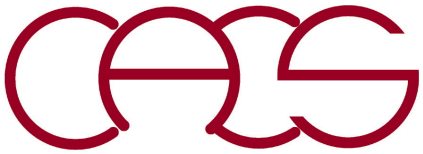
Neural-Network Quantum Molecular Dynamics

Aiichiro Nakano

Collaboratory for Advanced Computing & Simulations
Department of Computer Science
Department of Physics & Astronomy
Department of Quantitative & Computational Biology
University of Southern California

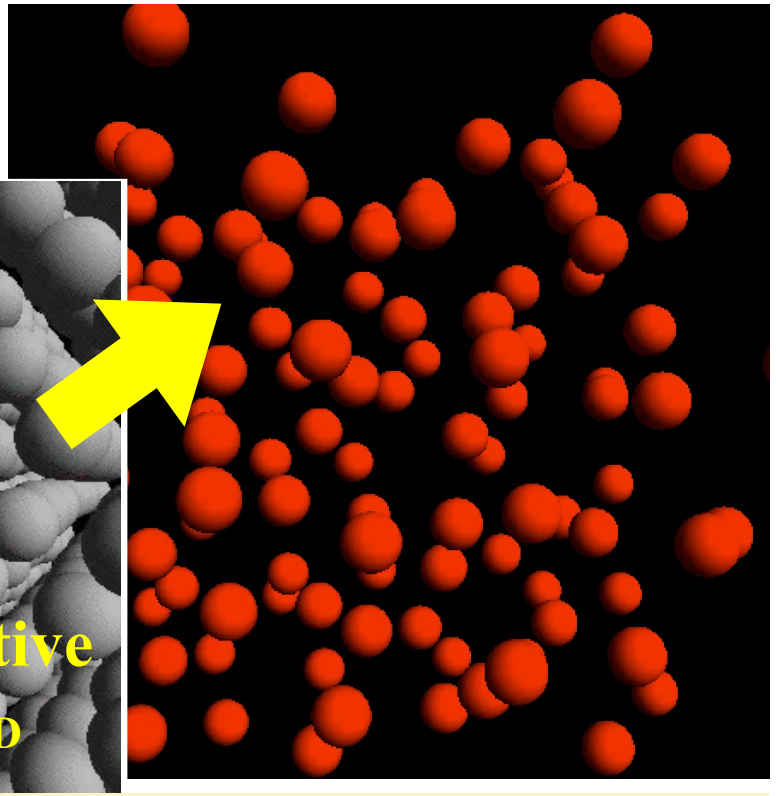
Email: anakano@usc.edu

Replace expensive electronic wave functions by deep neural networks to achieve orders-of-magnitude speedup

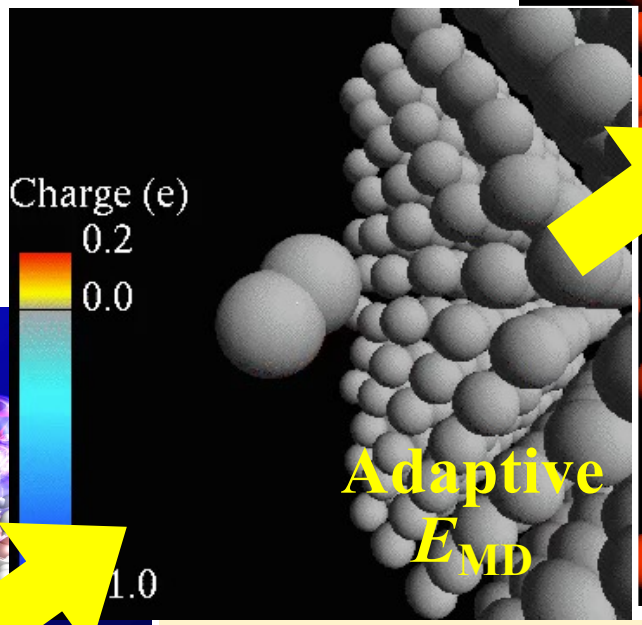


Molecular Dynamics & Machine Learning

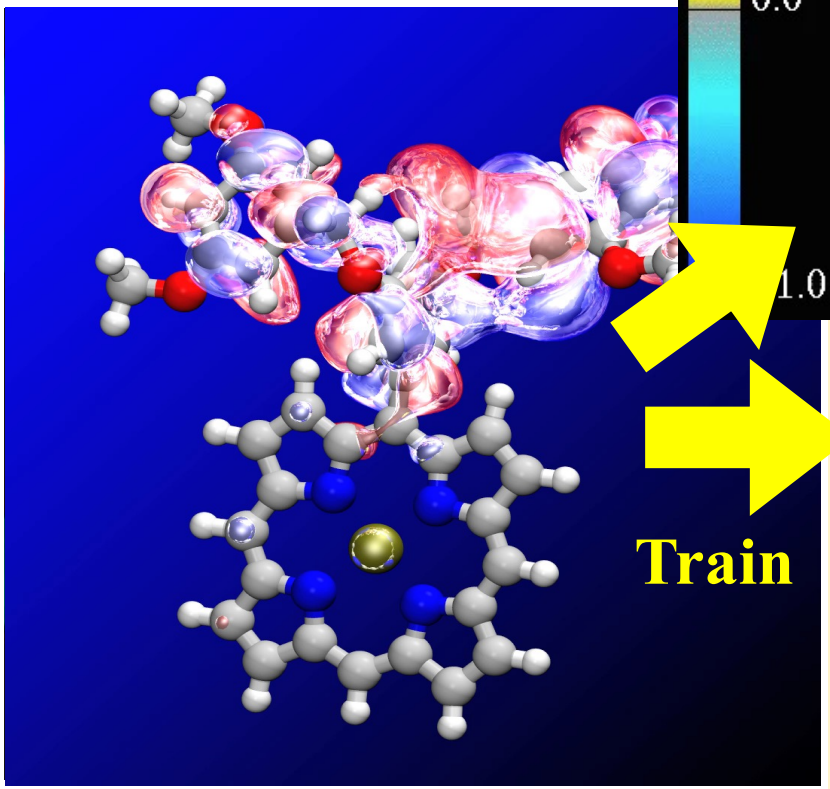
Molecular Dynamics (*MD*)



Reactive MD (*RMD*)

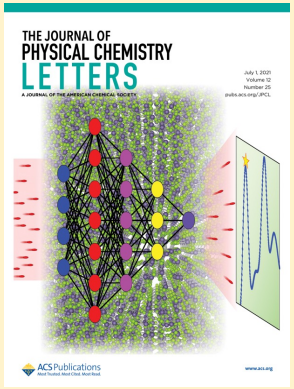


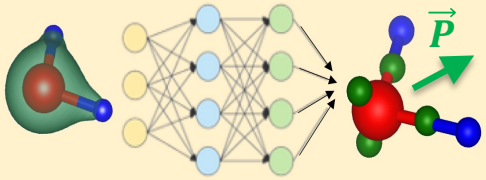
Nonadiabatic quantum MD (*NAQMD*)



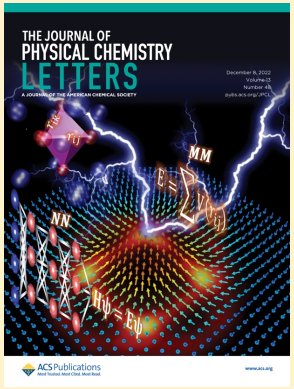
Train

First principles-based neural-network quantum molecular dynamics (*NNQMD*)



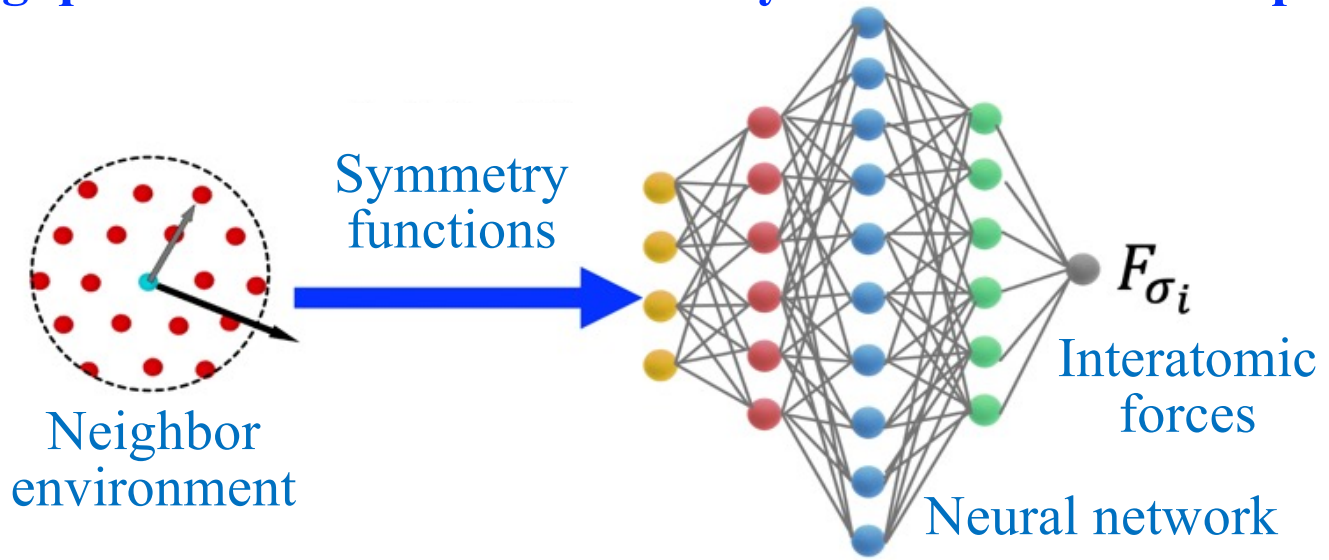


Physical Review Letters
Editor's choice
(May 25, 2021)

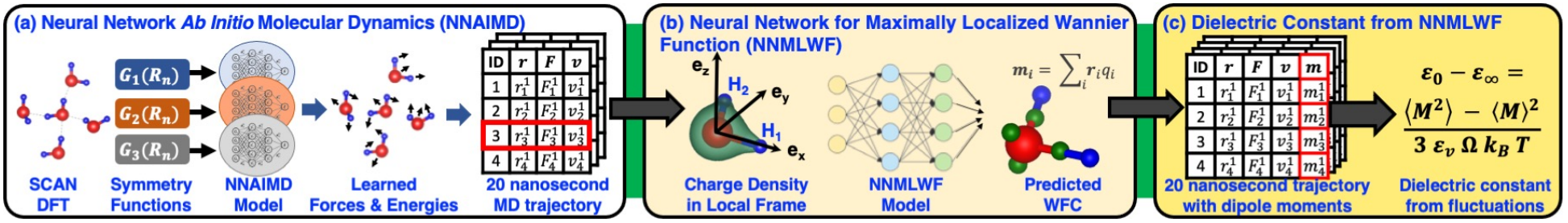


Neural-Network Quantum Molecular Dynamics

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



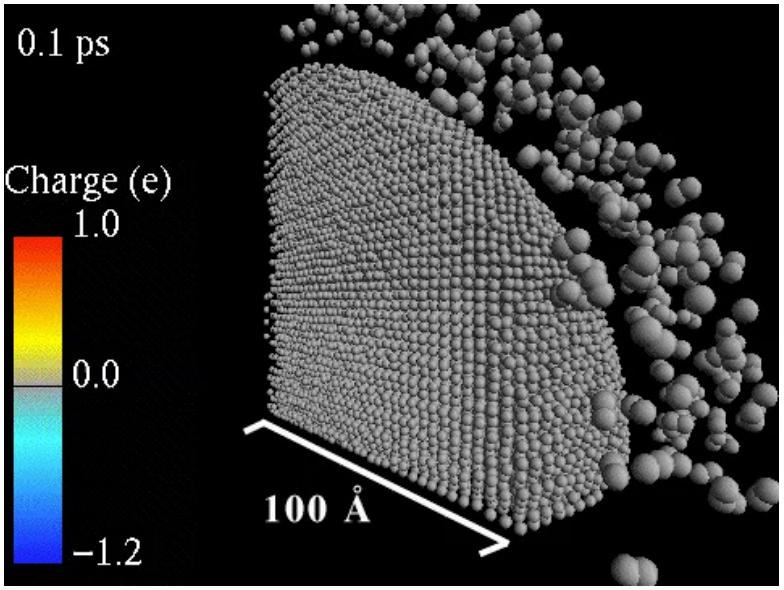
- **Neural networks predict: (1) atomic forces for performing MD simulations; & (2) maximally-localized Wannier-function (MLWF) centers for computing quantum properties like electronic dipoles**



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

Charge-Transfer NNQMD

- **Incorporated charge transfer for accurately describing chemical reactions through charge equilibration (QEq) in reactive molecular dynamics (RMD)**



Campbell, *Phys. Rev. Lett.* **82**, 4866 ('99)

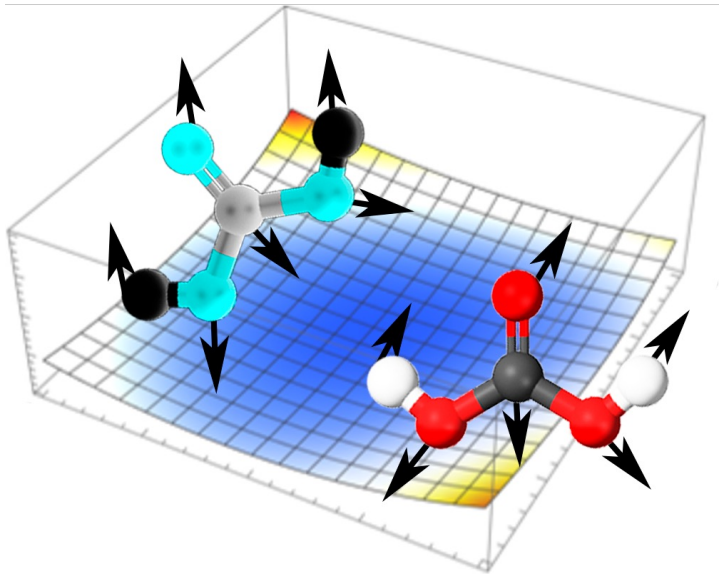
$$q_*^N = \underset{q^N}{\operatorname{argmin}} E_{\text{Coulomb}}(\mathbf{r}^N, q^N)\ s.t. \Sigma_i q_i = 0$$

$$E_{\text{Coulomb}}(\mathbf{r}^N, q^N) = \Sigma_i \chi_i q_i + \frac{1}{2}\Sigma_{i,j}\, q_i H(r_{ij}) q_j$$

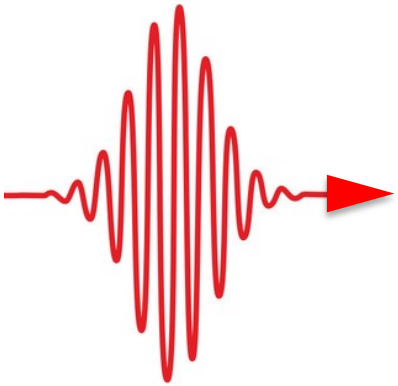
Electronegativity — Coulombic interaction

Rappe, *J. Phys. Chem.* **95**, 3358 ('91); van Duin, *J. Phys. Chem. A* **105**, 9396 ('01)
Nakano, *Comput. Phys. Commun.* **104**, 59 ('97); Nomura, *ibid.* **192**, 91 ('15)

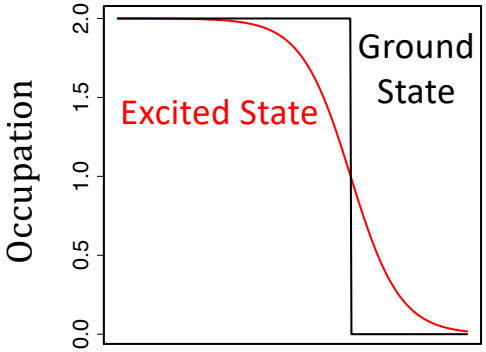
- **4G NNQMD: Separate neural network to predict electronegativity**
  Ko, *Nat. Commun.* **12**, 398 ('21)
- **Allegro-Legato-4G: Allegro-Legato to achieve smooth loss landscape & robust long-time dynamics in 4G-NNQMD**



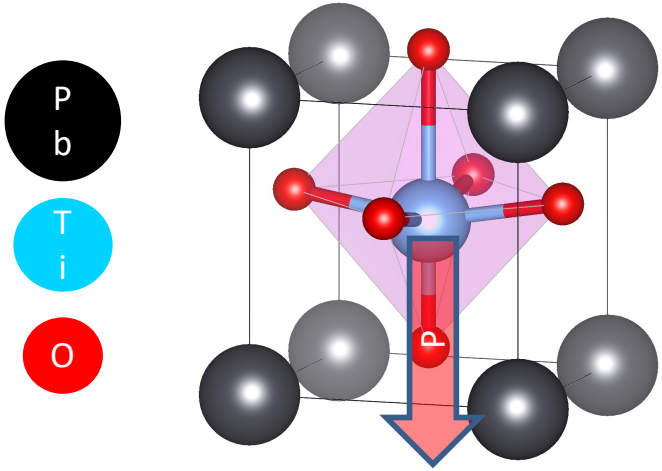
Multiscaling from NAQMD to XS-NNQMD



Electronic excitation by ultrafast laser pulse



Change in electronic occupation due to electron-electron interaction



Change in polarization dynamics due to electron-ion interaction

Maxwell + RT-TDDFT → Excitation density & effective electronic temperature from strong light-electron (e) interaction & e-e scattering

Excited-state QMD → Medium spatiotemporal scale excited electron-phonon (e-ph) dynamics

Large-scale XS-NNQMD → Large-scale structural change & defect formation

*DC-MESH: divide-&-conquer Maxwell-Ehrenfest-surface hopping NAQMD

DC-MESH* → **Excited-state (XS) NNQMD**

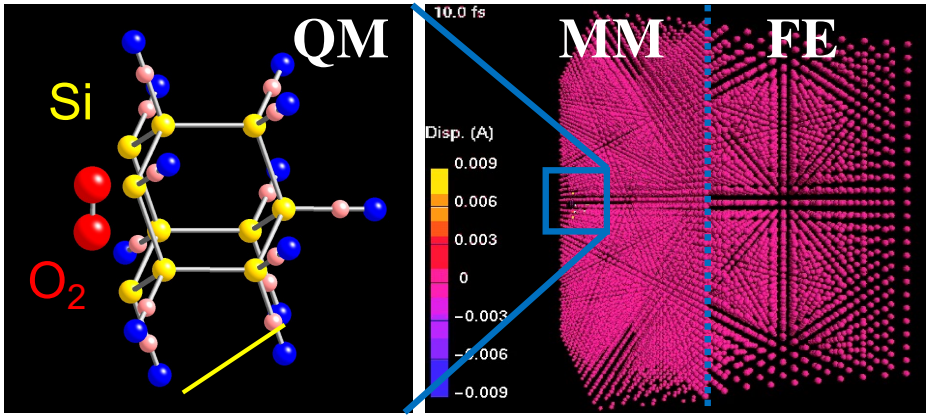
Excited energy landscape

Linker *et al.*, *Science Advances* **8**, eabk2625 ('22)

Multiscale QM/MM → NN/MM

- **Multiscale quantum challenge:** Complex response of ferroelectric topological defects to external stimuli encompasses picosecond-to-nanosecond time & nanometer-to-micrometer length scales
- **QM/MM:** Overcame the challenge taking cue from multiscale quantum-mechanics (QM)/molecular mechanics (MM) approach (2013 Nobel chemistry prize)

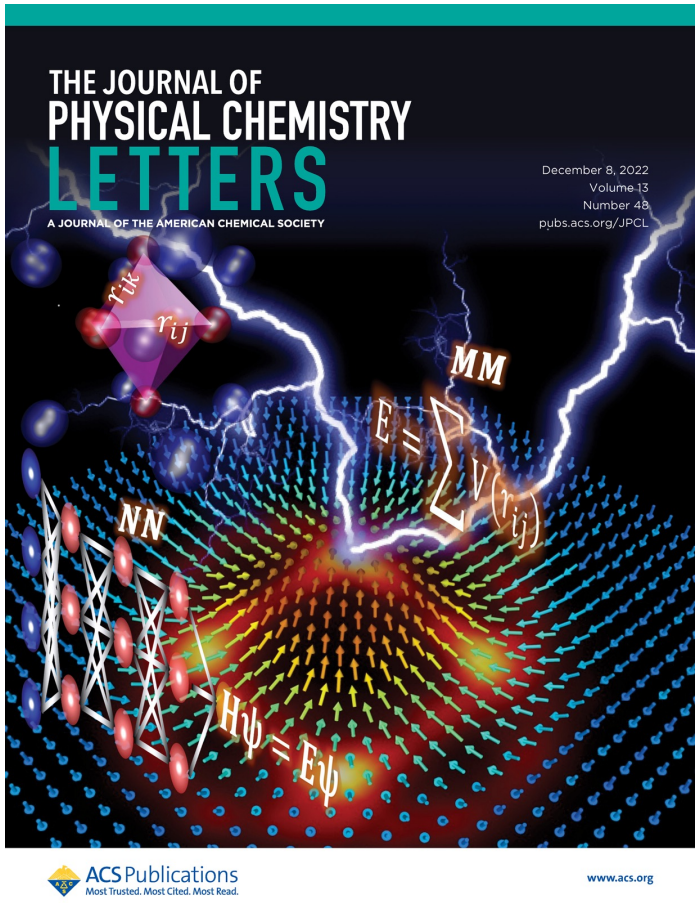
  Warshel, *Angew. Chem.* **53**, 10020 ('14)



QM/MM/FE (finite-element method)

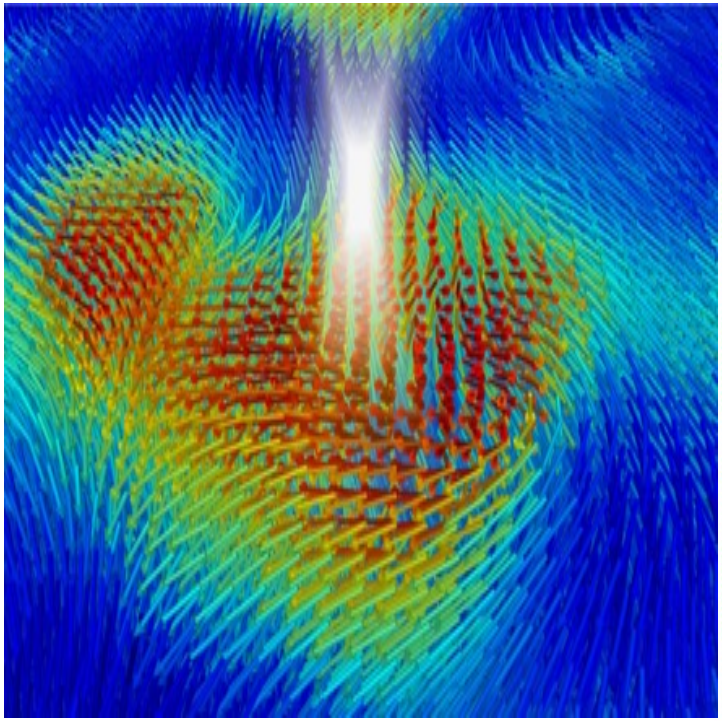
Ogata *et al*, *Comput. Phys. Commun.* **138**, 143 ('01)

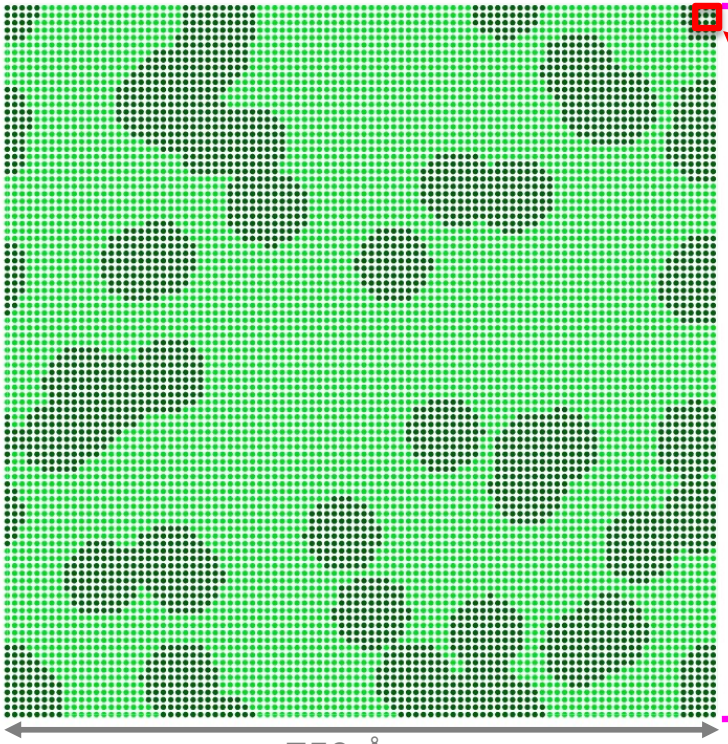
- **NN/MM:** NNQMD for ferroelectric ($PbTiO_3$: PTO) embedded in MM for paraelectric ($SrTiO_3$: STO) to apply appropriate strain boundary condition



Linker *et al.*, *J. Phys. Chem. Lett.* **13**, 11335 ('22); *Nano Lett.* **23**, 7456 ('23)

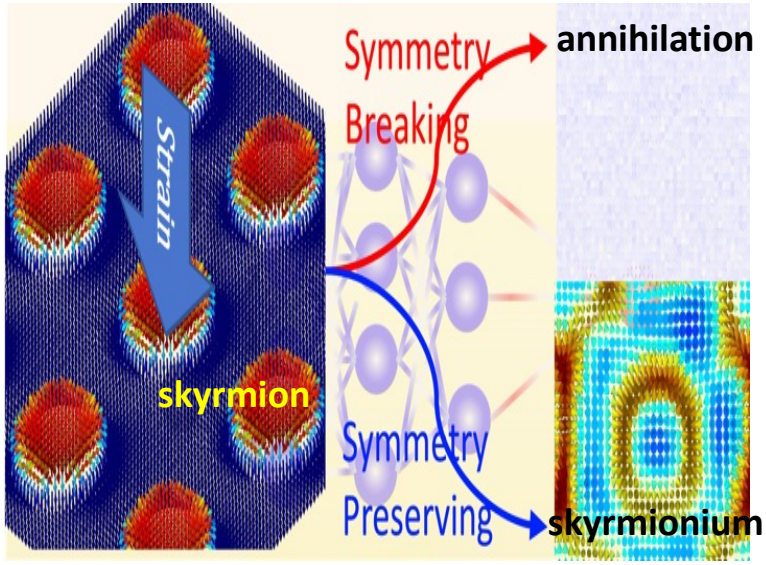
Application: Ferroelectric Opto-Topotronics





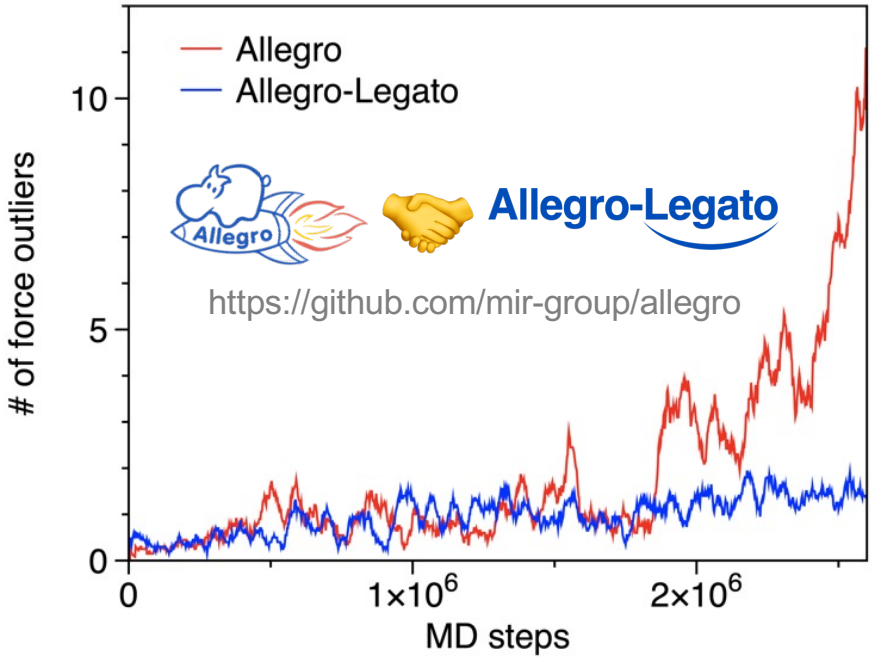
- **Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications**
- **Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)**
- **Symmetry-controlled skyrmion-to-skyrmionium*** **switching** *Composite of skyrmions with opposite topological charges

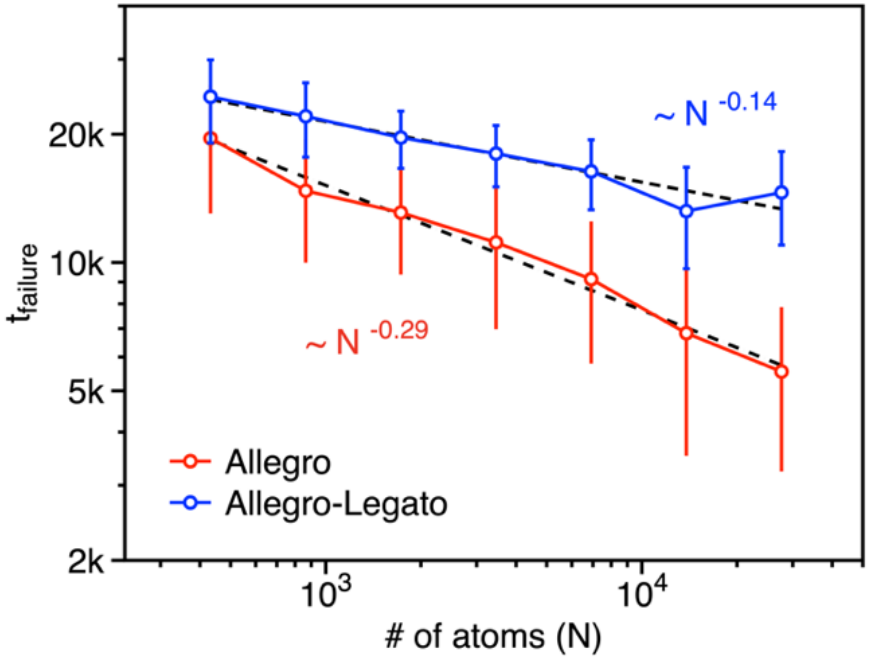
Linker *et al., Science Adv.* **8**, eabk2625 ('22); *JPCL* **13**, 11335 ('22); *Nano Lett.* **23**, 7456 ('23)



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

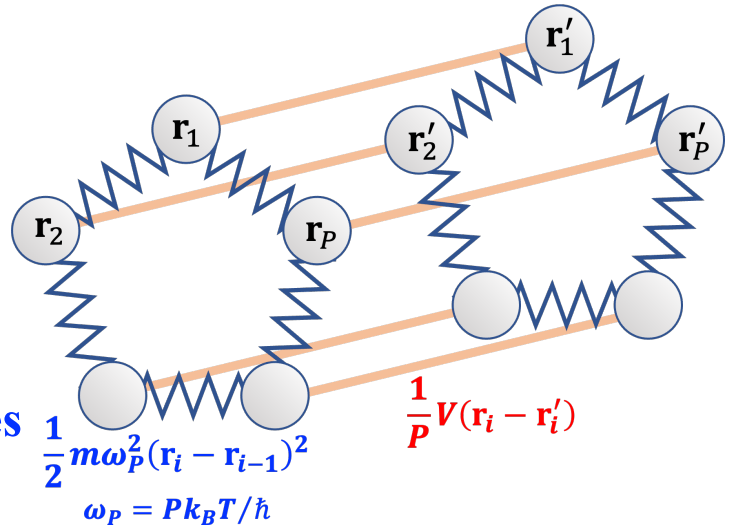


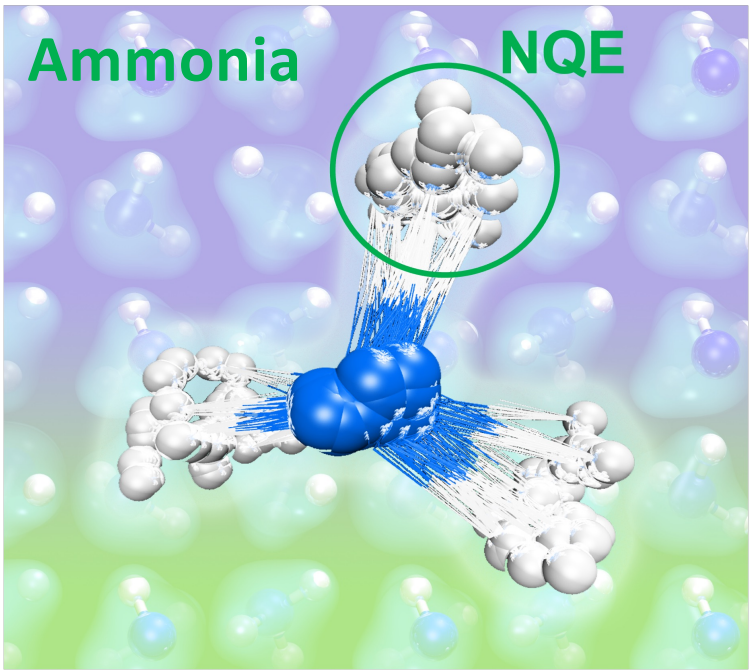


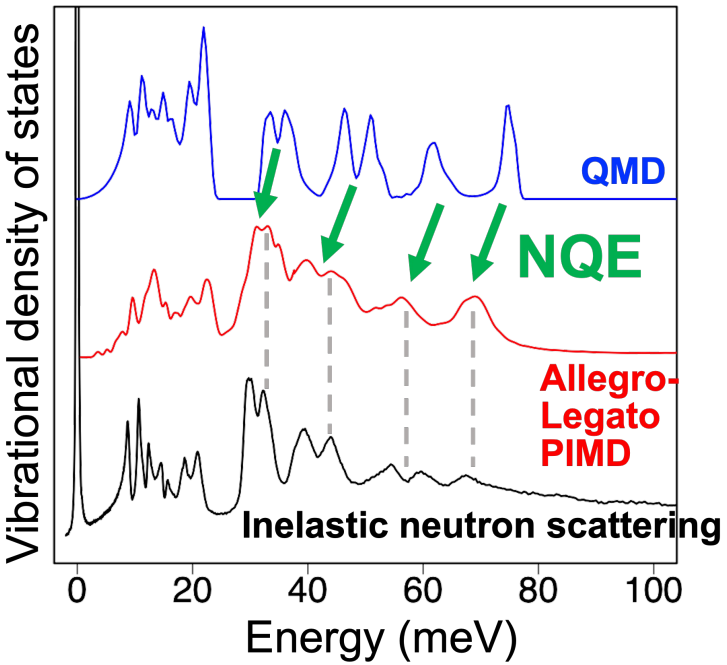
H. Ibayashi *et al.*, *ISC23—LNCS* **13948**, 223 ('23); *arXiv*: 2303.08169

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**







T. Linker *et al.*, *Nature Commun.* 15, 3911 ('24)