Path-Integral Molecular Dynamics

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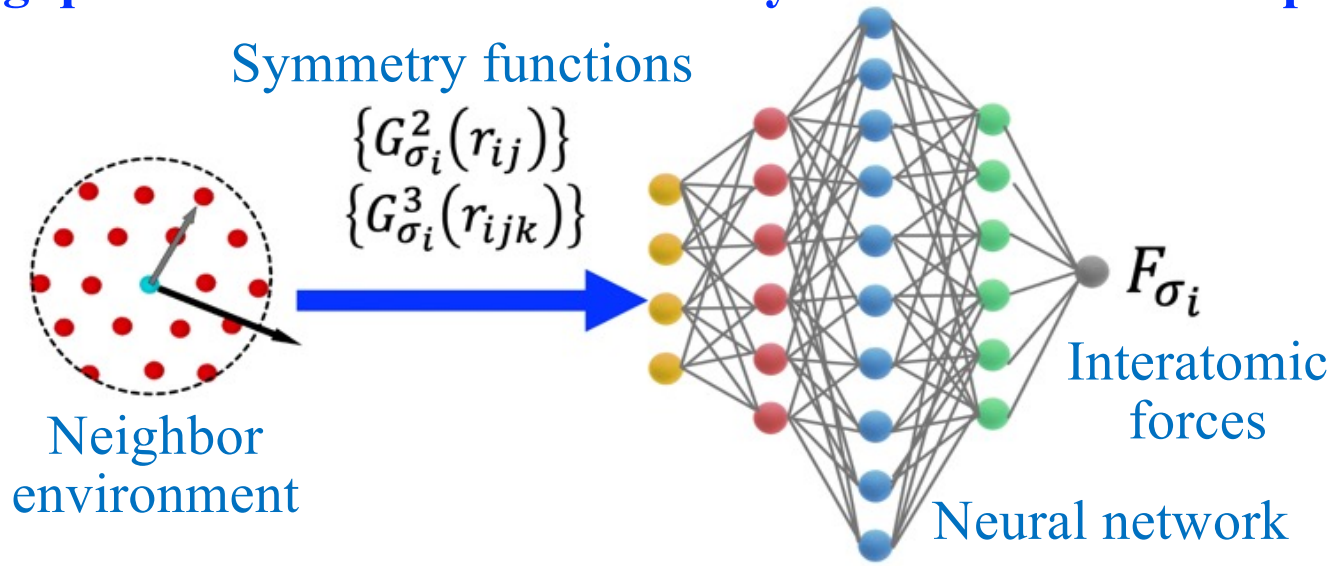
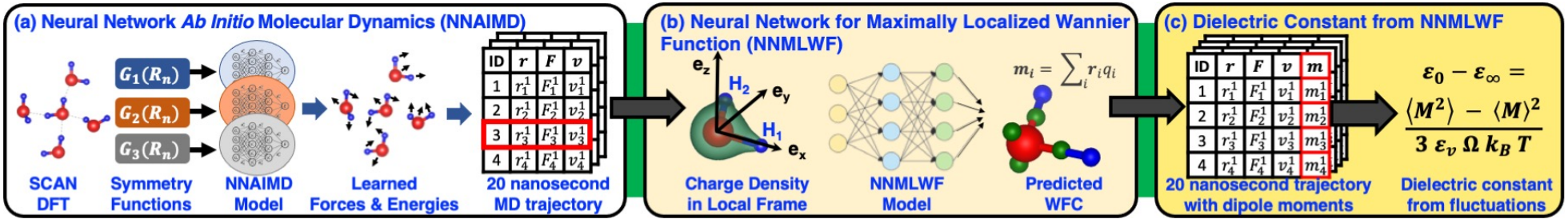
Neural-Network Quantum Molecular Dynamics

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



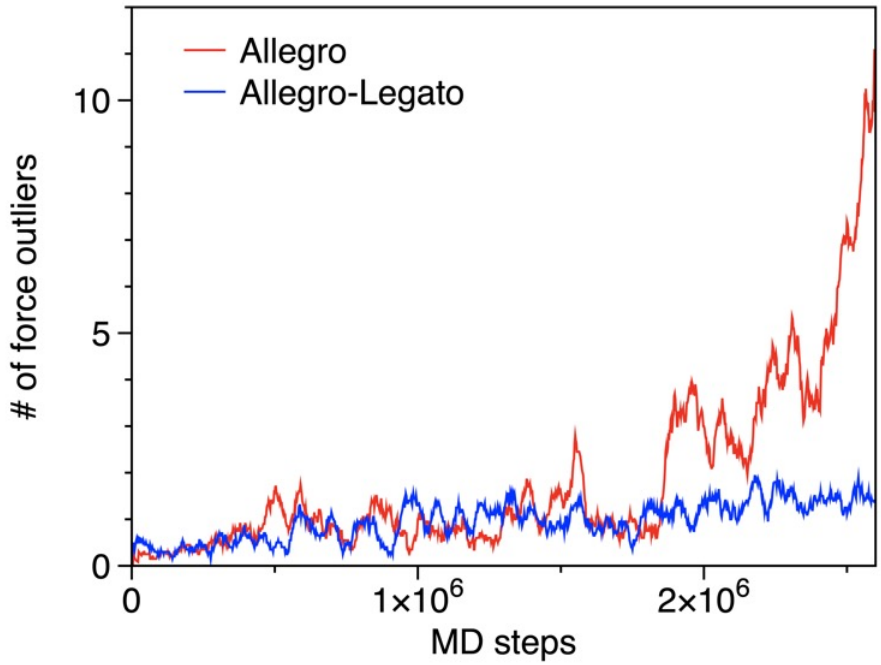
- **Neural networks predict: (1) atomic forces for performing MD simulations; & (2) maximally-localized Wannier-function (MLWF) centers for computing quantum properties like electronic dipoles**

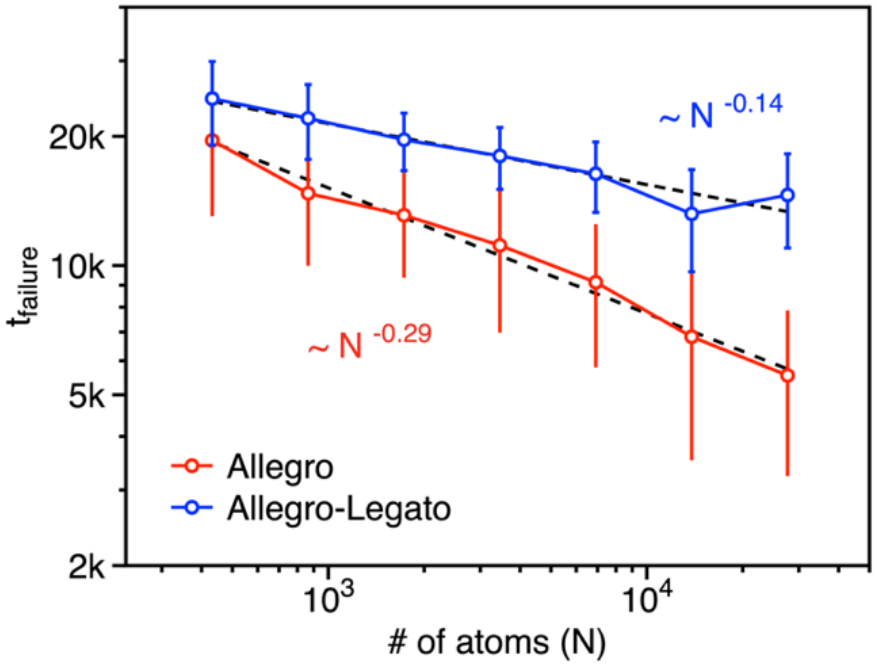


Krishnamoorthy *et al.*, *Phys. Rev. Lett.* **126**, 216403 ('21)

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**
  $\mathbf{w}_* = \mathrm{argmin}_{\mathbf{w}}\left[L(\mathbf{w}) + \max_{\|\epsilon\|_2 \le \rho}\{L(\mathbf{w}+\boldsymbol{\epsilon}) - L(\mathbf{w})\}\right]$ (L: loss; $\mathbf{w}$: model parameters)
- **Elongated time-to-failure scaling, $t_{\mathrm{failure}} = O(N^{-\beta})$, without sacrificing accuracy or speed, thereby achieving spectroscopically stable long-time Hamiltonian trajectory**

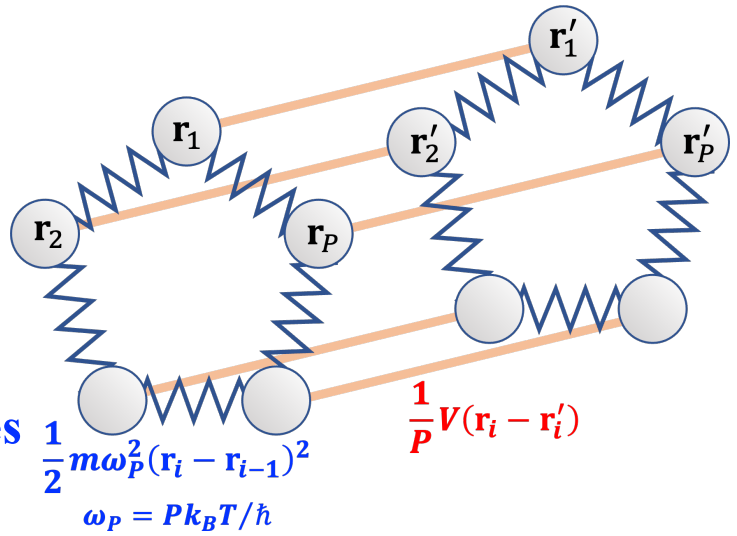


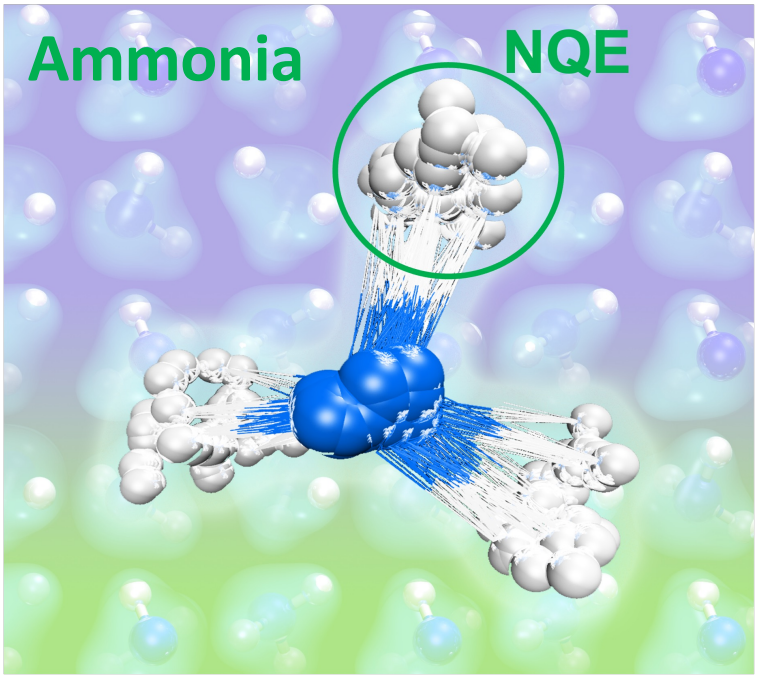


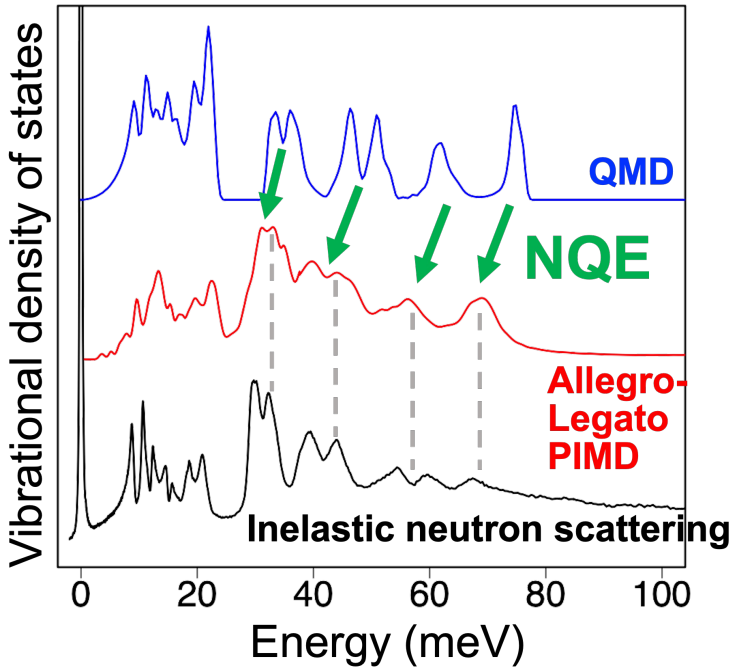
Ibayashi *et al.*, *ISC 2023* (*arXiv*: 2303.08169)

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)