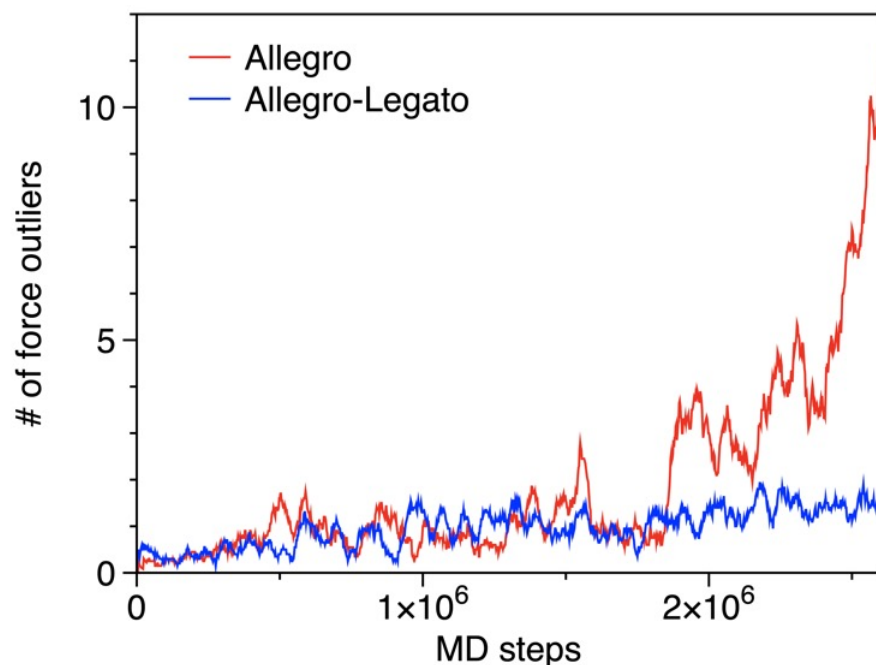


- Neural networks predict: (1) atomic forces for performing MD simulations; & (2) maximally-localized Wannier-function (MLWF) centers for computing quantum properties like electronic dipoles



Fast & Robust NNQMD: Allegro-Legato

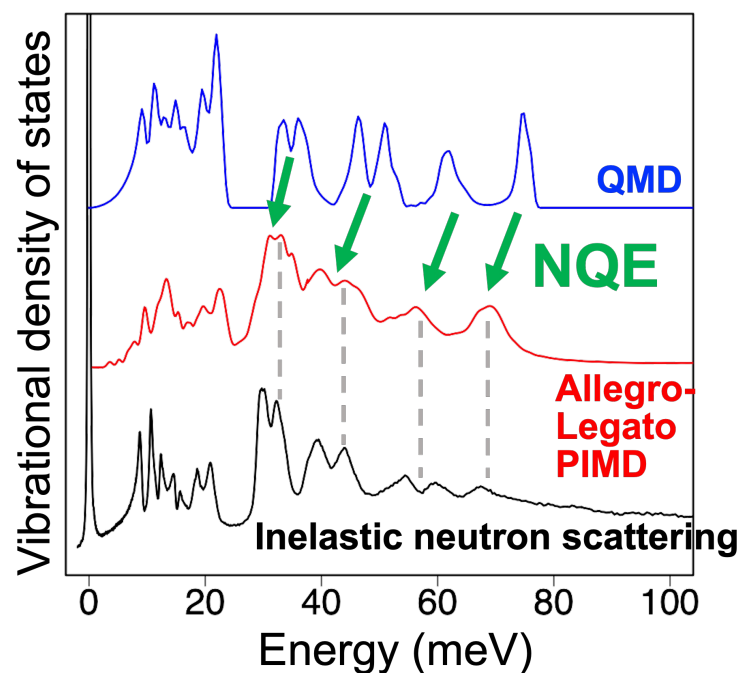
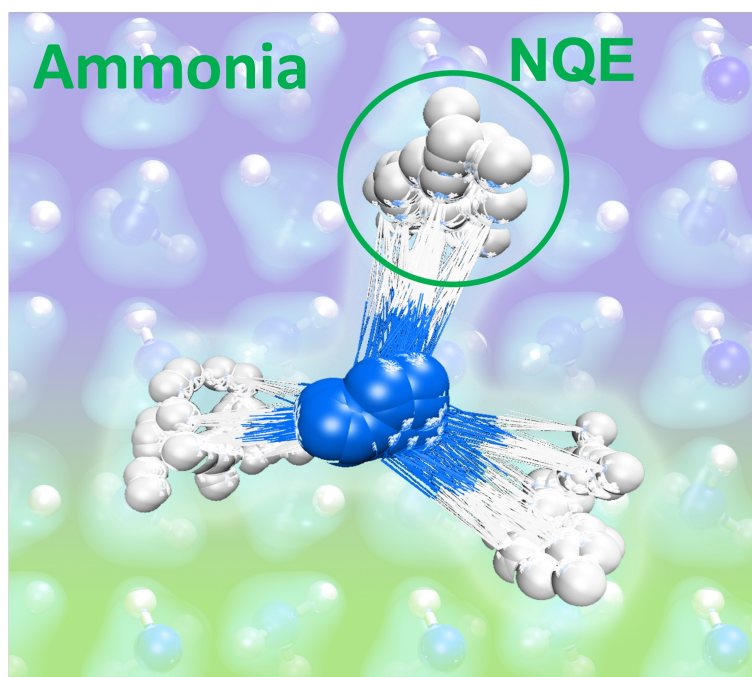
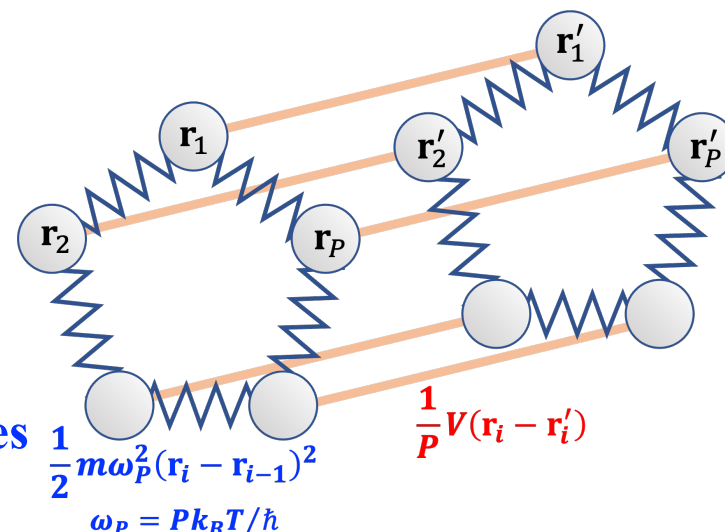
- **Allegro (fast) NNQMD:** State-of-the-art *accuracy & speed* founded on group-theoretical equivariance & local descriptors [Musaelian *et al.*, *Nat. Commun.* **14**, 579 ('23)]
- **Fidelity-scaling problem:** On massively parallel computers, growing number of unphysical (adversarial) force predictions prohibits simulations involving larger numbers of atoms for longer times
- **Allegro-Legato (fast and “smooth”):** *Sharpness aware minimization (SAM)* enhances the *robustness* of Allegro through improved smoothness of loss landscape
$$\mathbf{w}_* = \operatorname{argmin}_{\mathbf{w}} [L(\mathbf{w}) + \max_{\|\epsilon\|_2 \leq \rho} \{L(\mathbf{w} + \epsilon) - L(\mathbf{w})\}] \quad (L: \text{loss}; \mathbf{w}: \text{model parameters})$$
- **Elongated time-to-failure scaling, $t_{\text{failure}} = O(N^{-\beta})$,** without sacrificing accuracy or speed, thereby achieving spectroscopically stable long-time Hamiltonian trajectory



Ibayashi *et al.*, ISC 2023 (arXiv: 2303.08169)

Nuclear-Quantum NNQMD

- **Allegro-Legato-PIMD**: Incorporate nuclear quantum effect (NQE) through path-integral molecular dynamics (PIMD)
- NNQMD trained by QMD achieves the required large number (P) of replicas at low temperature & long-time Hamiltonian dynamics to resolve fine vibrational structures
- NQE down-shifts inter-molecular vibrational modes in ammonia to explain high-resolution inelastic neutron scattering experiments



Linker *et al.*, *Nature Commun.* **15**, 3911 ('24)