Quantum Materials Dynamics at the Nexus of Exascale Computing, Artificial Intelligence, and Quantum Computing

Aiichiro Nakano, Rajiv K. Kalia, Ken-ichi Nomura, Priya Vashishta, Anikeya Aditya, Lindsay Bassman, Hikaru Ibayashi, Thomas Linker, Ankit Mishra, Pankaj Rajak, Taufeq Razakh, Liqiu Yang

Collaboratory for Advanced Computing & Simulations
Depts. of Computer Science, Physics & Astronomy, Chemical Engineering & Materials Science, and Quantitative & Computational Biology
University of Southern California

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Collaborators:
F. Shimojo, K. Shimamura (*Kumamoto University, Japan*)

UCLA-IPAM Workshop on "Increasing the Length, Time, and Accuracy of Materials Modeling Using Exascale Computing"
Organizers: Dr. V. Ehrlacher, Dr. V. Gavini, Dr. D. Perez, Dr. S. Plimpton

March 27, 2023, Los Angeles, CA

Current & Future Supercomputing

- **Won two DOE supercomputing awards to develop & deploy metascalable ("design once, scale on future platforms") simulation algorithms**

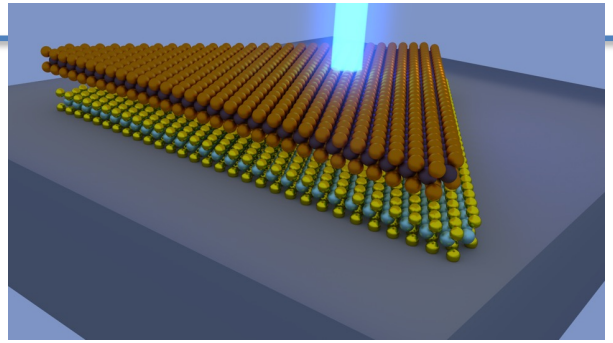
U.S. DEPARTMENT OF ENERGY
INCITE
LEADERSHIP COMPUTING
Innovative & Novel Computational Impact on Theory & Experiment

- **Atomistic simulations on million cores (pre-exascale)**

Title: AI-Guided Exascale Simulations of Quantum Materials Manufacturing and Control
PI and Co-PIs: Aiichiro Nakano–PI, Rajiv K. Kalia, Ken-ichi Nomura, Priya Vasishta

786,432-core IBM Blue Gene/Q
281,088-core Intel Xeon Phi
560-node (2,240-GPU) AMD/NVIDIA Polaris

AURORA | EARLY SCIENCE PROGRAM



Early Science Projects for Aurora Supercomputer Announced
Metascalable layered materials genome
Investigator: Aiichiro Nakano, University of Southern California

2 exaflop/s
Intel Aurora (forthcoming)

exaflop/s = 10^{18} mathematical operations per second

- **One of the initial simulation users of the next-generation DOE supercomputer**

Changing Computing Landscape for Science

Post-exascale Computing for Science

Compute Cambrian explosion

AI for Science

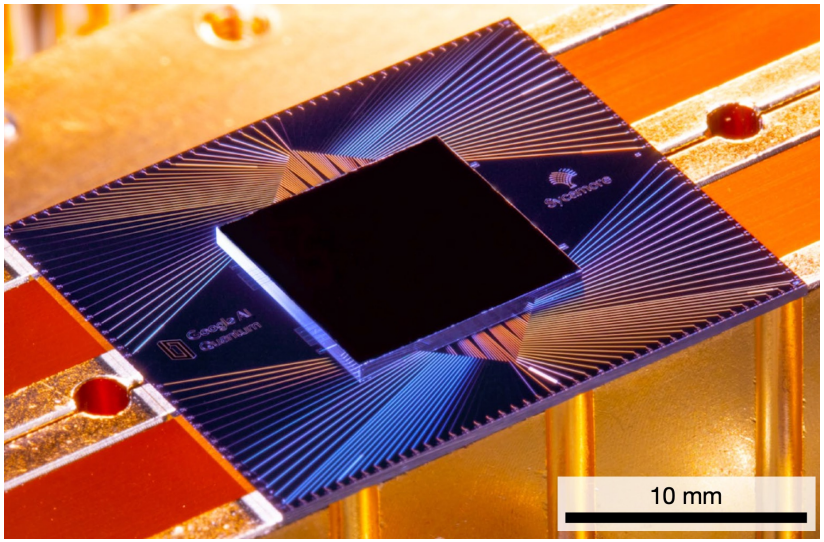
DOE readies multibillion-dollar AI push

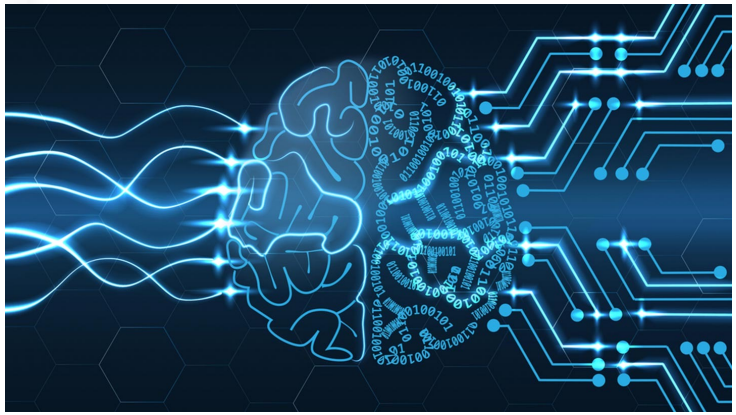
U.S. supercomputing leader is the latest big backer in a globally crowded field

By **Robert F. Service**, *in Washington, D.C.*

Science **366**, 559 (Nov. 1, '19)

Quantum Computing for Science

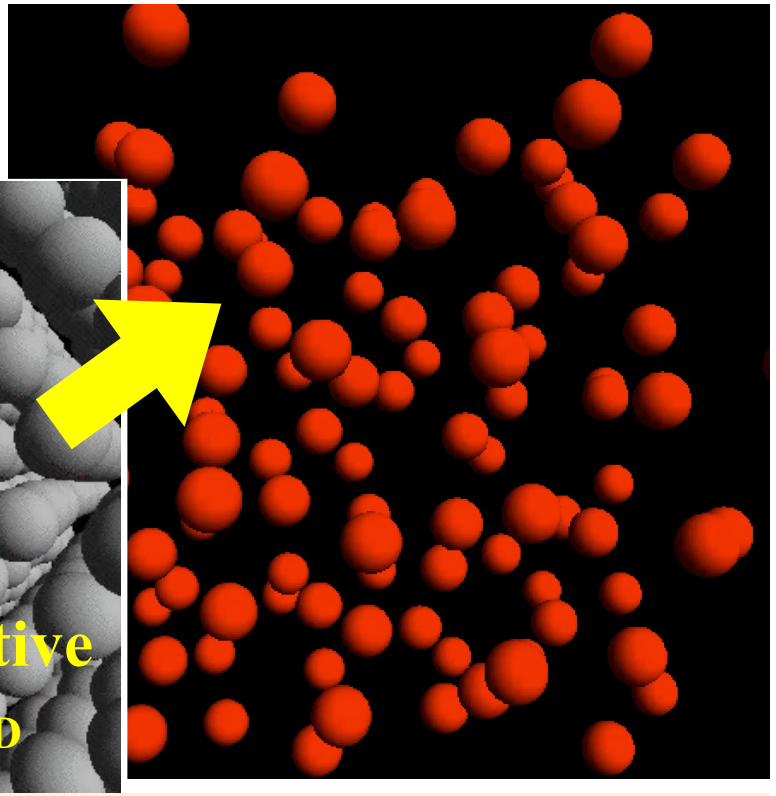




Use all to advance science!

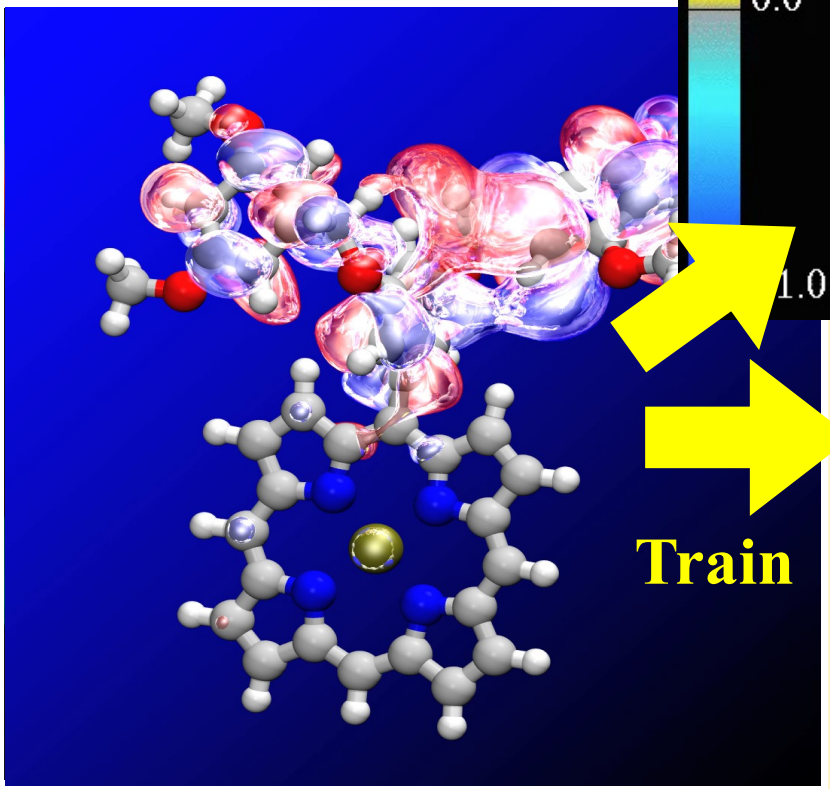
Molecular Dynamics & Machine Learning

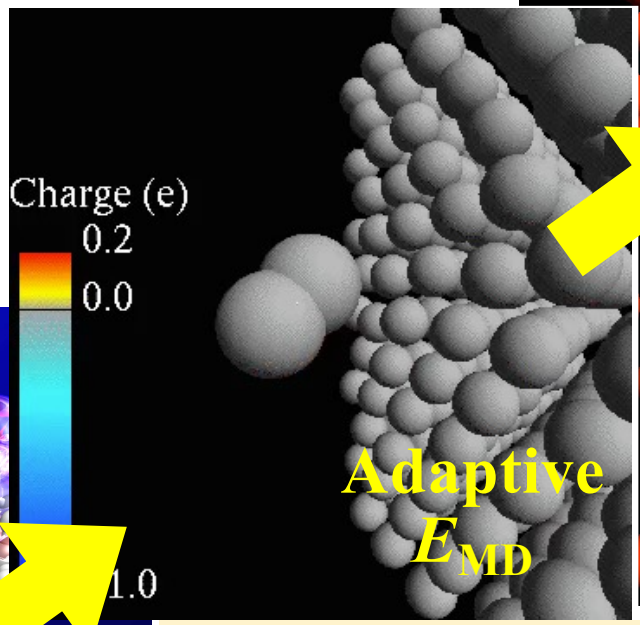
Molecular Dynamics (*MD*)



Reactive MD (*RMD*)

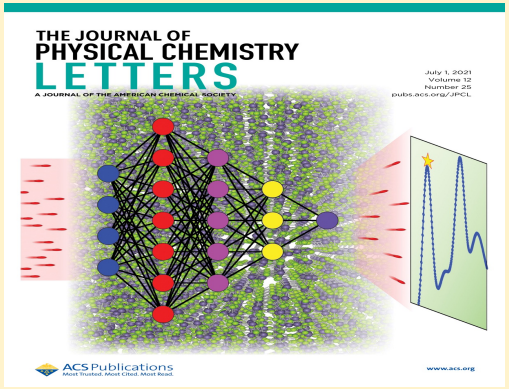
Nonadiabatic quantum MD (*NAQMD*)

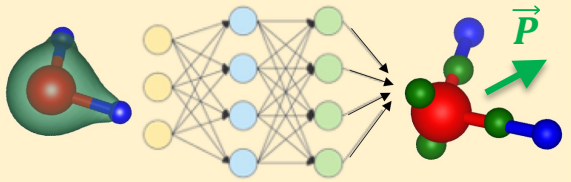




Train

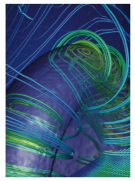
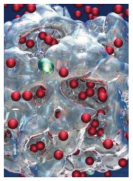
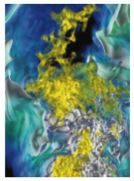
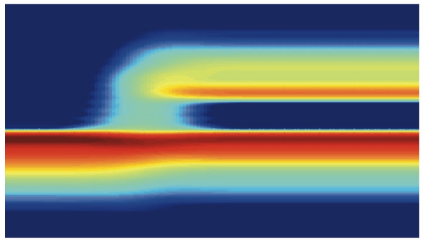
First principles-based neural-network quantum molecular dynamics (*NNQMD*)





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

BES

Exa-scale

BASIC ENERGY SCIENCES

EXASCALE REQUIREMENTS REVIEW

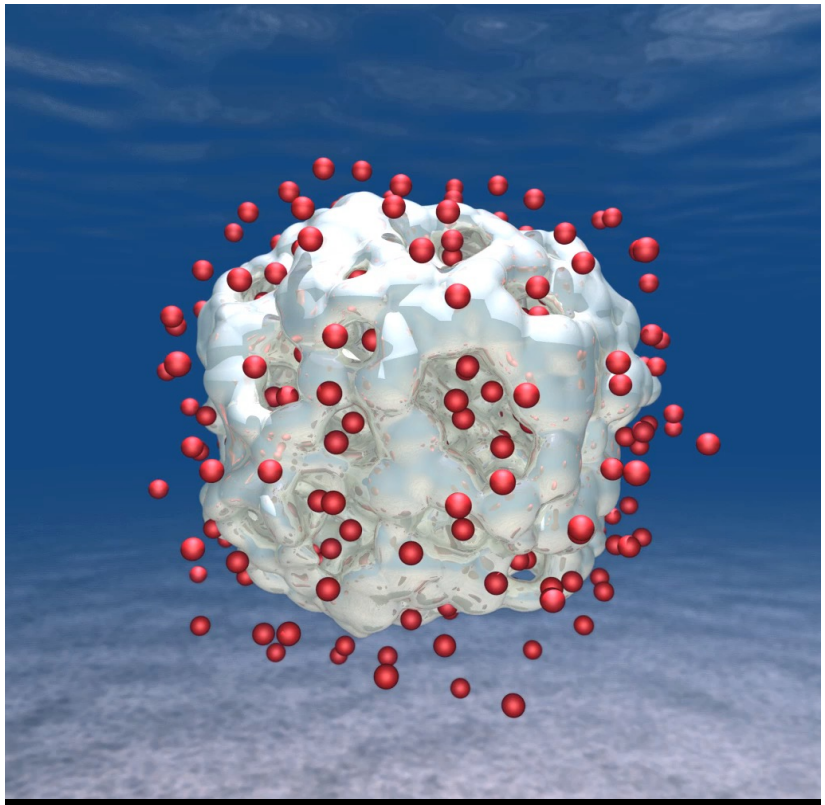
An Office of Science review sponsored jointly by
Advanced Scientific Computing Research and Basic Energy Sciences

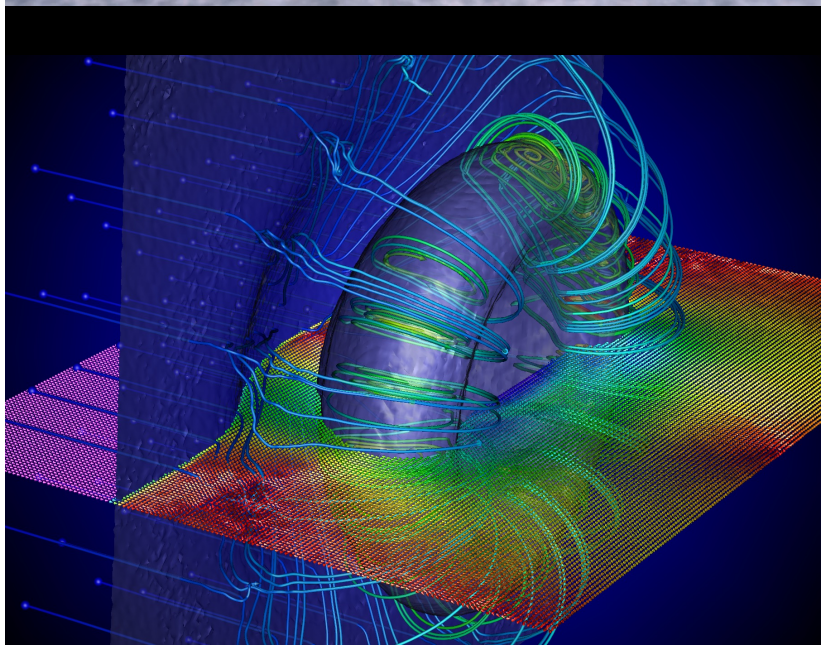
16,661-atom QMD

Shimamura *et al.*,
Nano Lett.
14, 4090 ('14)

10^9-atom RMD

Shekhar *et al.*,
Phys. Rev. Lett.
111, 184503 ('13)



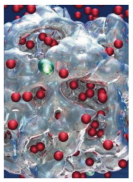
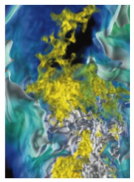
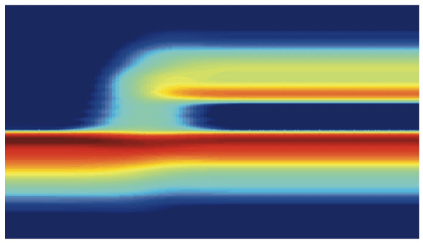


NOVEMBER 3–5, 2015

ROCKVILLE, MARYLAND

U.S. DEPARTMENT OF ENERGY

BES

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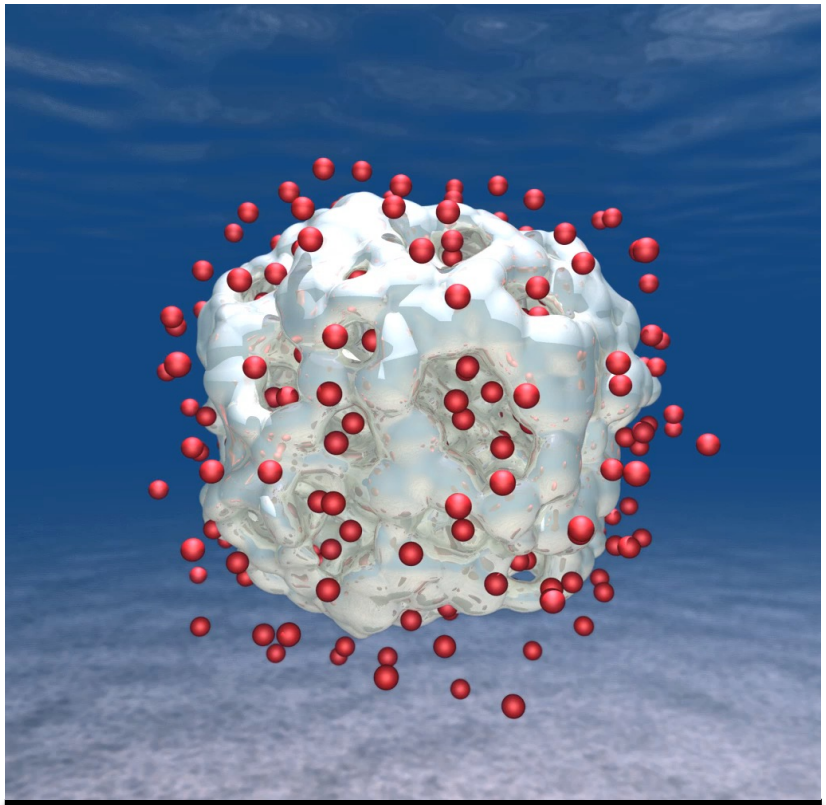

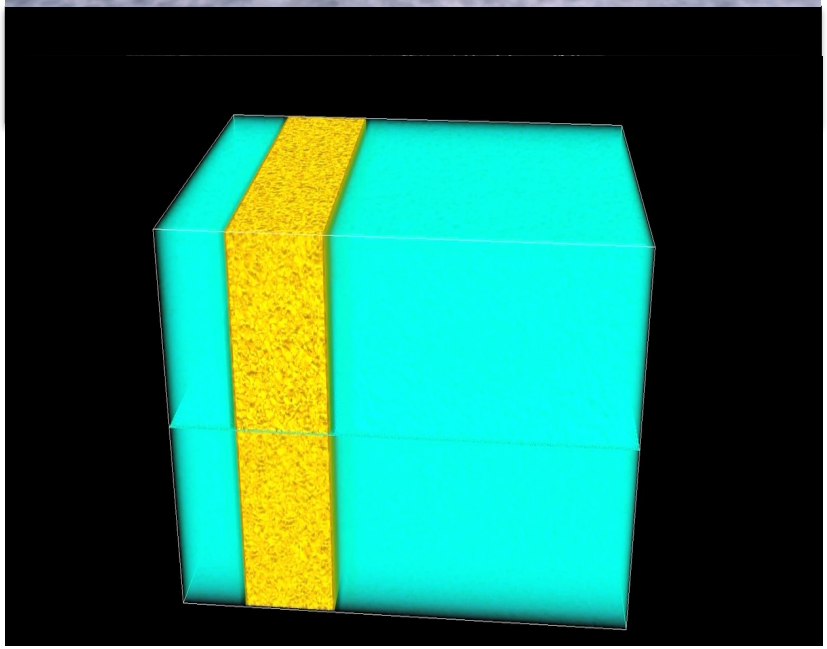

NOVEMBER 3–5, 2015

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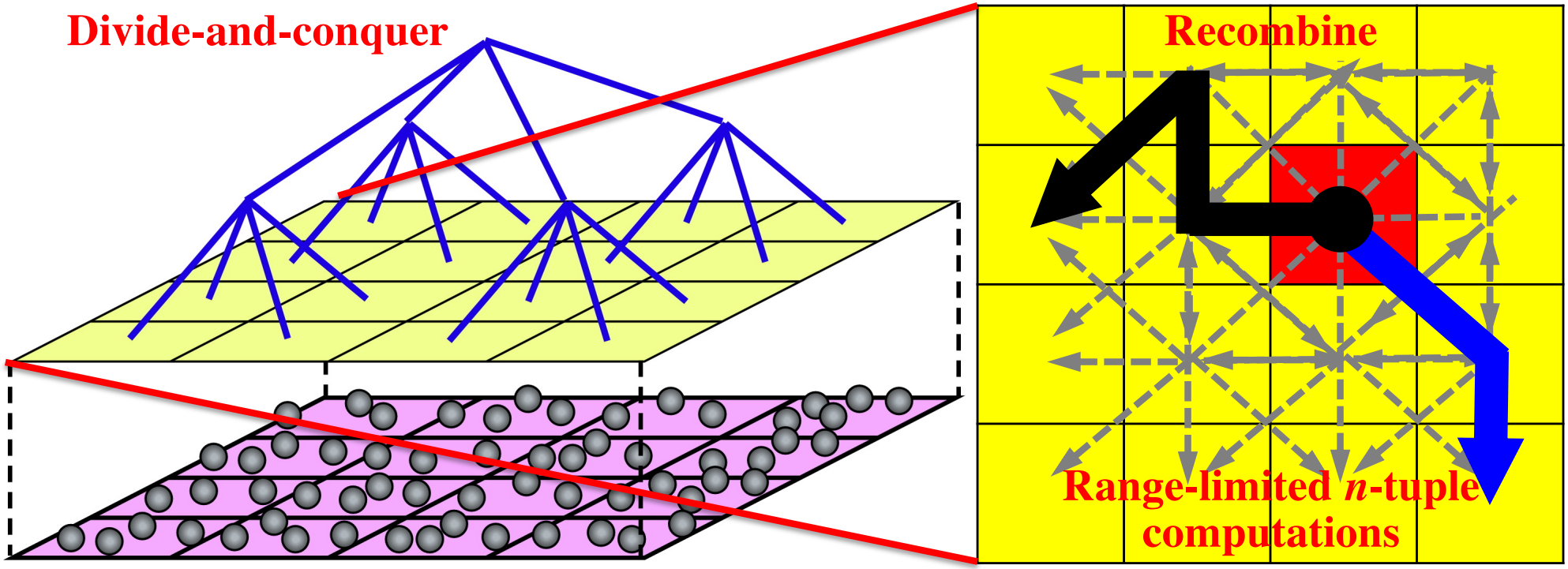
U.S. DEPARTMENT OF ENERGY

Divide-Conquer-Recombine (DCR) Engines



M. Kunaseth *et al.*, *ACM/IEEE SC13*

- **Variable *N*-charge problem: $O(N^3) \rightarrow O(N)$: Extended-Lagrangian reactive molecular dynamics algorithm eliminates speed-limiting charge iteration**

  Nomura *et al.*, *Comput. Phys. Commun.* **192**, 91 ('15); Liu *et al.*, *IEEE/ACM ScalA18*

  cf. Niklasson, *Phys. Rev. Lett.* **100**, 123004 ('08)

- **Quantum *N*-body problem: $O(C^N) \rightarrow O(N)$: Lean divide-&-conquer density functional theory algorithm for QMD minimizes $O(N)$ prefactor**

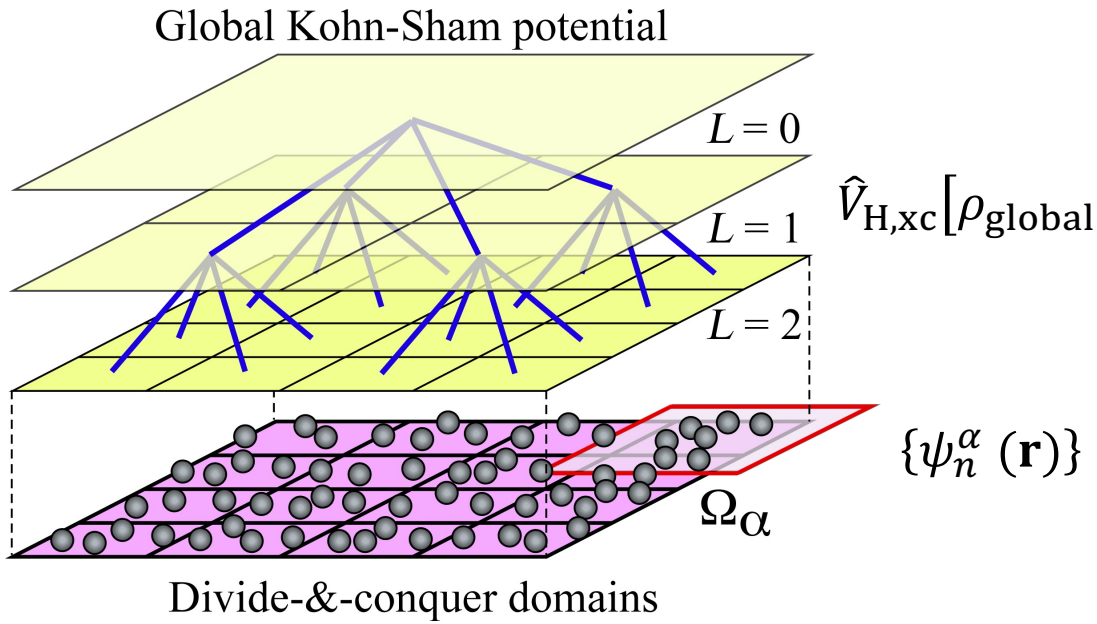
  Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14); Nomura *et al.*, *IEEE/ACM SC14*; Tiwari *et al.*, *ACM HPCAsia20*, best paper award

Divide-&-Conquer Density Functional Theory

Yang, *Phys. Rev. Lett.* **66**, 1438 ('91)

Shimojo *et al.*, *Comput. Phys. Commun.* **167**, 151 ('05)

Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14)



- **Overlapping spatial domains:** $\Omega = \cup_\alpha \Omega_\alpha$
- **Domain Kohn-Sham equations**

Global-local self-consistent field (SCF) iteration

$$\left(-\frac{1}{2}\nabla^2 + \hat{V}_{\mathrm{ion}} + \hat{V}_{\mathrm{H,xc}}[\rho_{\mathrm{global}}(\mathbf{r})]\right)\psi_n^\alpha(\mathbf{r}) = \epsilon_n^\alpha \psi_n^\alpha(\mathbf{r})$$

- **Global & domain electron densities**

$$\rho_{\mathrm{global}}(\mathbf{r}) = \Sigma_\alpha\, p_\alpha(\mathbf{r})\rho_\alpha(\mathbf{r}) \leftarrow \rho_\alpha(\mathbf{r}) = \Sigma_n |\psi_n^\alpha|^2\, \Theta(\mu - \epsilon_n^\alpha)$$

Domain support function

$$\Sigma_\alpha\, p_\alpha(\mathbf{r}) = 1$$

Global chemical potential

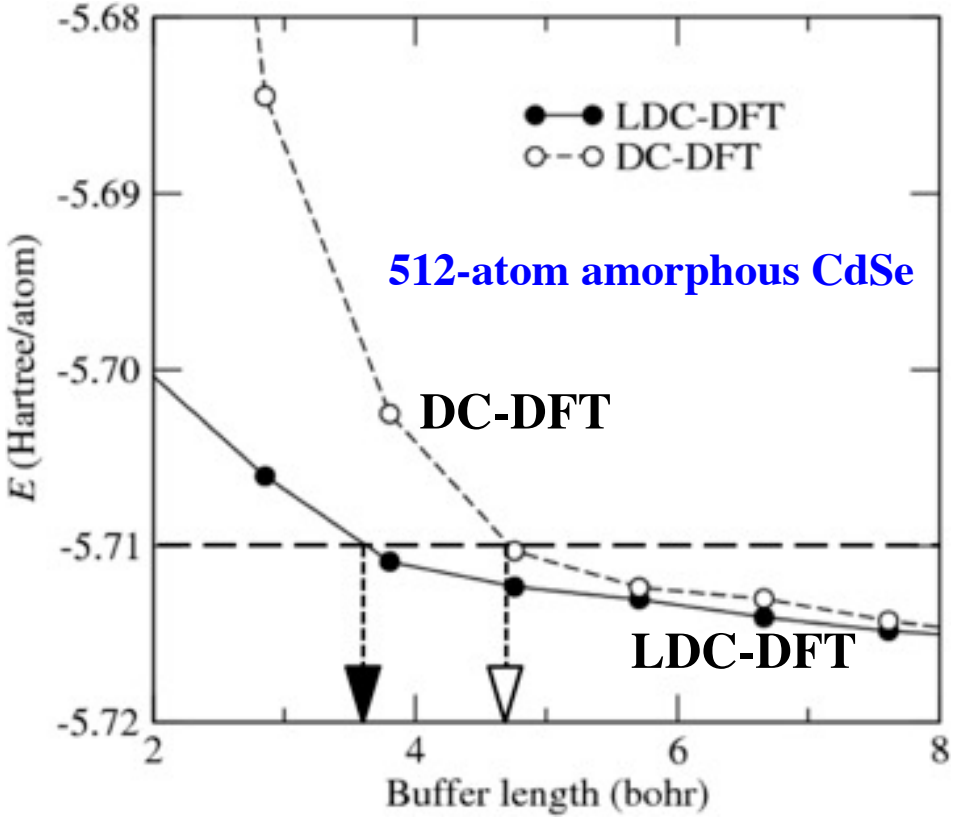
$$N = \int d\mathbf{r}\, \rho_{\mathrm{global}}(\mathbf{r})$$

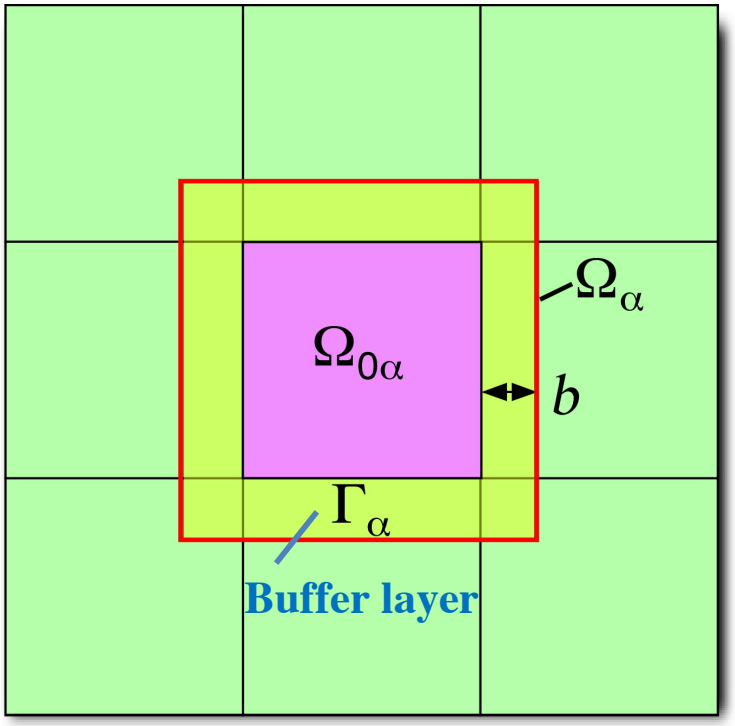
Lean Divide-&-Conquer (LDC) DFT

- **Density-adaptive boundary potential to reduce the *O*(*N*) prefactor**

$$v_\alpha^{\mathrm{bc}}(\mathbf{r}) = \int d\mathbf{r}' \frac{\partial v(\mathbf{r})}{\partial \rho(\mathbf{r}')}\Big(\rho_\alpha(\mathbf{r}) - \rho_{\mathrm{global}}(\mathbf{r})\Big) \cong \frac{\rho_\alpha(\mathbf{r}) - \rho_{\mathrm{global}}(\mathbf{r})}{\xi}$$

- **More rapid energy convergence of LDC-DFT compared with nonadaptive DC-DFT**



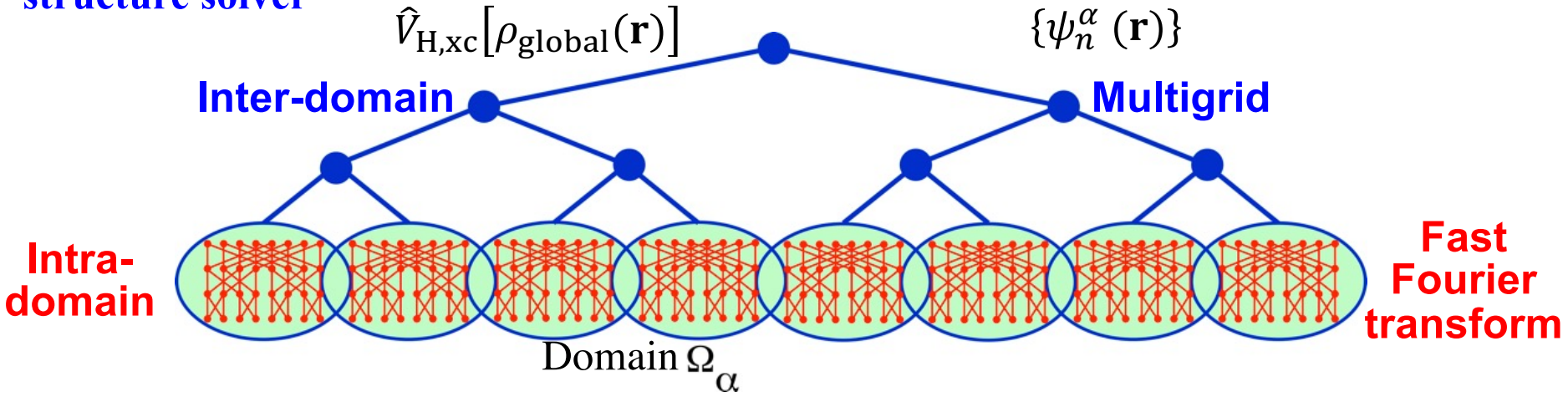


- **Factor 2.03 (for ν = 2) ~ 2.89 (for ν = 3) reduction of the computational cost with an error tolerance of 5×10^{-3} a.u. (per-domain complexity: n^ν)**

Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14)

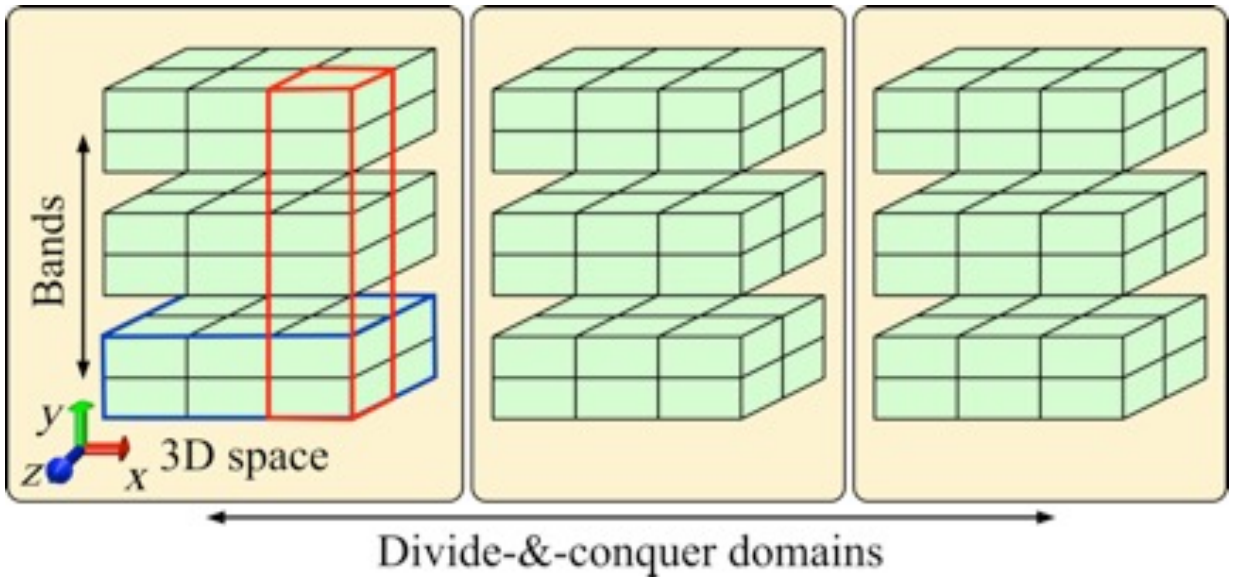
Hierarchical Computing: QMD

- **GSLF: Globally scalable (real-space multigrid) + locally fast (plane wave) electronic-structure solver**



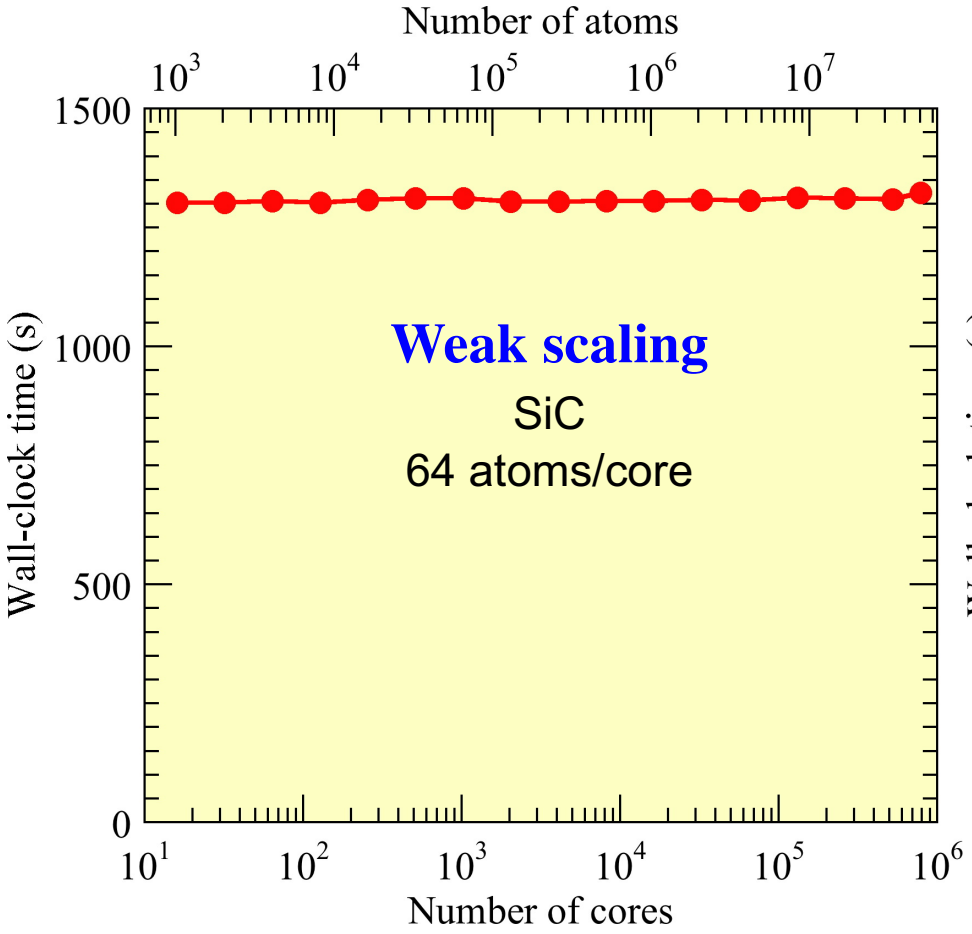
GSLF maximally exposes data locality → scalability

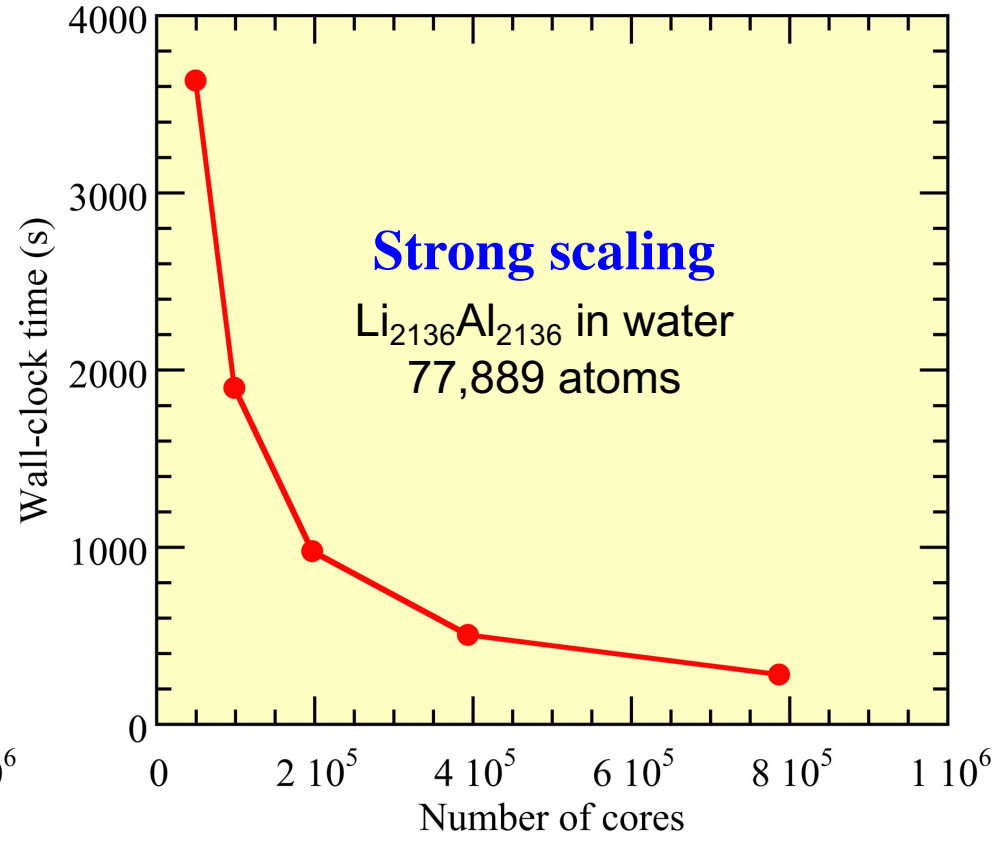
- **Hierarchical band (*i.e.*, Kohn-Sham orbital) + space + domain (BSD) decomposition**



Parallel Performance: QMD

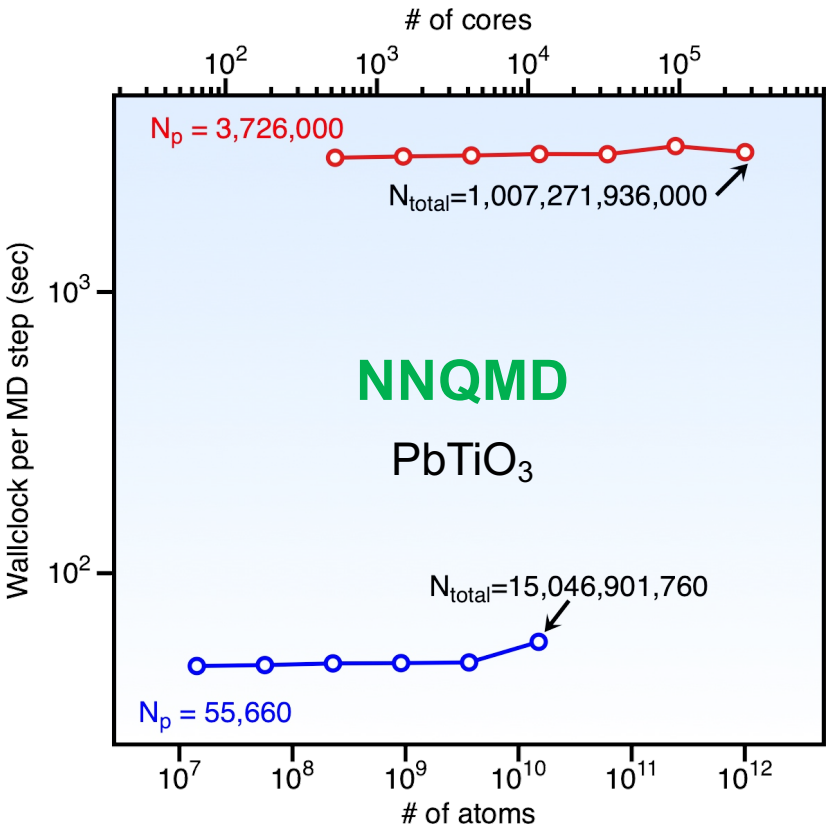
- **Weak-scaling parallel efficiency is 0.984 on 786,432 cores for a 50,331,648-atom SiC system**
- **Strong-scale parallel efficiency is 0.803 on 786,432 cores**





Nomura *et al.*, *IEEE/ACM Supercomputing, SC14* ('14)

Parallel Performance: NNQMD

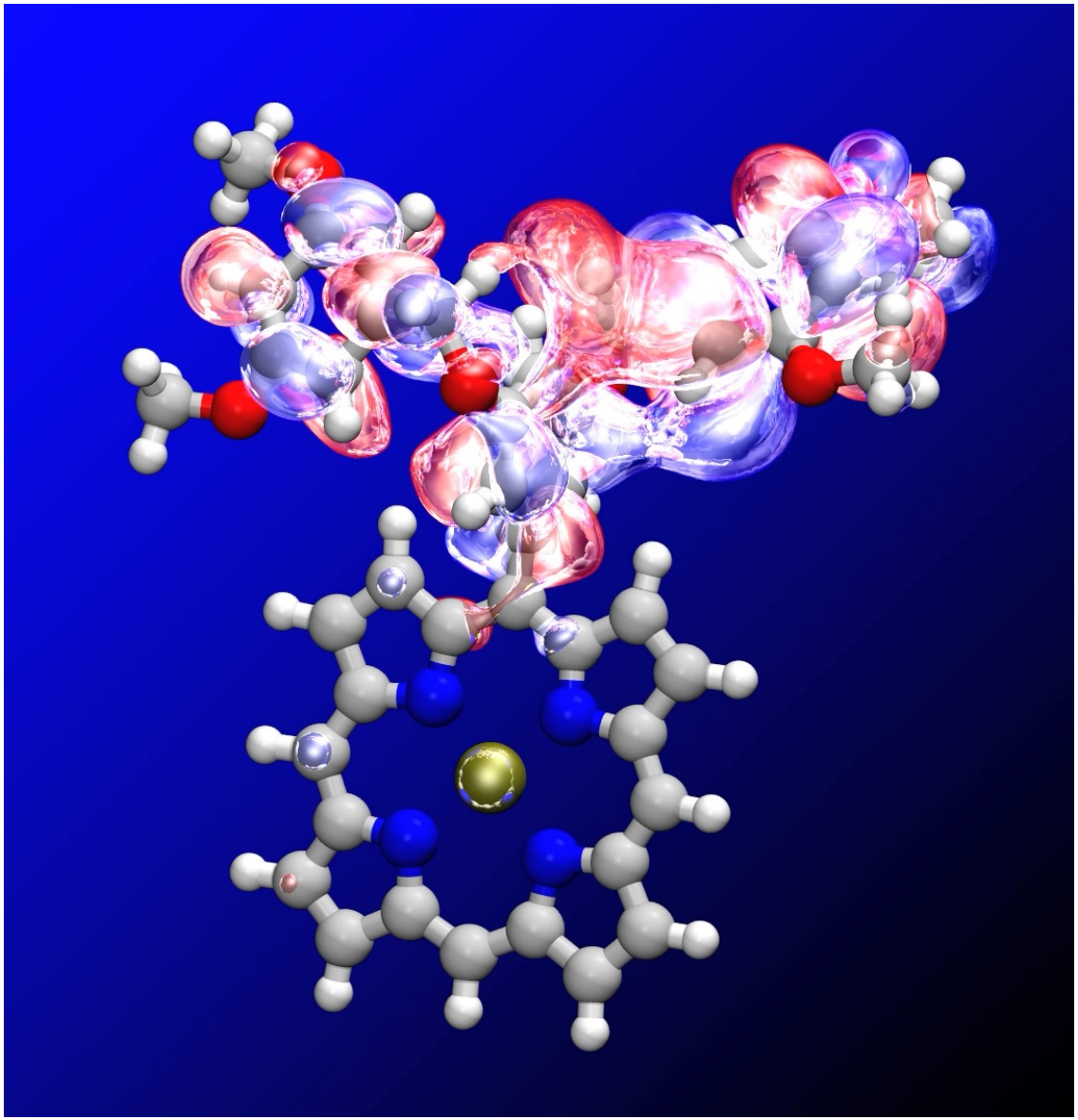


- **Trillion-atom scalability: Weak-scaling parallel efficiency is 0.984 on 4,224 compute nodes (270,336 cores) of Theta supercomputer at Argonne National Laboratory for a trillion-atom neural-network quantum molecular dynamics (NNQMD) of $PbTiO_3$**

- **GPU acceleration: 70× speedup on GPU (NVIDIA A100) over CPU (AMD EPYC 7513) of Polaris supercomputer at Argonne**

Number of atoms	1,000	10,000	100,000
Speedup (A100 GPU over EPYC CPU)	4.37	24.7	68.9

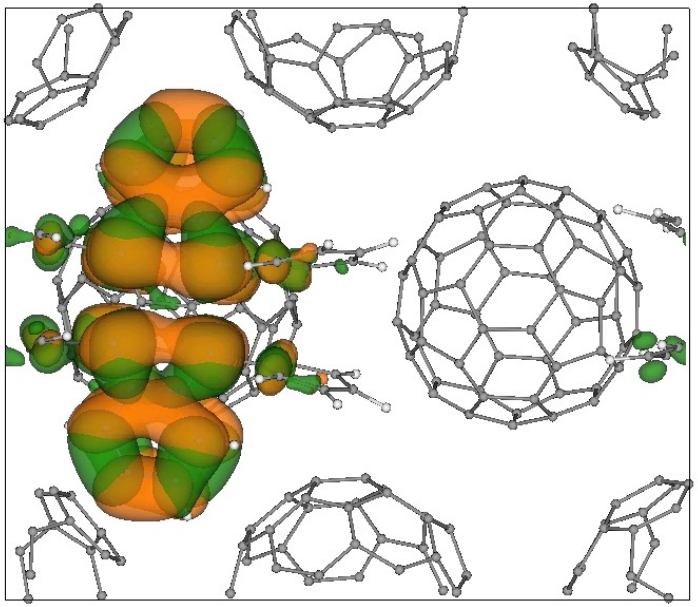
Nonadiabatic Quantum Molecular Dynamics



Appl. Phys. Lett. **98**, 113301 ('11); *ibid*. **100**, 203306 ('12); *J. Chem. Phys*. **136**, 184705 ('12); *Comput. Phys. Commun*. **184**, 1 ('13); *Appl. Phys. Lett*. **102**, 093302 ('13); *ibid*. **102**, 173301 ('13); *J. Chem*. *Phys*. **140**, 18A529 ('14); *IEEE Computer* **48(11)**, 33 ('15); *Sci. Rep*. **5**, 19599 ('16); *Nature Commun*. **8**, 1745 ('17); *Nano Lett*. **18**, 4653 ('18); *Nature Photon.* **13**, 425 ('19)

Zn porphyrin

Rubrene/C_{60}

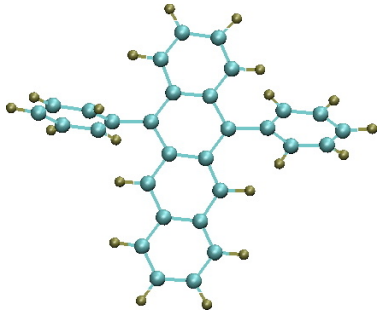


quasi-electron; quasi-hole

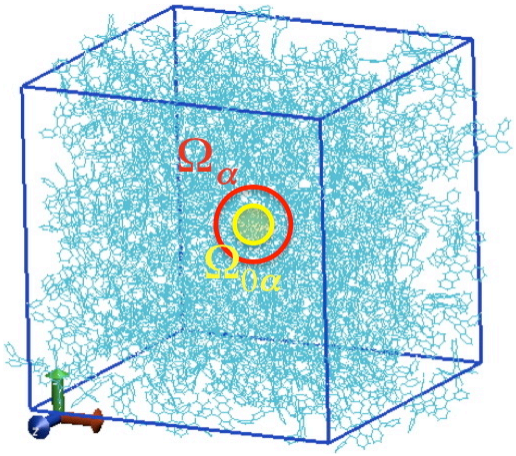
- **Excited states: Linear-response time-dependent density functional theory** [Casida, '95]
- **Interstate transitions: Surface hopping** [Tully, '90; Jaeger, Fisher & Prezhdo, '12]

Singlet Fission in Amorphous DPT

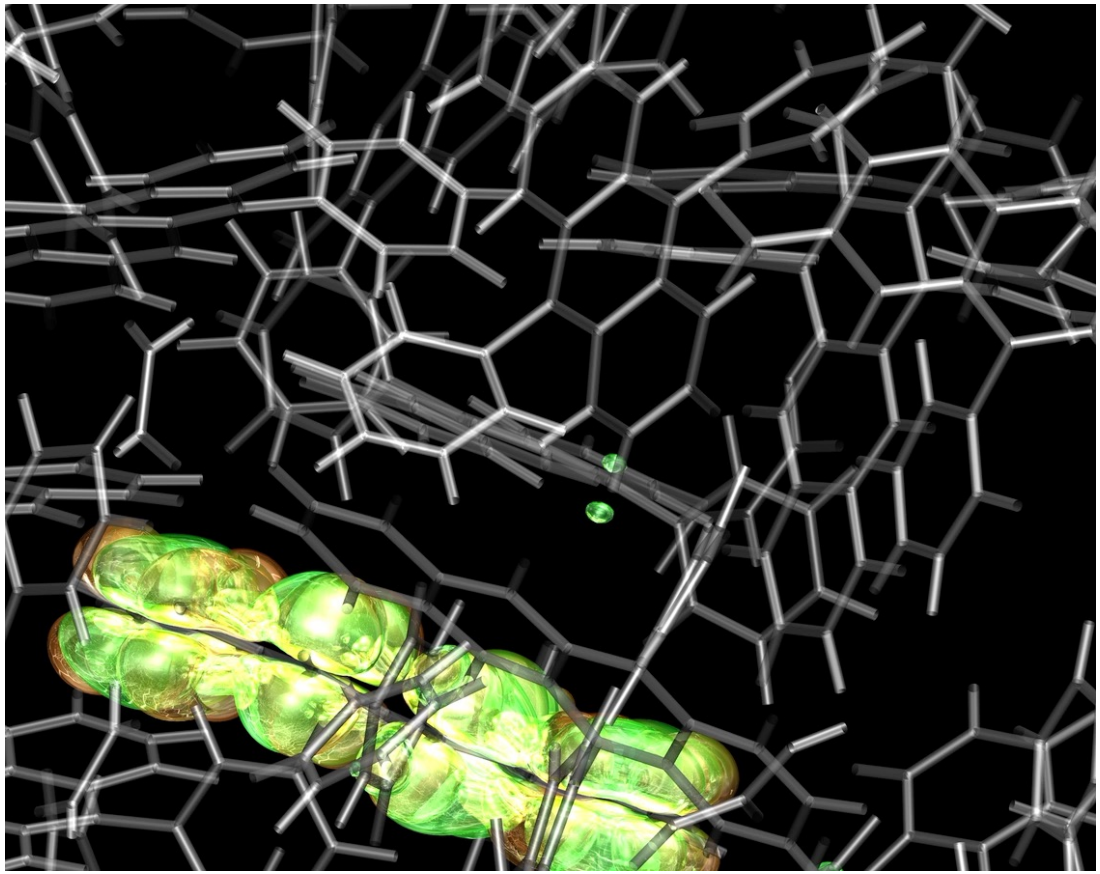
- **Photo-current doubling by splitting a singlet exciton into 2 triplet excitons**
- **Singlet fission in mass-produced disordered organic solid → efficient low-cost solar cells**
- **Experimental breakthrough: SF found in amorphous diphenyl tetracene (DPT)**



DPT molecule



Amorphous DPT



Quasi-electron

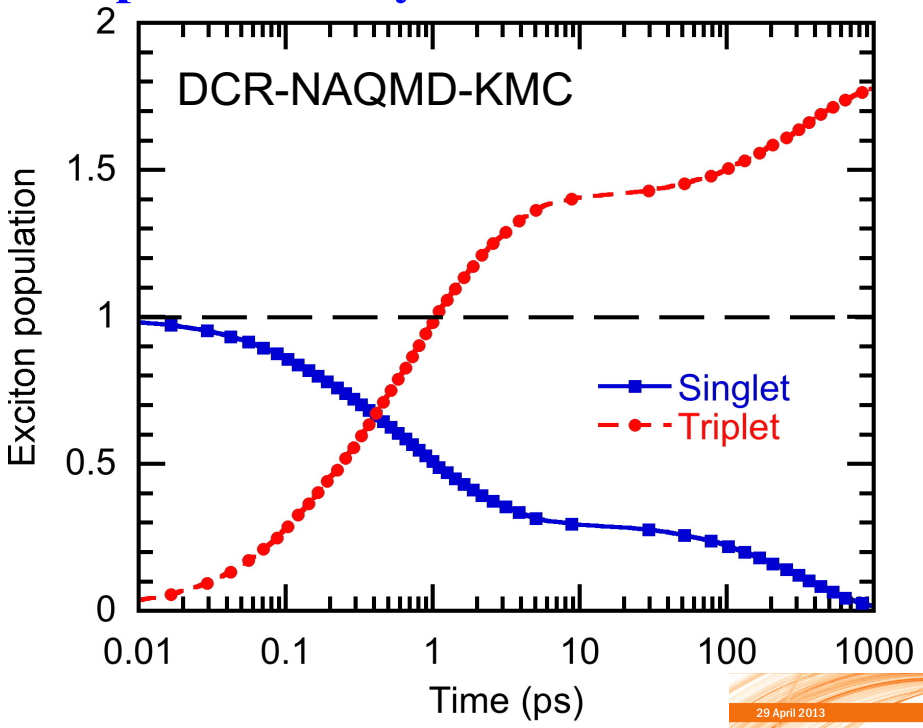
Quasi-hole

W. Mou *et al.*, *Appl. Phys. Lett.* **102**, 173301 ('13)

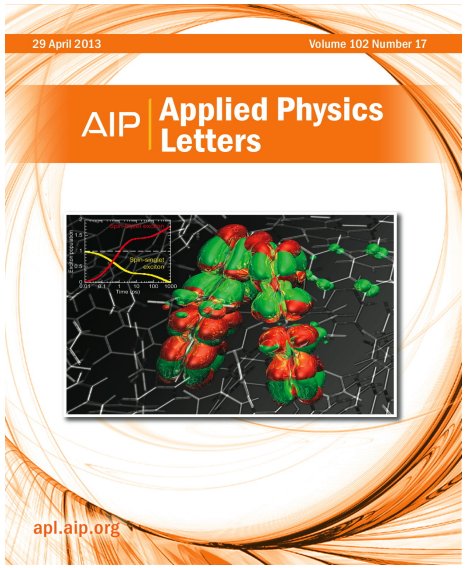
- **Divide-conquer-recombine nonadiabatic QMD (phonon-assisted exciton dynamics) + time-dependent perturbation theory (singlet-fission rate) + kinetic Monte Carlo calculations of exciton population dynamics in 6,400-atom amorphous DPT**

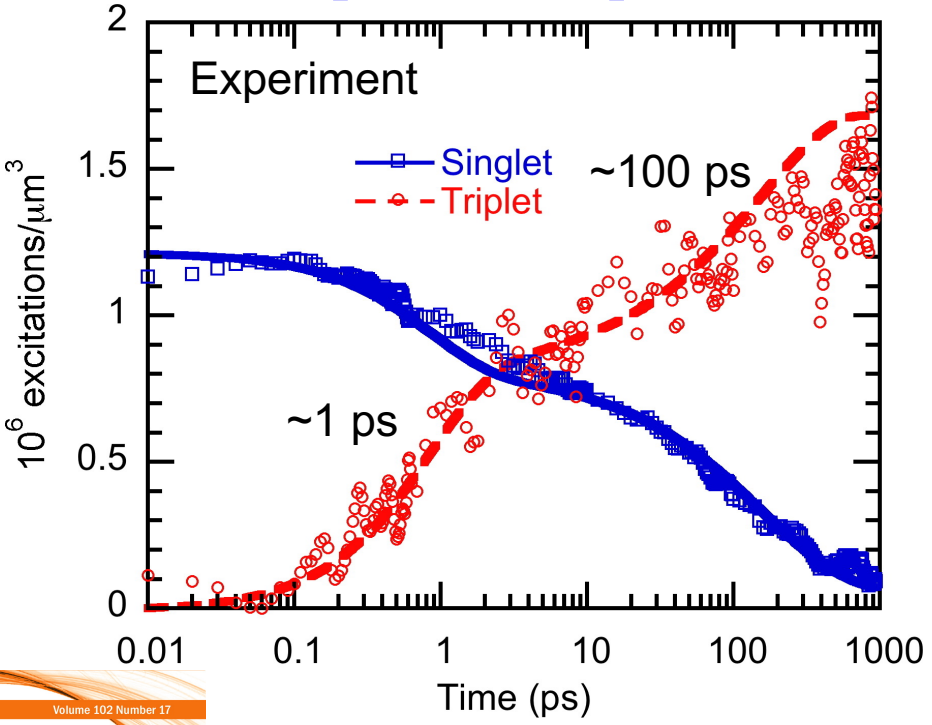
DCR-NAQMD Informed Kinetic Monte Carlo

- **DCR-NAQMD-KMC exciton population dynamics reproduces the experimentally observed two time-scales (~ 1 & 100 ps) in amorphous DPT**



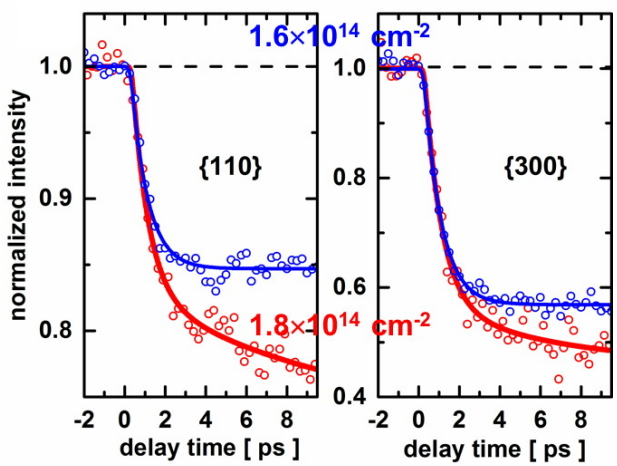
Mou *et al.*,
Appl. Phys. Lett.
100, 173301 ('13)

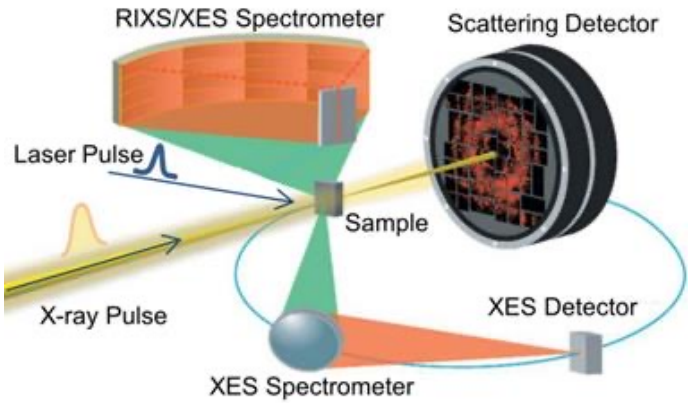


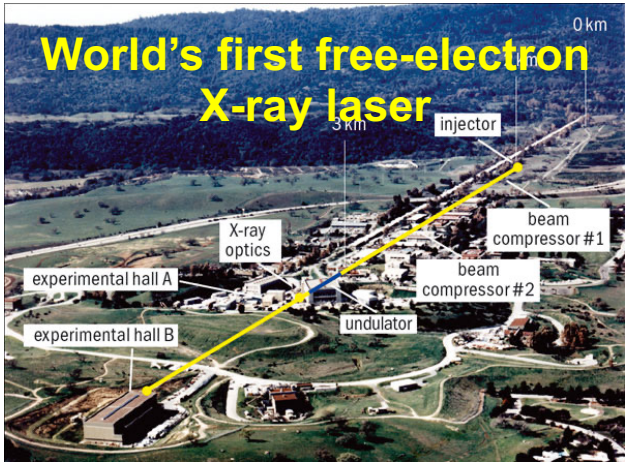


Roberts *et al.*,
J. Am. Chem. Soc.
134, 6388 ('12)

INCITE|AURORA–MAGICS–LCLS Synergy





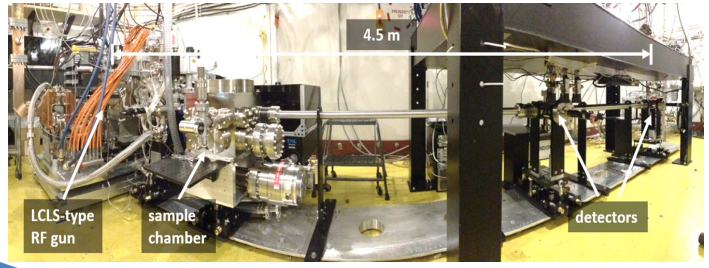


Linac Coherent Light Source

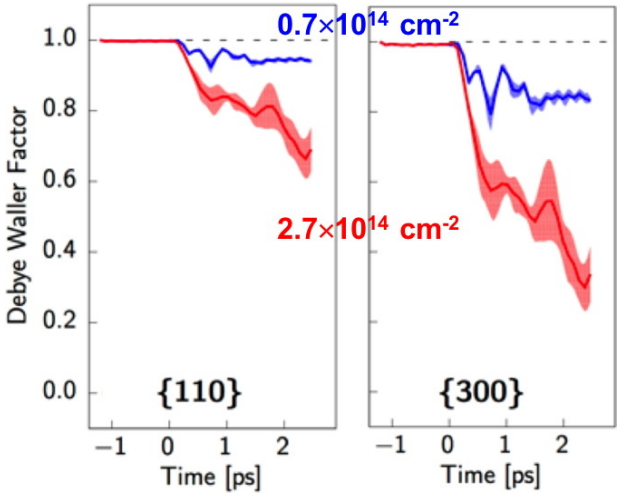
LCLS

DOE INCITE & Aurora ESP Awards

MAGICS
Materials Genome Innovation for Computational Software

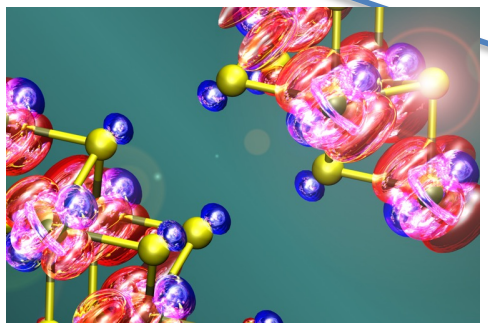


Ultrafast electron diffraction (UED) at SLAC



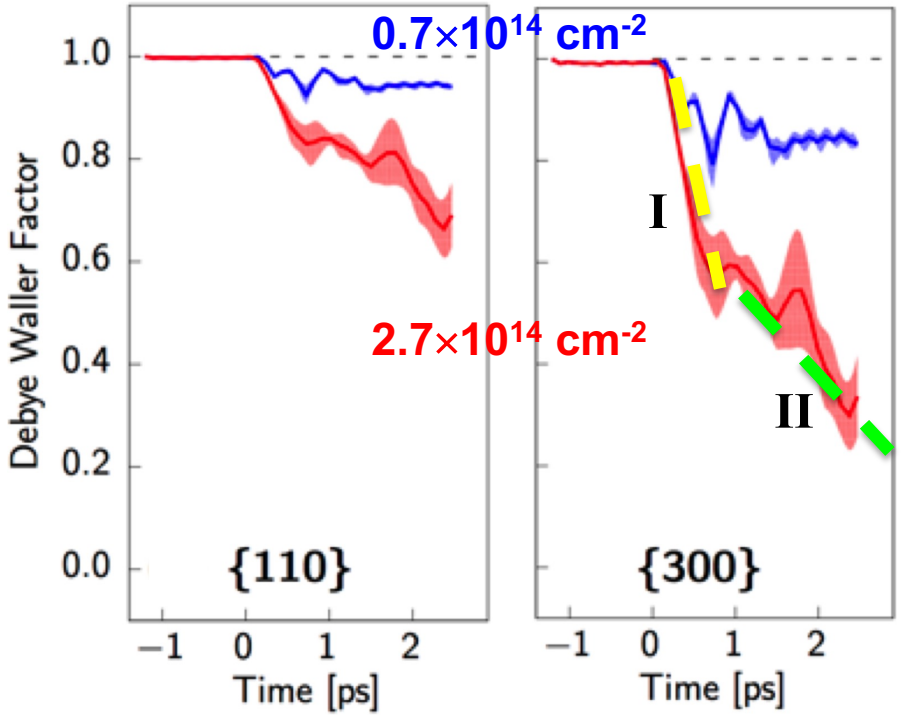
U.S. DEPARTMENT OF ENERGY
INCITE
LEADERSHIP COMPUTING

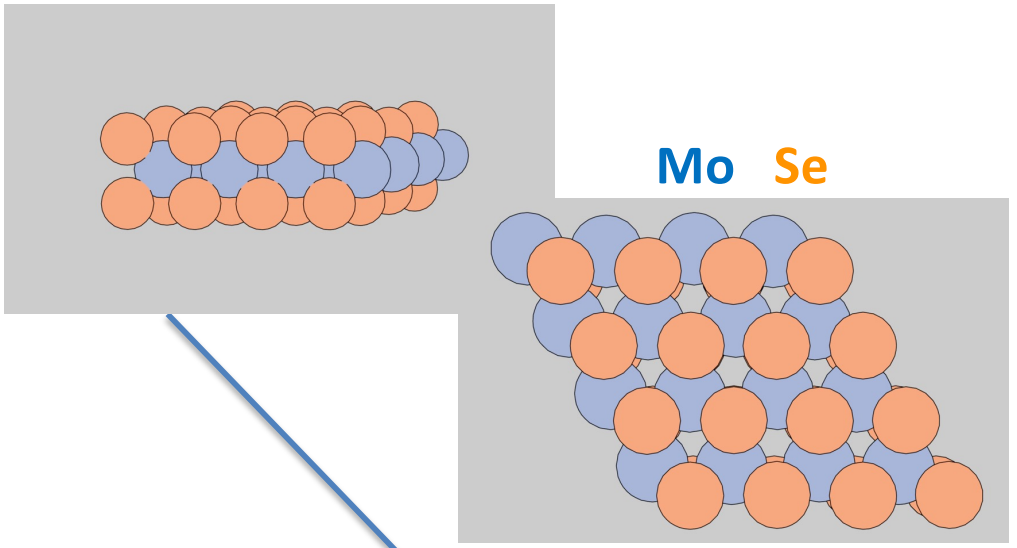
AURORA | EARLY SCIENCE PROGRAM

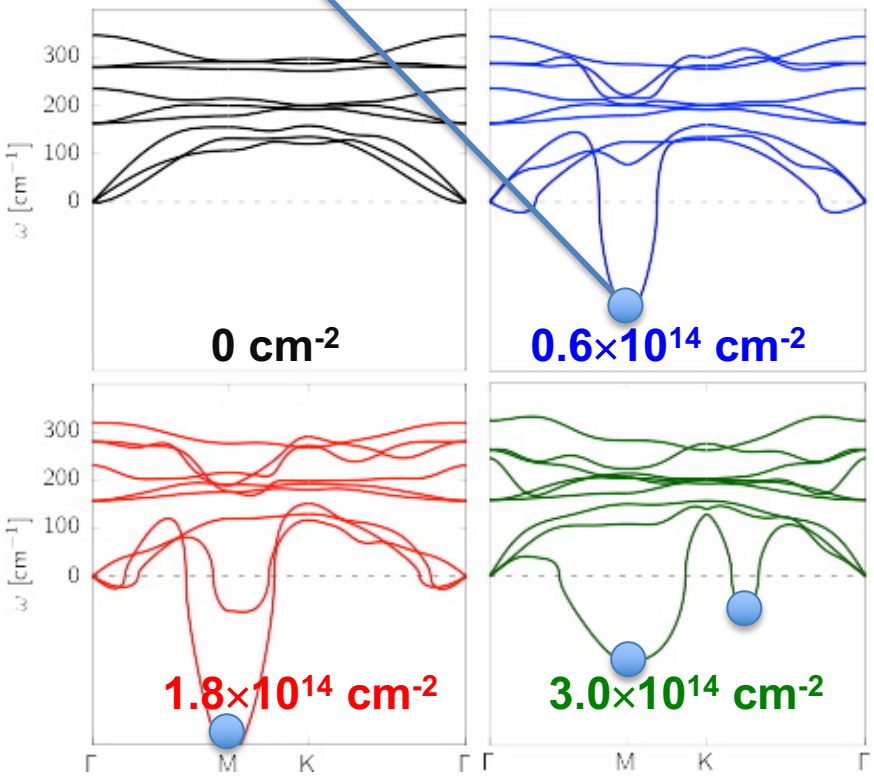


Strong Electron-Lattice Coupling in $MoSe_2$

- **NAQMD simulations reproduce (1) rapid photo-induced lattice dynamics & (2) mono- to bi-exponential transition at higher electron-hole density**







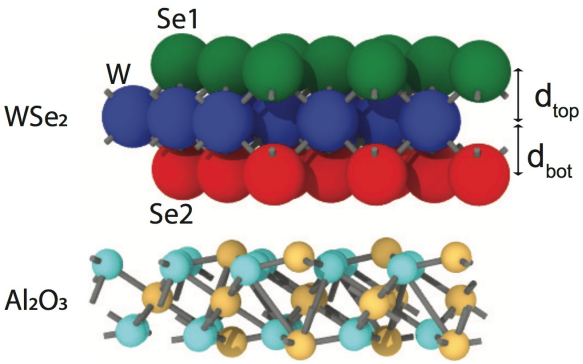
- **Rapid lattice dynamics is explained by the softening of M-point (1/2 0 0) phonon**
- **Bi-exponential transition is explained by the softening of additional phonon modes at higher electron-hole densities**

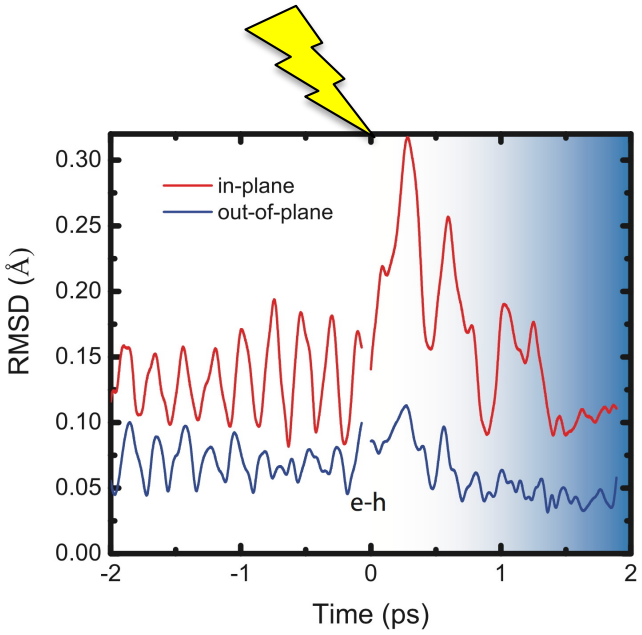
Lin *et al.*, *Nature Commun.* **8**, 1745 ('17)

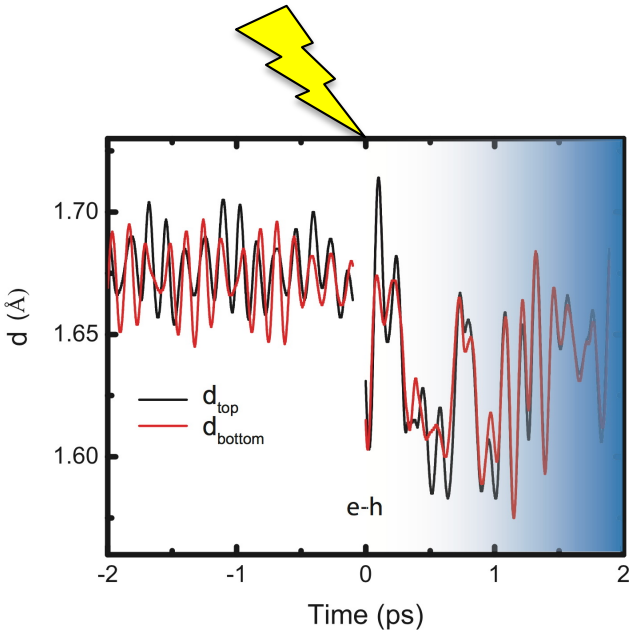
Bassman *et al.*, *Nano Lett.* **18**, 4653 ('18)

WSe_2 Monolayer on Al_2O_3 Substrate

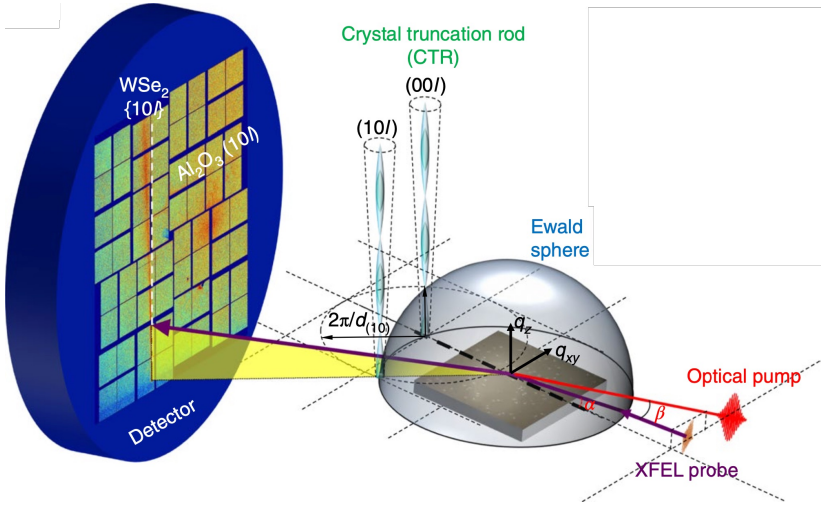
- **NAQMD simulation to study photoexcitation dynamics of WSe_2 monolayer on Al_2O_3 substrate**







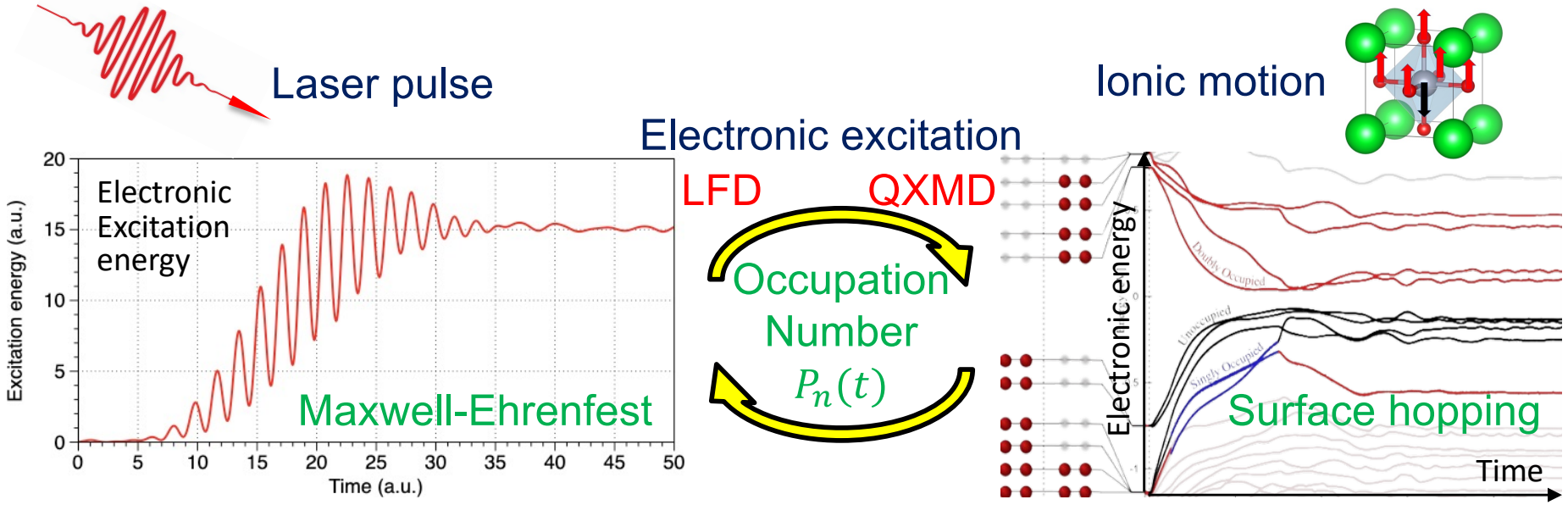
- **Enhanced in-plane atomic displacements upon photoexcitation**
- **Photo-induced intralayer contraction of W-Se distances**
- **Good agreement with femtosecond surface X-ray scattering experiments at LCLS**



Tung, *et al.*, *Nature Photonics* **13**, 425 ('19)

Light-Matter Interaction: DC-MESH

- **DC-MESH (divide-&-conquer Maxwell + Ehrenfest + surface-hopping): *O*(*N*) algorithm to simulate photo-induced quantum materials dynamics**
- **LFD (local field dynamics) solves Maxwell equations for light & real-time time-dependent density functional theory (RT-TDDFT) equations for electrons to describe light-matter interaction**
- **QXMD (quantum molecular dynamics with excitation) describes nonadiabatic coupling of excited electrons & ionic motions based on surface-hopping approach** [*Nature Commun*. **8**, 1745 ('17); *Nature Photon.*, **13**, 425 ('19)]
- **LFD-QXMD handshaking *via* electronic occupation numbers**
- **GSLD: Globally sparse (interdomain Hartree coupling *via* multigrid) & locally dense (intradomain nonlocal exchange-correlation computation *via* BLAS) solver**



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

LFD Algorithm

- **Hamiltonian in the α-th domain** [Yabana, *Phys. Rev. B* **85**, 045134 ('12)]

$$\hat{h}\big(t,\mathbf{R}(t)\big) = \overbrace{\frac{1}{2}\left(\frac{\nabla}{i}+\frac{1}{c}\mathbf{A}(\mathbf{r}_\alpha,t)\right)^2 - \phi(\mathbf{r}_\alpha,t) + \hat{v}_{\rm xc} + v_{\rm ion}(\mathbf{r},\mathbf{R})}^{\hat{h}_{\rm el}(t)} + \overbrace{\Delta\dot{\mathbf{R}}\bullet\frac{\partial}{\partial\mathbf{R}}v_{\rm ion}}^{\hat{h}_{\rm el-ion}}$$

Electromagnetic vector & scalar potentials at the α-th domain — Nonadiabatic coupling

- **Trotter expansion of time propagator**

$$\exp\big(-i\hat{h}\Delta_{\rm MD}\big) \cong \exp\big(-i\hat{h}_{\rm el-ion}\Delta_{\rm MD}/2\big)\mathcal{T}\exp\left(-i\int_t^{t+\Delta_{\rm MD}} dt\hat{h}_{\rm el}(t)dt\right)\exp\big(-i\hat{h}_{\rm el-ion}\Delta_{\rm MD}/2\big)$$

QXMD — LFD — QXMD

- **Self-consistent propagator** [Sato, *J. Chem. Phys.* **143**, 224116 ('15); Lian, *Adv. Theo. Sim.* **1**, 1800055 ('18)]

$$\mathcal{T}\exp\left(-i\int_t^{t+\Delta_{\rm MD}} dt\hat{h}_{\rm el}(t)\right) \cong \prod_{n=1}^{N_{\rm QD}=\Delta_{\rm MD}/\Delta_{\rm QD}} \exp\left(-i\Delta_{\rm QD}\hat{h}_{\rm el}\left(t+\left(n-\frac{1}{2}\right)\Delta_{\rm QD}\right)\right)$$

- **Nonlocal exchange-correlation propagator** [Vlcek, *J. Chem. Phys.* **150**, 184118 ('19)]

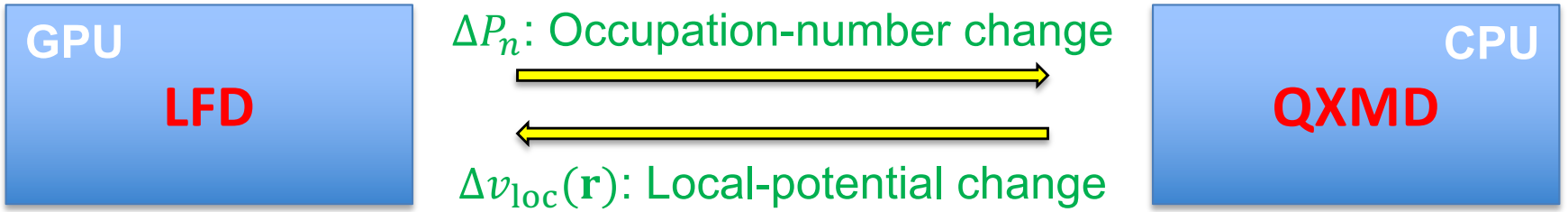
$$\exp\big(-i\Delta_{\rm QD}\hat{h}_{\rm el}\big) \cong \frac{1-i\hat{v}_{\rm nl}\Delta_{\rm QD}/2}{\big\|\big(1-i\hat{v}_{\rm nl}\Delta_{\rm QD}/2\big)|\psi_n(t)\rangle\big\|}\exp\big(-i\Delta_{\rm QD}\hat{h}_{\rm loc}\big)\frac{1-i\hat{v}_{\rm nl}\Delta_{\rm QD}/2}{\big\|\big(1-i\hat{v}_{\rm nl}\Delta_{\rm QD}/2\big)|\psi_n(t)\rangle\big\|}$$

$\hat{v}_{\rm xc} = \hat{v}_{\rm loc} + \hat{v}_{\rm nl}$; $\hat{h}_{\rm el} = \hat{h}_{\rm loc} + \hat{v}_{\rm nl}$

local — nonlocal

Reduced-Communication Shadow Dynamics

- **At each molecular-dynamics step, LFD informs QXMD of occupation-number change due to light-electron & electron-electron interactions**
- **QXMD performs excited-state quantum molecular dynamics & informs LFD of local-potential change for the next N_{QD} $(= \Delta_{\mathrm{MD}}/\Delta_{\mathrm{QD}})$ quantum-dynamics steps**
- **"Shadow" electronic wave functions in LFD are resident on GPU, while QXMD wave functions on CPU, to minimize CPU-GPU data transfers**



Shadow dynamics: real-time time-dependent density functional theory (RT-TDDFT)

Pseudopotentials: projector augmented wave (PAW), ultrasoft (Vanderbilt)

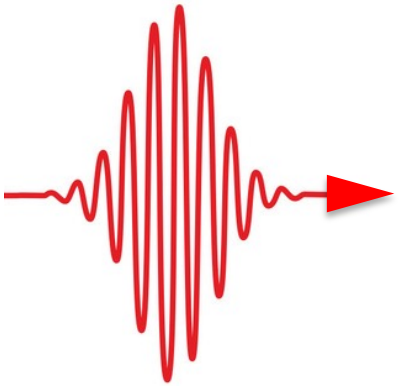
Exchange-correlation functionals: metaGGA (SCAN), hybrid exact-exchange (HSE *etc.*), DFT+U, DFT-D, nonlocal vdW

- **BLASified nonlocal propagator in LFD on GPU**

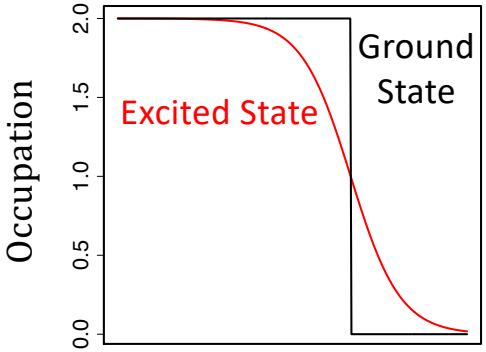
  Real-time scissor approximation [Wang, *J. Phys. Condens. Mat.* **31**, 214002 ('19)]

$$\hat{v}_{\mathrm{nl}}|\psi_n(t)\rangle \cong \Delta_{\mathrm{sci}} \sum_{m \geq \mathrm{LUMO}} |\psi_m\rangle\langle\psi_m|\psi_n(t)\rangle$$

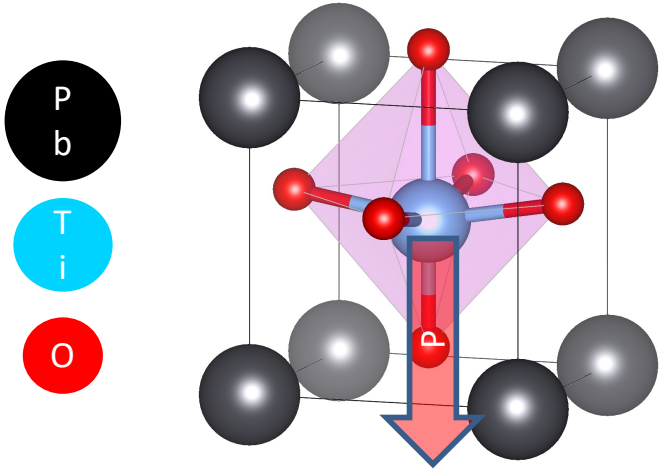
Multiscaling from DC-MESH to XS-NNQMD



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 → **Excited-state (XS) NNQMD**

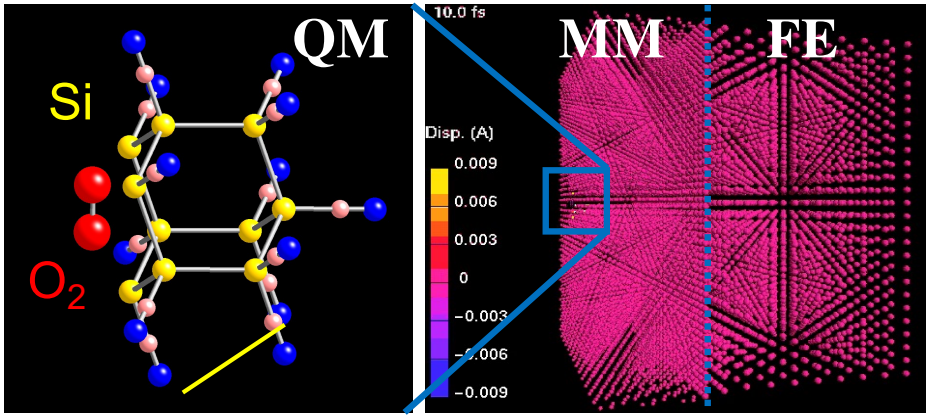
Excited energy landscape

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)

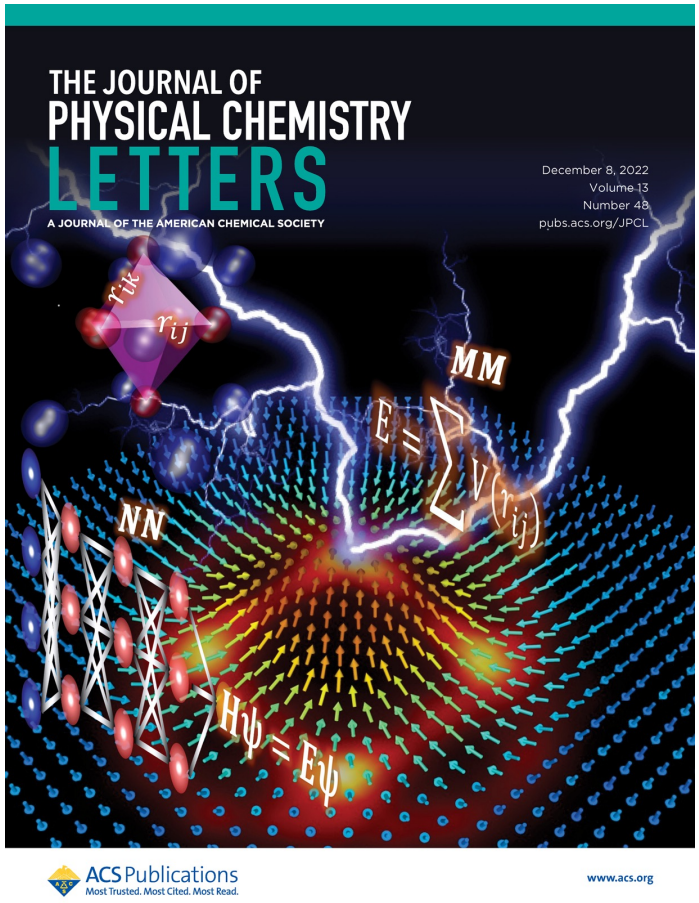
  Warshel, *Angew. Chem.* **53**, 10020 ('14)



QM/MM/FE (finite-element method)

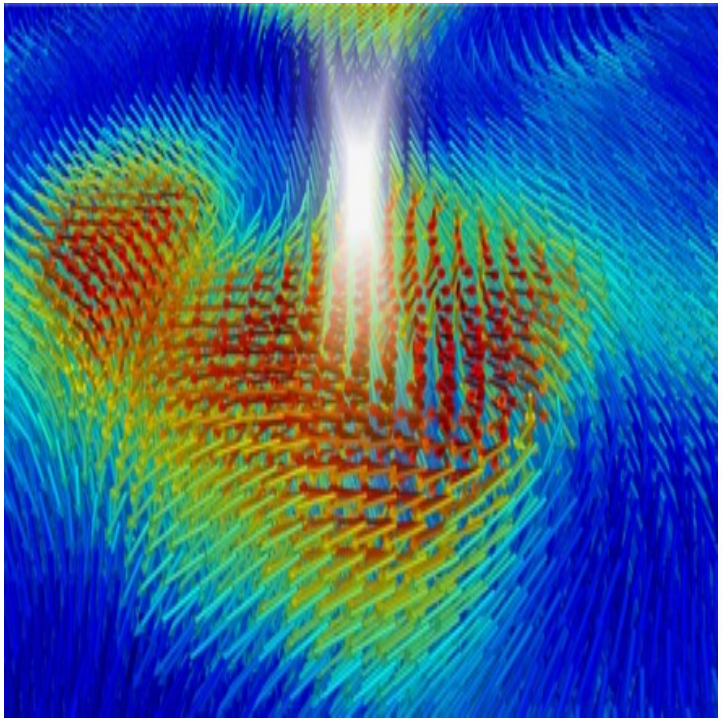
Ogata *et al*, *Comput. Phys. Commun.* **138**, 143 ('01)

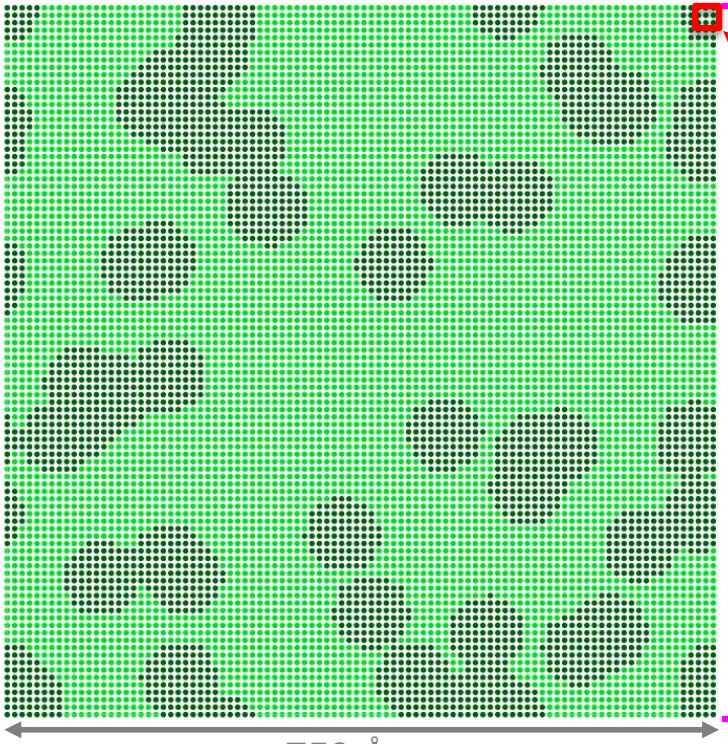
- **NN/MM:** NNQMD for ferroelectric ($PbTiO_3$: PTO) embedded in MM for paraelectric ($SrTiO_3$: STO) to apply appropriate strain boundary condition



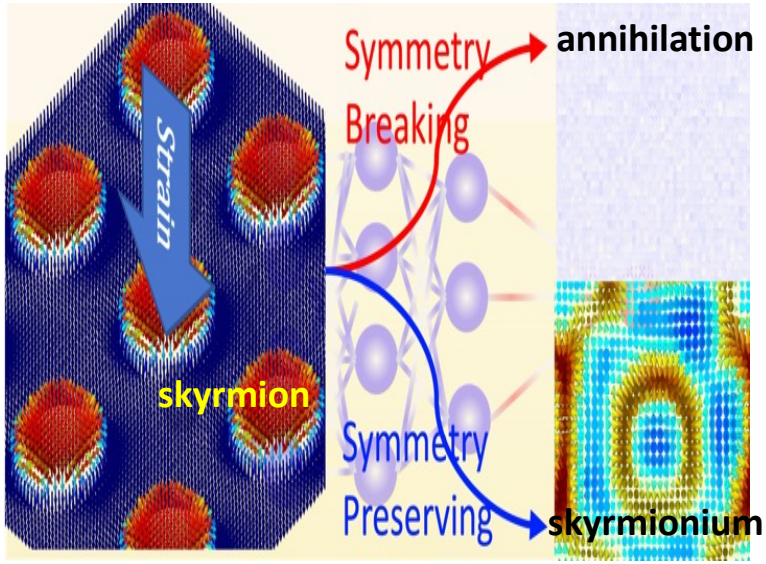
Linker *et al.*, *J. Phys. Chem. Lett.* **13**, 11335 ('22)

Application: Ferroelectric Opto-Topotronics





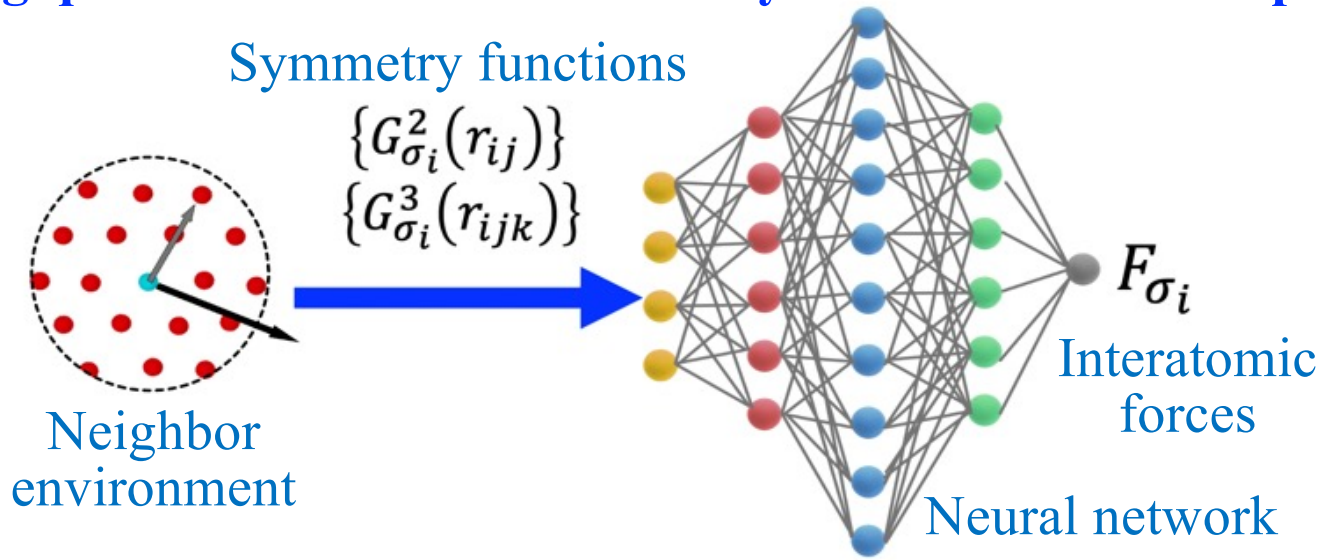
- **Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications**
- **Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)**
- **Symmetry-controlled skyrmion-to-skyrmionium*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



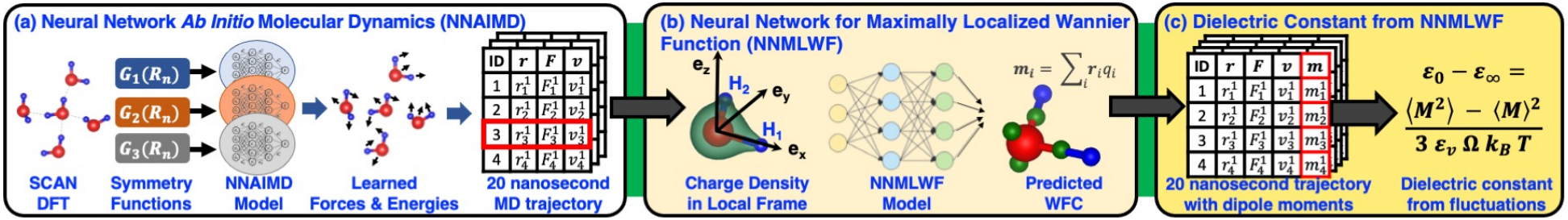
Linker *et al., Science Adv.* **8**, eabk2625 ('22); *JPCL* **13**, 11335 ('22)

Neural-Network Quantum Molecular Dynamics

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



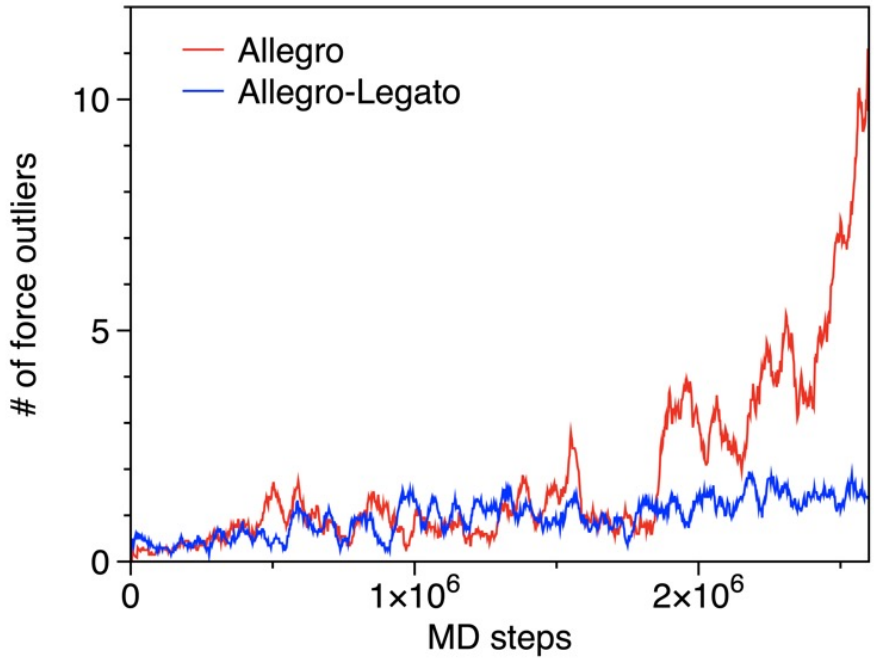
- **Neural networks predict: (1) atomic forces for performing MD simulations; & (2) maximally-localized Wannier-function (MLWF) centers for computing quantum properties like electronic dipoles**

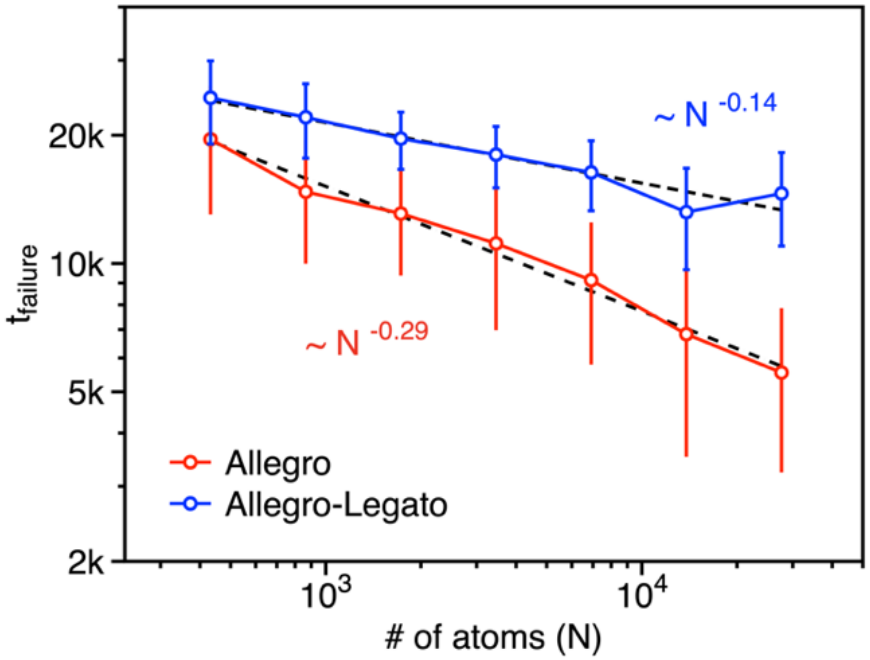


Krishnamoorthy *et al.*, *Phys. Rev. Lett.* **126**, 216403 ('21)

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**

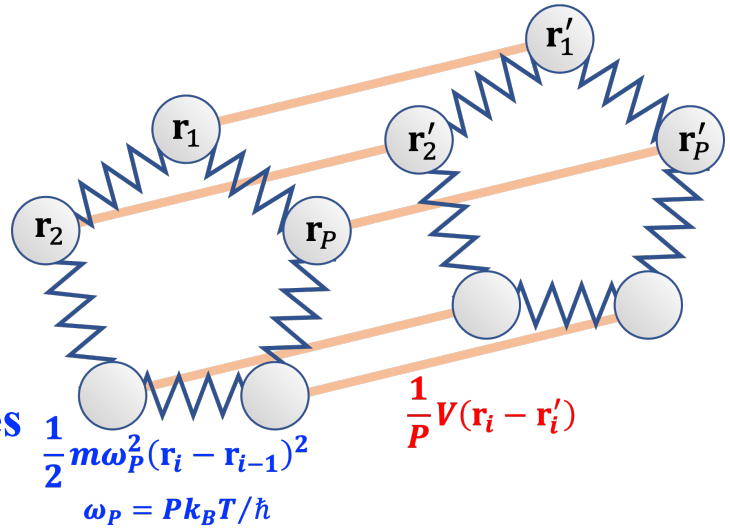


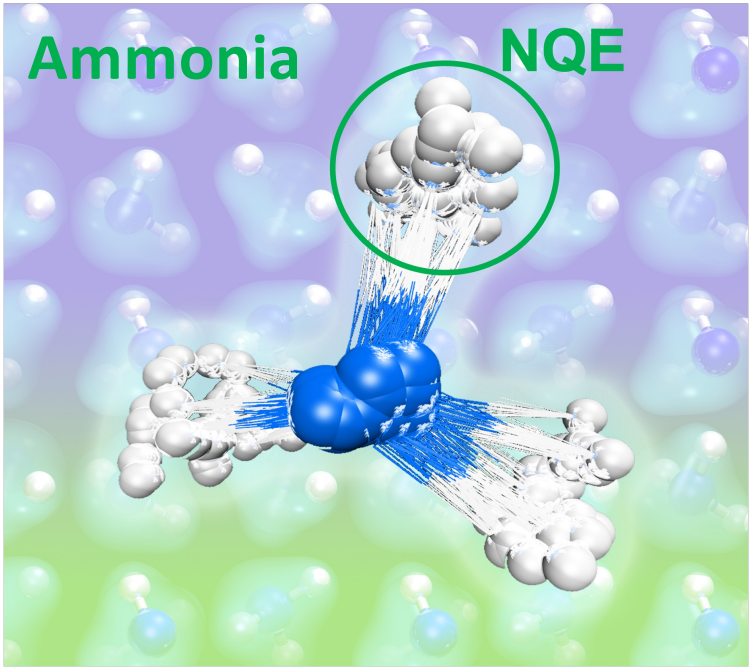


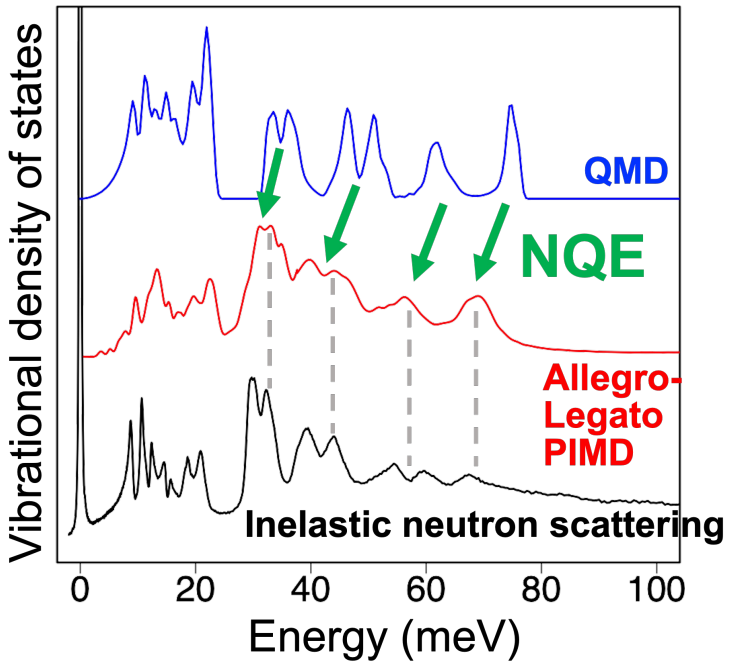
H. Ibayashi *et al.*, *ISC 2023* (*arXiv*: 2303.08169)

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**



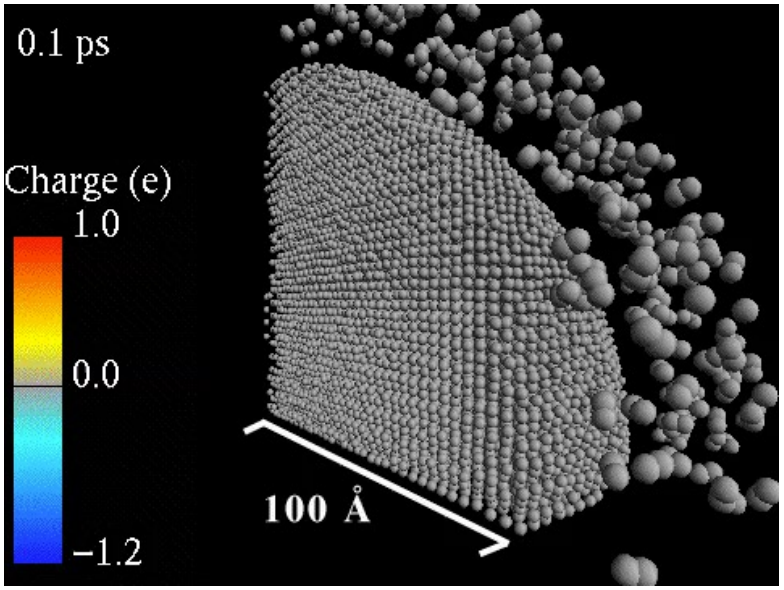




H. Ibayashi *et al.*, *ISC 2023* (*arXiv*: 2303.08169); Linker *et al.*, under submission

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)

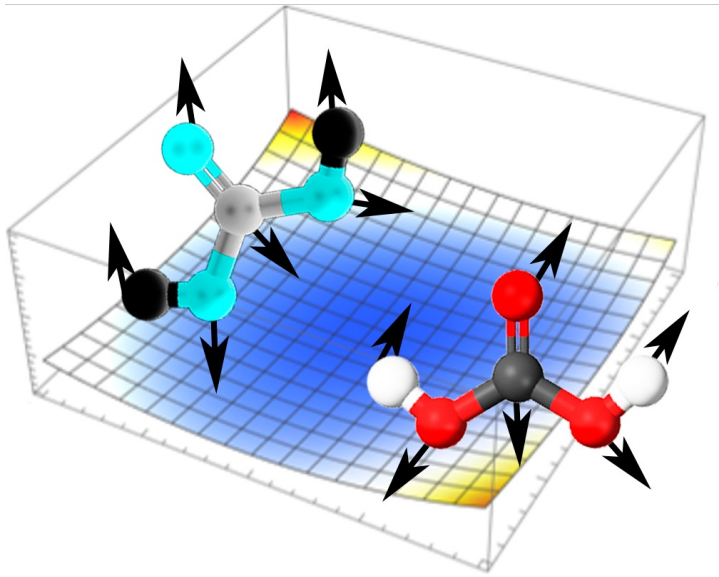
$$q_*^N = \underset{q^N}{\operatorname{argmin}} E_{\text{Coulomb}}(\mathbf{r}^N, q^N)\ s.t. \Sigma_i q_i = 0$$

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

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**



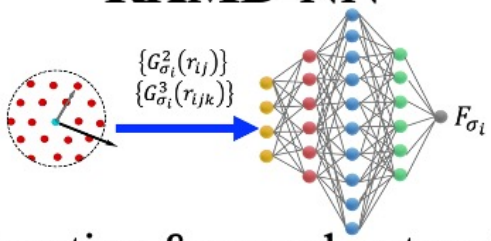
AIQ-XMaS Software Suite

AI & Quantum-Computing Enabled Exa Quantum Materials Simulator

QXMD

Nonadiabatic quantum molecular dynamics

RXMD-NN



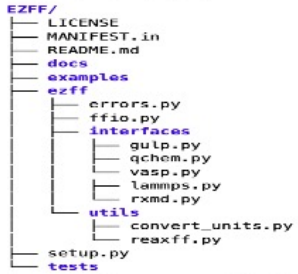
Reactive & neural-network molecular dynamics

Aurora ESP engine

GEARS

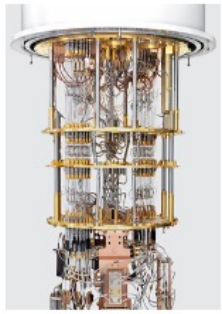
Augmented-reality user interface

EZFF



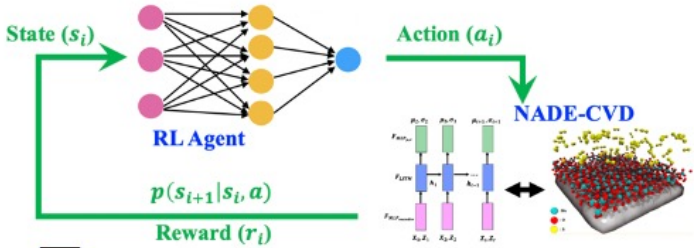
Easy force-field parameterization & uncertain quantification

MISTIQS



Quantum many-body dynamics on quantum computers

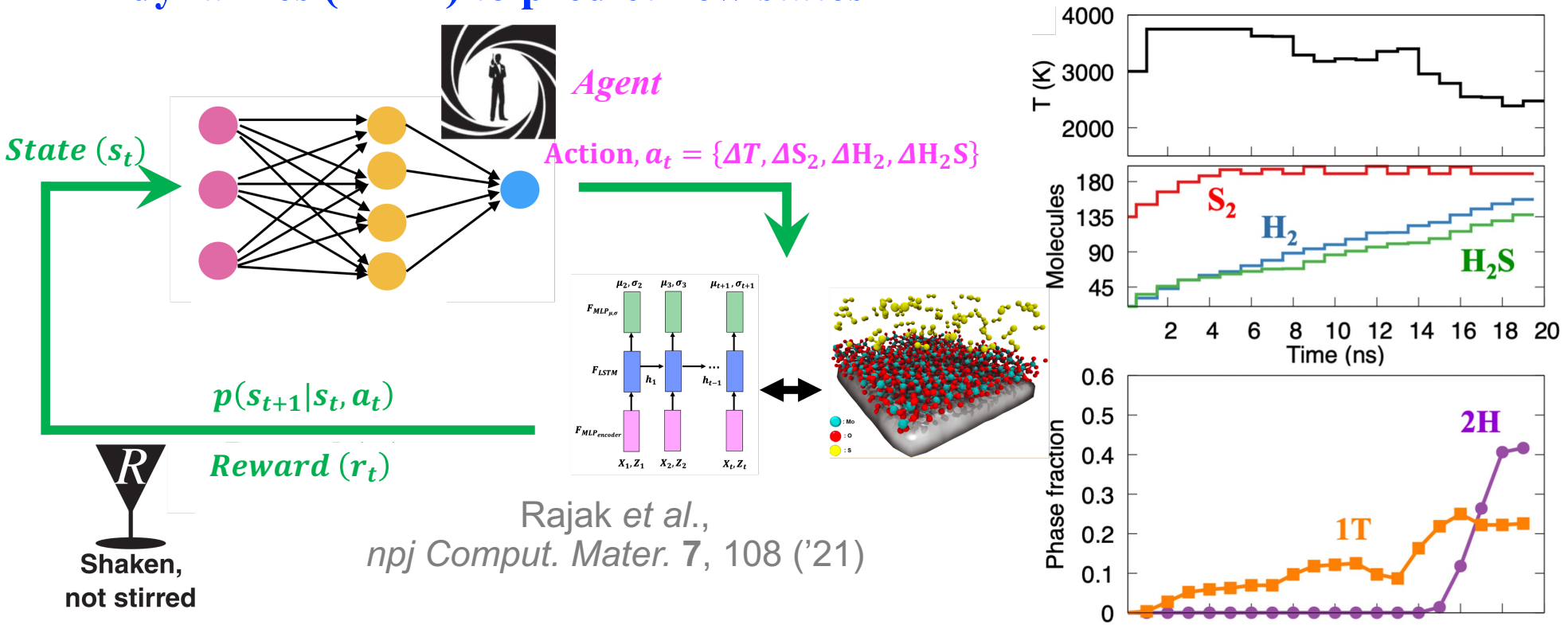
MAITAI



AI tools for materials design

Reinforcement Learning for Growth

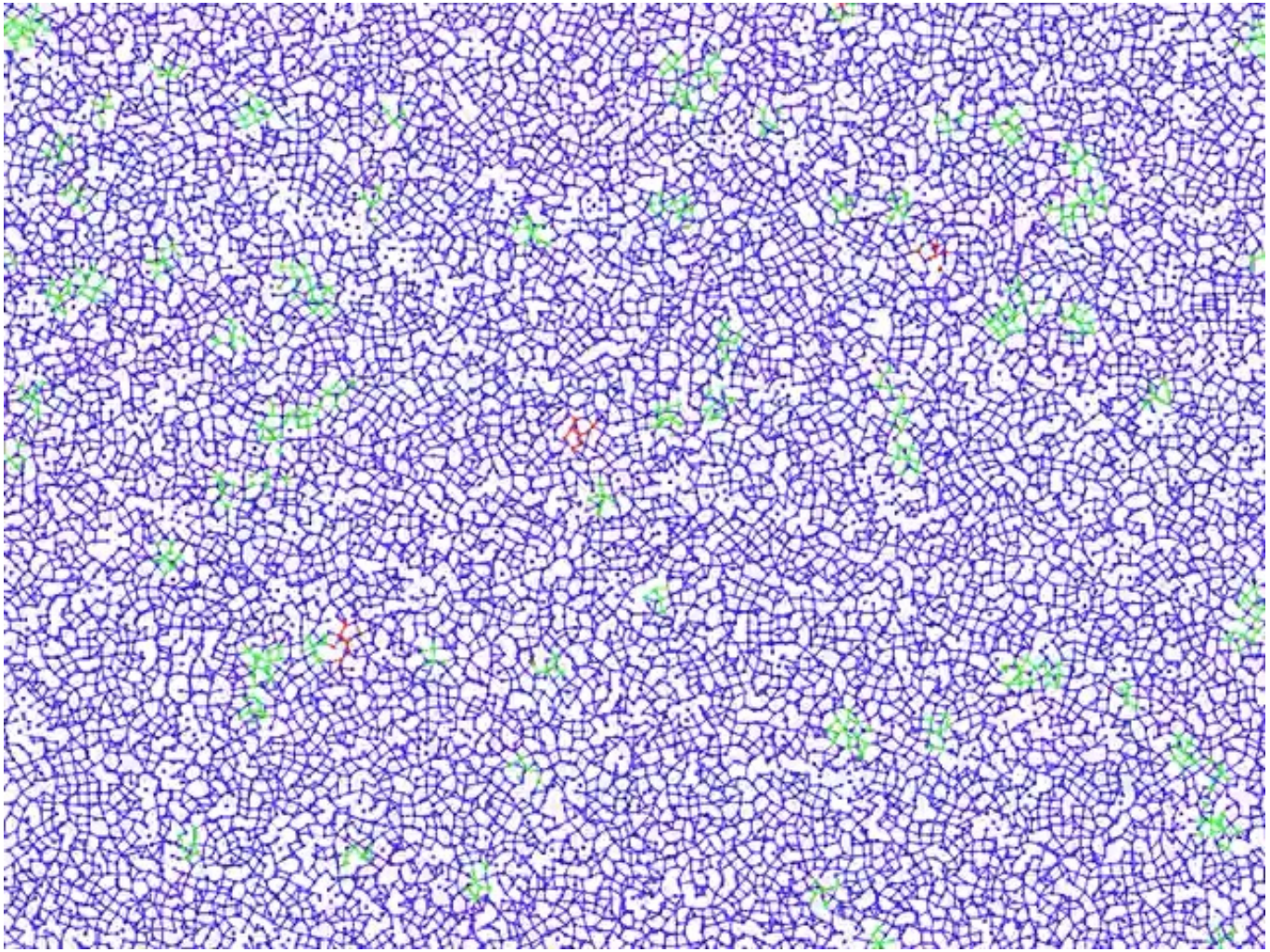
- **In a manner AI plays a board game of Go, use reinforcement learning (RL) to design optimal growth conditions (*e.g.*, temperature & gas-pressure control) to achieve desired properties such as minimal defect density**
- **AI model combines:**
  1. **RL agent to design actions**
  2. **Neural network-based dynamic model trained by reactive molecular-dynamics (RMD) to predict new states**



Rajak *et al.*,
npj Comput. Mater. **7**, 108 ('21)

cf. Sgroi *et al.*, *Phys. Rev. Lett.* ***126***, 020601 ('21)

Computational Synthesis of MoS_2



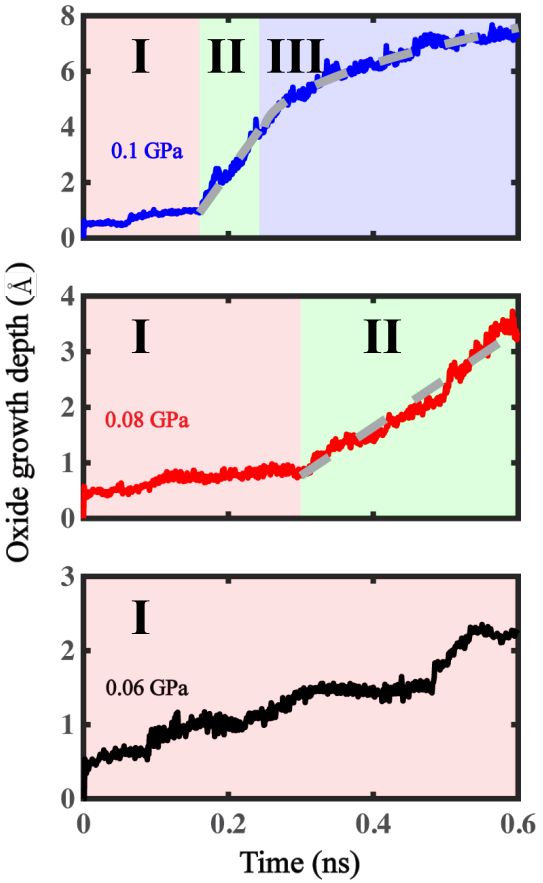
2H
1T
disorder

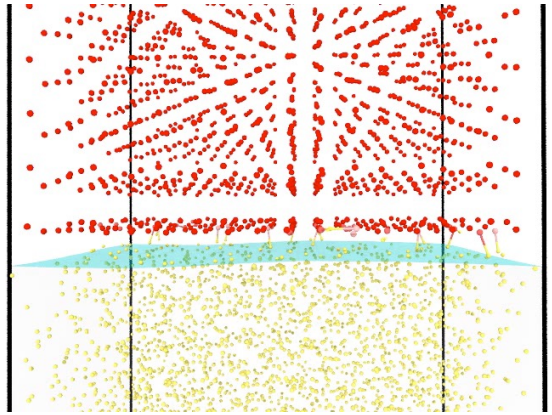
- **Found novel transformation pathways to the stable 2H phase *via* the metastable 1T phase during chemical vapor deposition (CVD) growth of MoS_2**

S. Hong *et al.*, *J. Phys. Chem. Lett.* **10**, 2739 ('19)

Active Control of TMDC Oxidation

- **Transition-metal dichalcogenide (TMDC) semiconductors form the basis of future low-power two-dimensional (2D) electronics**
- **Oxidation of TMDC was proposed as a scalable synthetic pathway to critical semiconductor-insulator interfaces** [Jo, Yang *et al*., *Nano Lett.* ***20***, 8592 ('20); Illarionov, *Nat. Commun*. **11**, 3385 ('20); Liu, *IEEE T. Electron. Dev*., early edition ('23)]
- **Active oxidation (enhanced by plasma, ultraviolet, laser & pressure) is being explored for controlled oxidation of TMDC** [Lai, *Nanoscale* **10**, 18758 ('18); Reidy, *arXiv*:2211.16789 ('23)]
- **Reactive molecular dynamics simulations reveal pressure control of oxidation stages during oxidation of ZrS_2: (I) layer-by-layer to (II) reaction-controlled to (III) diffusion-controlled**



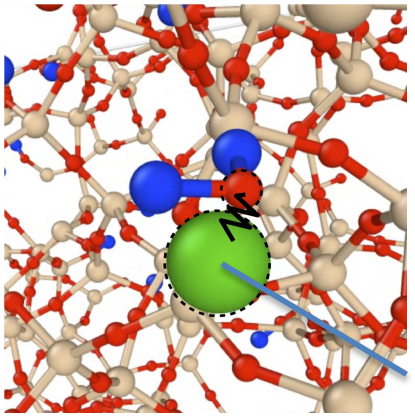


O
Zr (non-bonded)
Zr (bonded)

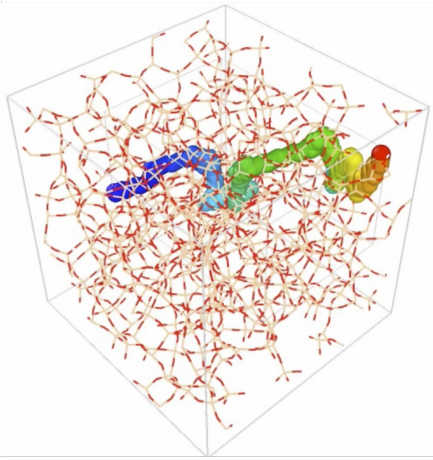
Jo, Yang *et al*., *Nano Lett.* ***20***, 8592 ('20)
Yang, *ACS Nano*, accepted ('23)

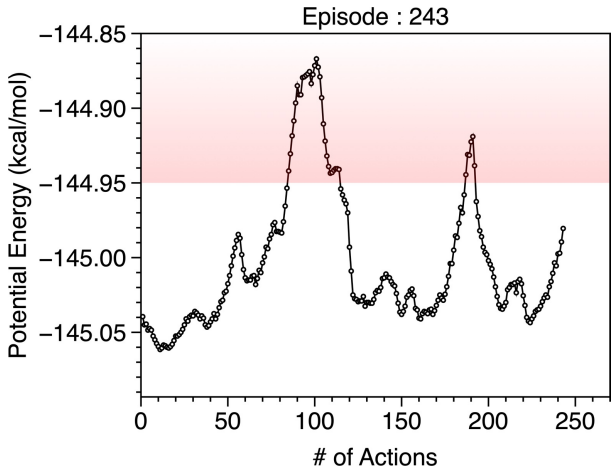
Reinforcement Learning for Long-Time Dynamics

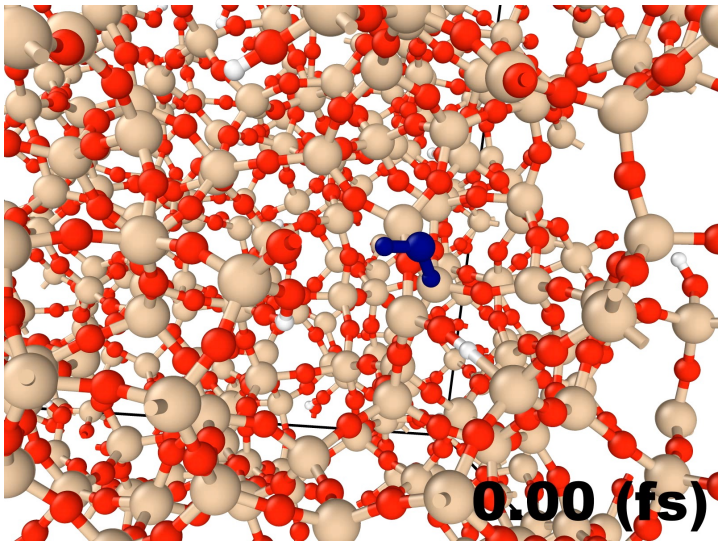
- **Reinforcement learning agents autonomously discover low-activation-barrier migration pathways to study long-time dynamics**
- **Multiple agents share experience using an asynchronously updated replay buffer** [Silver *et al.*, *Nature* **529**, 484 ('16)]
- **Estimate migration time based on the transition-state theory:**



$$t_{\text{migration}} = \sum_{i \in \{\text{activation events}\}} \frac{\hbar}{k_B T} \exp\left(\frac{E_i^{\text{activation}}}{k_B T}\right)$$







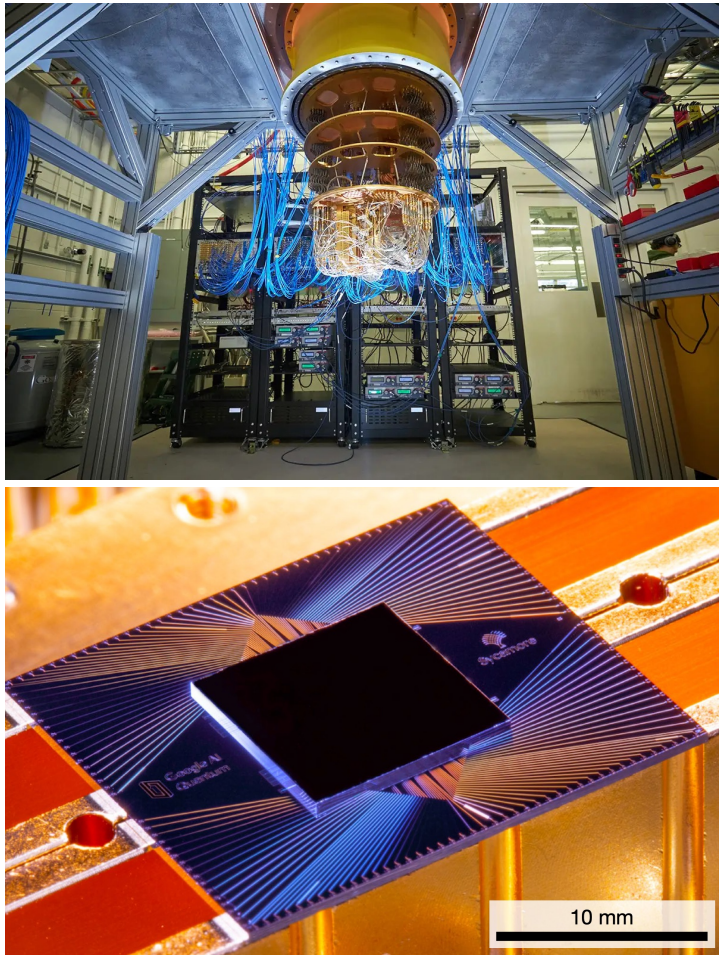
2-seconds trajectory

Low-activation-barrier H_2O migration pathway in SiO_2
Application to CO_2 storage through mineralization in wet silicates

Quantum Computing (QC) for Science

Quantum computing utilizes quantum properties such as superposition & entanglement for computation

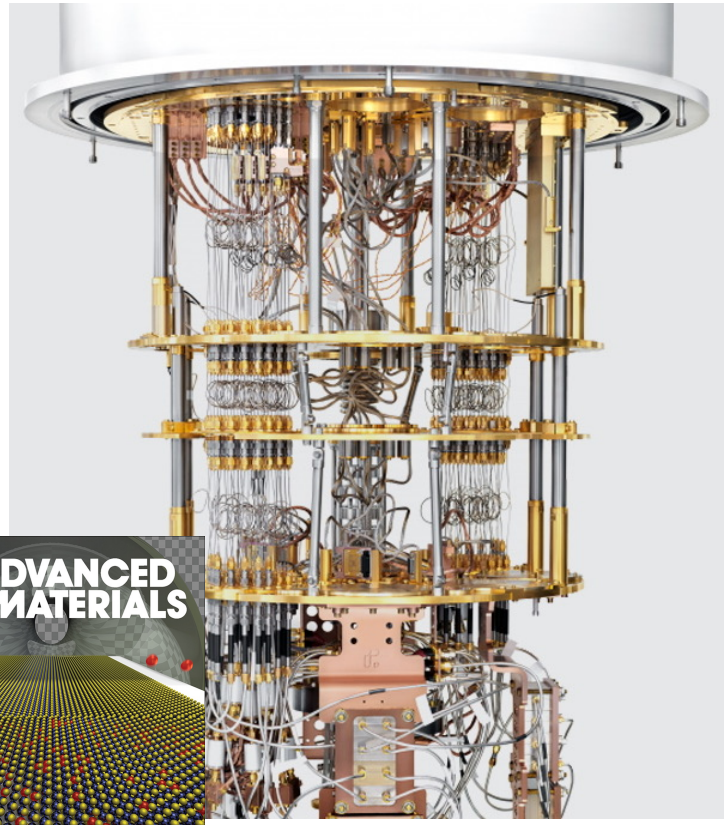
- **U.S. Congress (Dec. 21, '18) signed National Quantum Initiative Act to ensure leadership in quantum computing & its applications**
- **Quantum supremacy demonstrated by Google**
  F. Arute, *Nature* **574**, 505 ('19)
- **Quantum computing for science: *Universal simulator of quantum many-body systems***
  R. P. Feynman, *Int. J. Theo. Phys.* **21**, 467 ('82);
  S. Lloyd, *Science* **273**, 1073 ('96)
- **Success in simulating *static* properties of quantum systems (*i.e.*, ground-state energy of small molecules)**
  A. Aspuru-Guzik *et al.*, *Science* **309**, 1704 ('05)
- **Challenge: Simulate quantum many-body *dynamics* on current-to-near-future noisy intermediate-scale quantum (NISQ) computers**
  J. Preskill, *Quantum* **2**, 79 ('18)

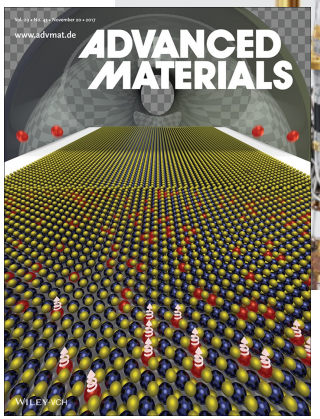


54-qubit Google Sycamore

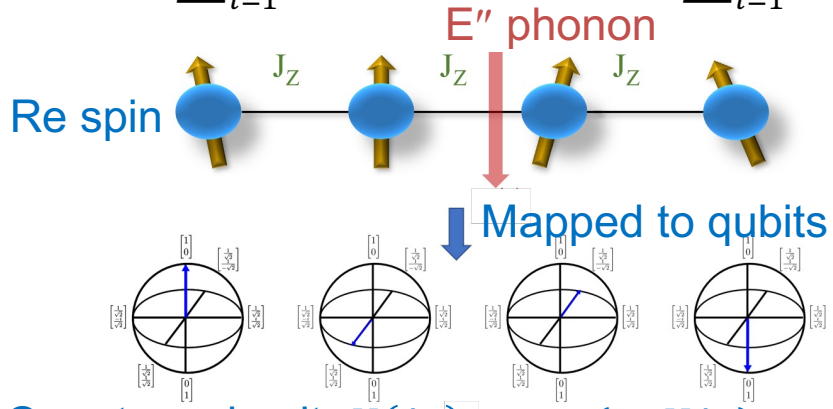
Quantum Computing of Magnetism

- **Simulated quantum many-body dynamics on IBM's Q16 Melbourne & Rigetti's Aspen quantum processors**
- **Electromagnetic-field control of quantum states in a chain of rhenium-magnets in $MoSe_2$ monolayer to realize desired material properties on demand, thereby pushing the envelope of "quantum materials science"**

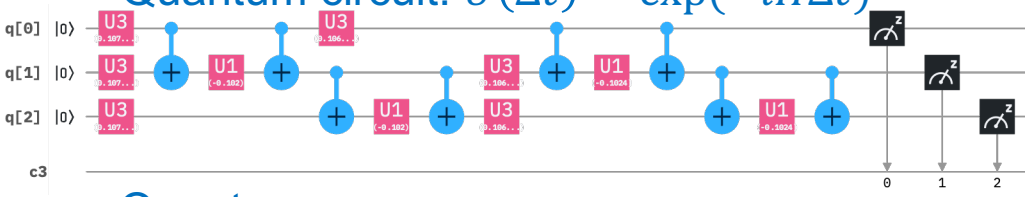




$$H(t) = -J_z \sum_{i=1}^{N-1} \boldsymbol{\sigma}_z^i \boldsymbol{\sigma}_z^{i+1} - \varepsilon_{ph} \sin(\omega_{ph} t) \sum_{i=1}^{N} \boldsymbol{\sigma}_x^i$$



Quantum circuit: $U(\Delta t) = \exp(-iH\Delta t)$



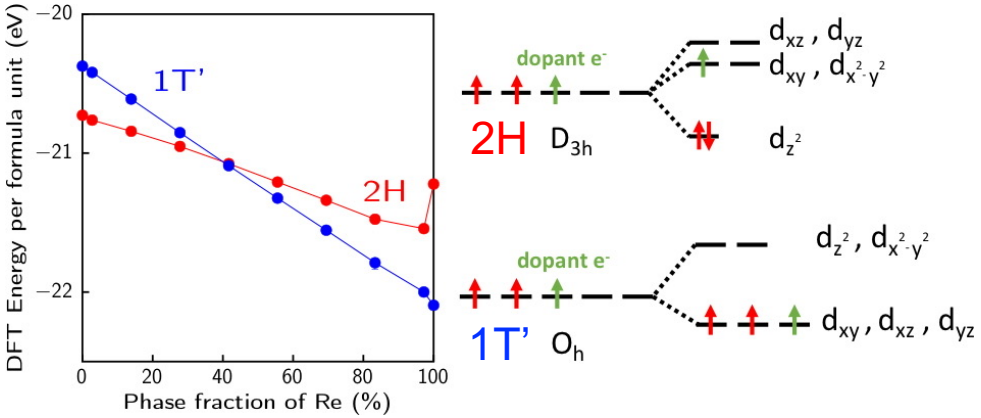
Quantum program

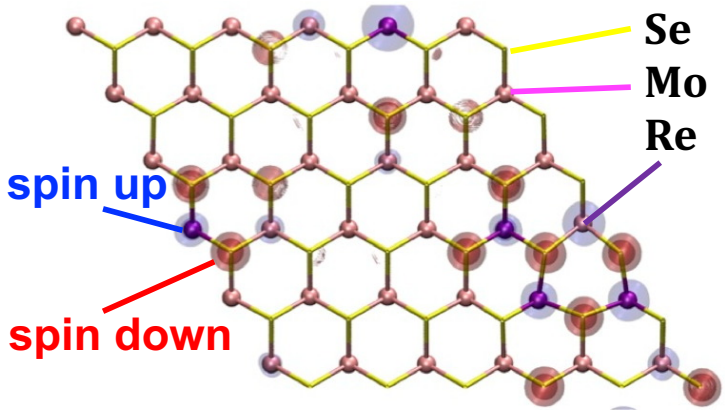
```
32  ····#define·the·two·non-commuting·terms·that·comprise·the·Hamiltonian¬
33  ····Hz·=·PauliTerm("Z",·0,·epsilon_0)¬
34  ····Hy·=·PauliTerm("Y",·0,·epsilon_ph*np.sin(w_ph*t))¬
35  ····#exponentiate·the·terms·of·the·Hamiltonian·for·use·in·Trotter·approx¬
36  ····exp_Hz·=·exponential_map(Hz)(delta_t/(2.0*hbar))¬
37  ····exp_Hy·=·exponential_map(Hy)(delta_t/hbar)¬
```

Do it yourself at https://quantum-computing.ibm.com

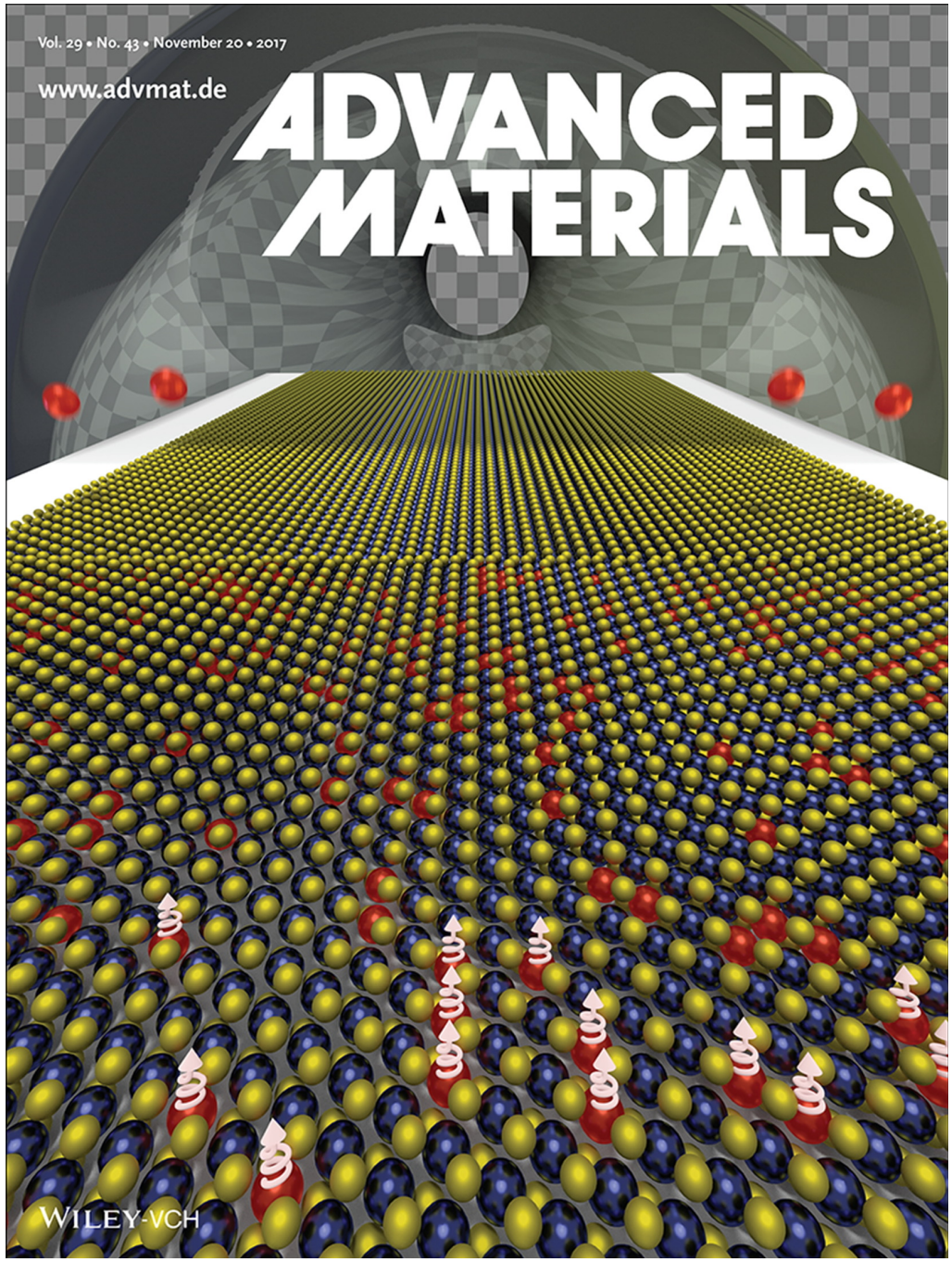
Emergent Magnetism: Structural Transition *via* Doping

- **Experiment at Rice shows 2H-to-1T' phase transformation by alloying $MoSe_2$ with Re**
- **QMD simulations at USC elucidate its electronic origin**
- **Simulation & experiment show novel magnetism centered at Re atoms**





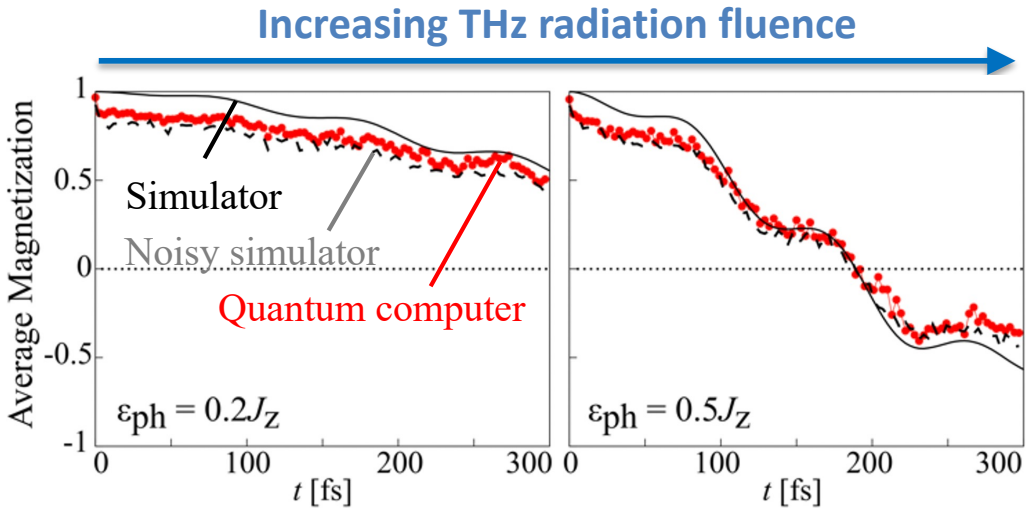
Kochat *et al.*, *Adv. Mater.* **29**, 1703754 ('17)



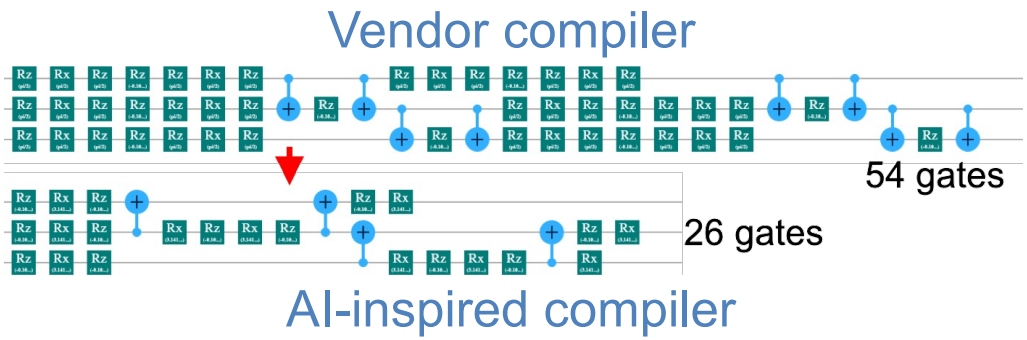
Quantum Dynamics on Quantum Computers

- **Quantum-dynamics simulations on quantum computers show dynamic suppression of magnetization by THz radiation**

Bassman *et al.*,
Phys. Rev. **101**, 184305 ('20)



- **AI-inspired quantum compiler reduced the circuit size by 30% to mitigate environmental noise**

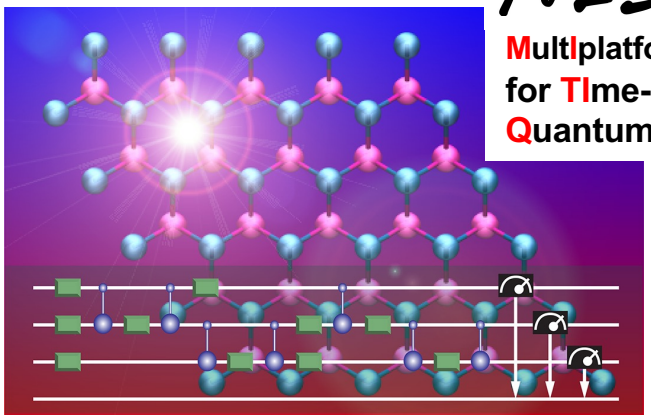


Bassman *et al.*,
Quantum Sci. Tech. **6**, 014007 ('21)

Lindsay Bassman: *Maria Curie Fellow* ('22-);
Science, She Says Award ('23)

- **Full-stack, cross-platform software for quantum dynamics simulations on NISQ computers**

MISTIQS
Mult**I**platform **S**oftware for **TI**me-dependent **Q**uantum **S**imulation

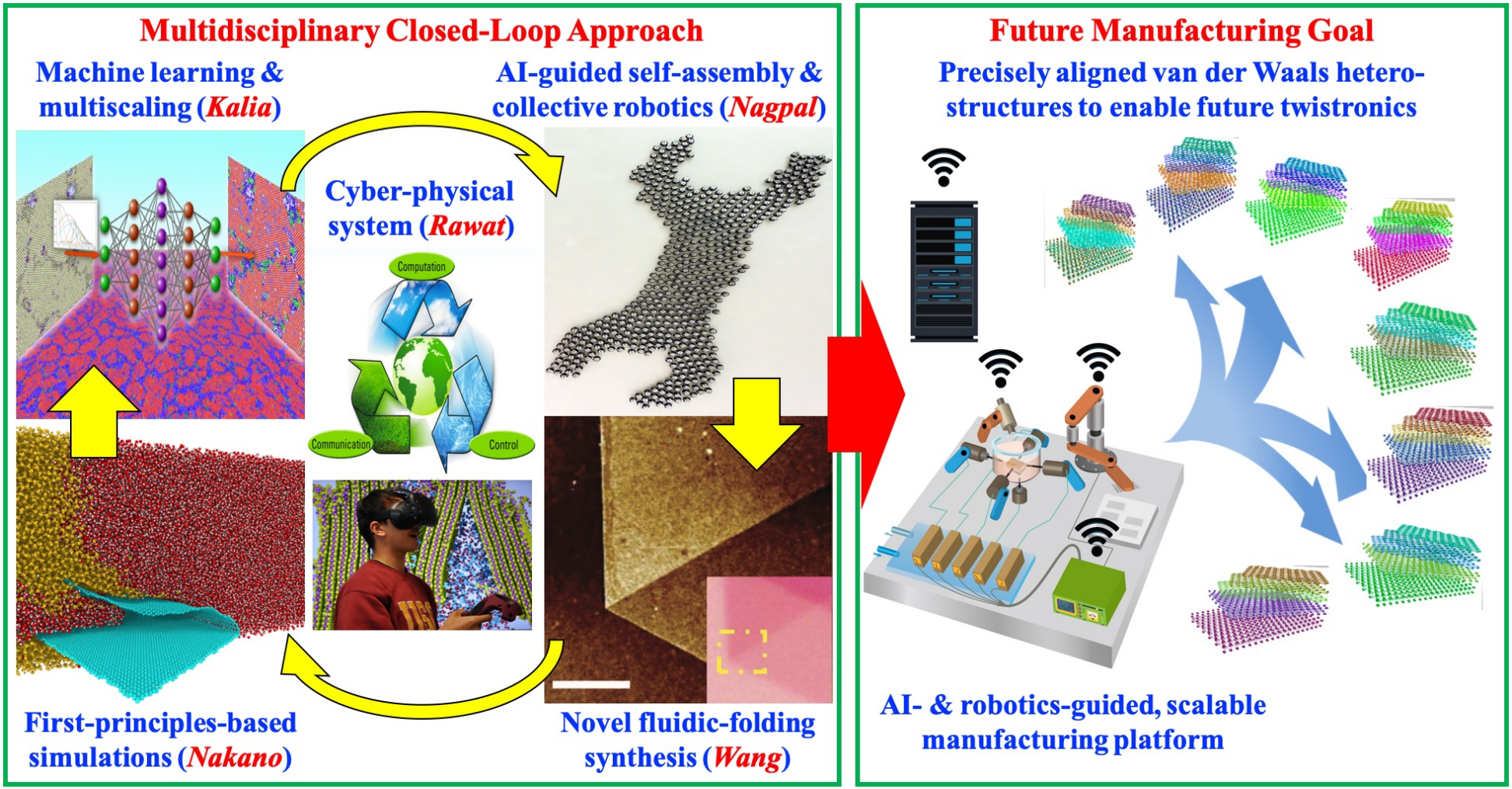


Powers *et al.*, *SoftwareX* **14**, 100696 ('21)
https://github.com/USCCACS/MISTIQS

Princeton-USC-Howard Future Manufacturing

FMRG: Artificial Intelligence Driven Cybermanufacturing of Quantum Material Architectures

$3.75M NSF project (2020-2025)

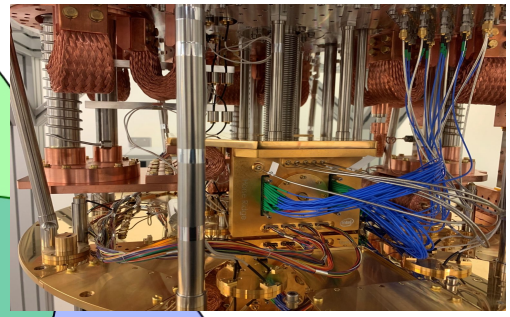


Nagpal (Princeton); Kalia, Nakano, Wang (USC); Rawat (Howard)

Training Cyber Science Workforce

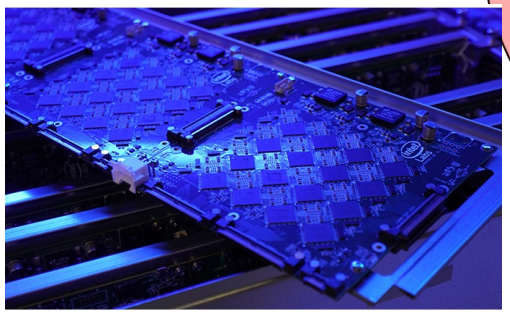
- New generation of computational scientists at the nexus of exascale computing, quantum computing & AI
- Unique dual-degree program: Ph.D. in materials science or physics, along with MS in computer science specialized in high-performance computing & simulations, MS in quantum information science or MS in materials engineering with AI

Horse Ridge II
Quantum computer



Cybertraining on exa + quantum + AI platforms

Neuromorphic
Pohoiki Springs



Exascale
Aurora

USC-Howard Cybertraining

CyberMAGICS: Cyber Training on Materials Genome Innovation for Computational Software

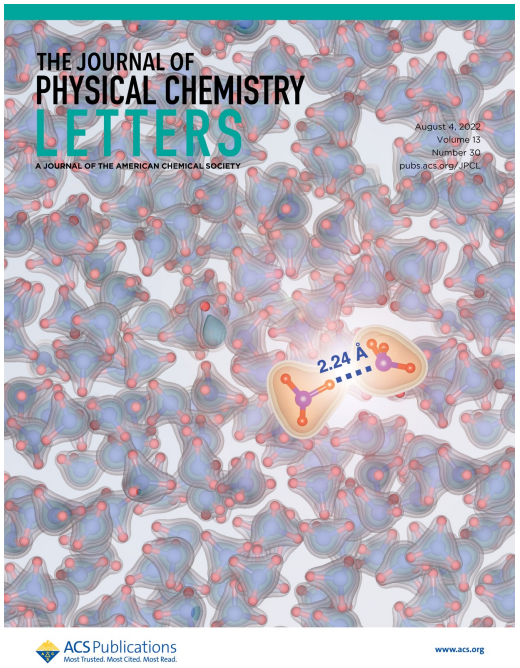
- **Train a new generation of materials cyberworkforce, who will solve challenging materials genome problems through innovative use of advanced cyberinfrastructure at the exa-quantum-AI nexus**

$1M NSF CyberTraining (2021-25) project

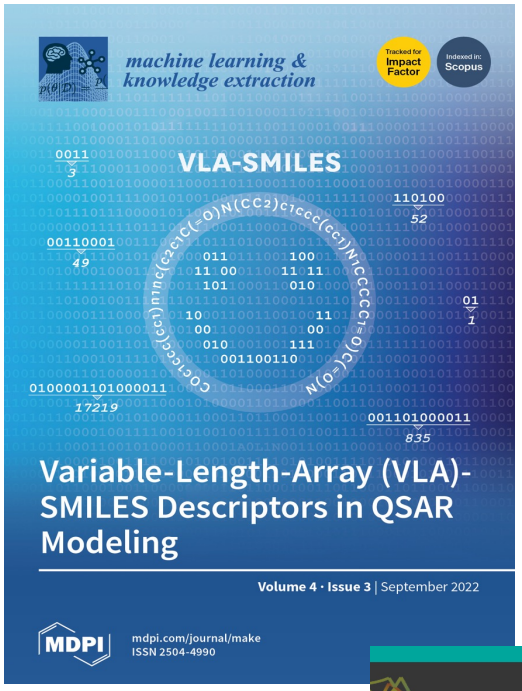
Nakano, Nomura, Vashishta (USC); Dev, Wei (Howard)

External advisors: T. Germann, S. Plimpton, *et al*.

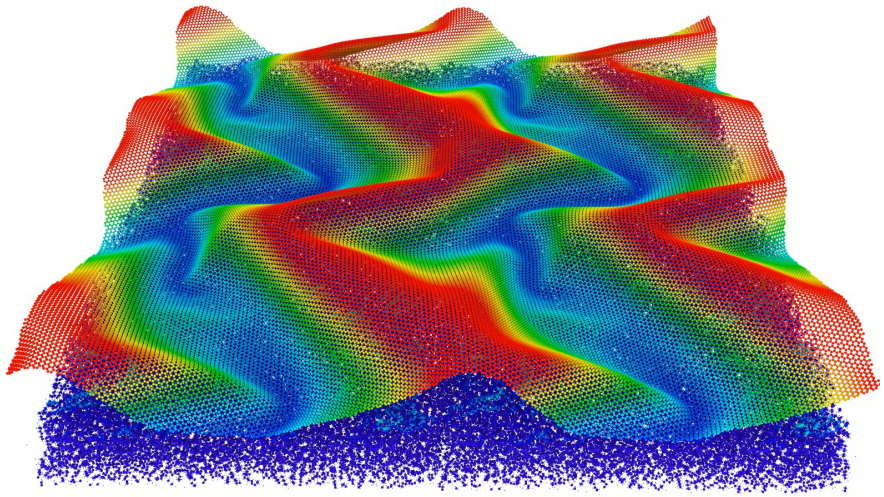
Other Applications



Green ammonia for renewable power (Aug. 4, ’22)

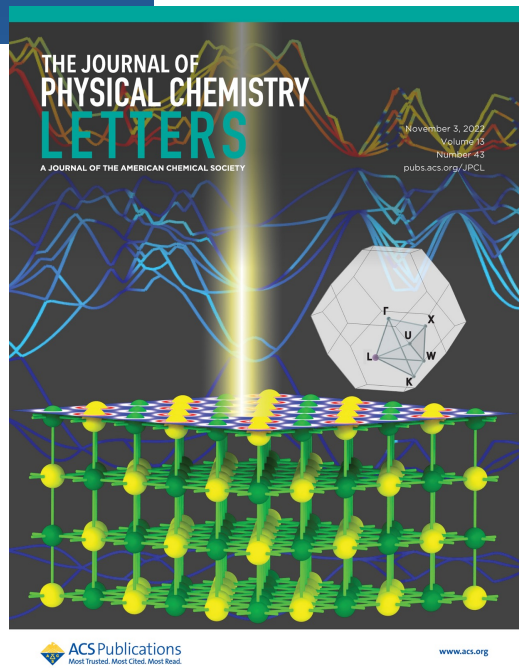


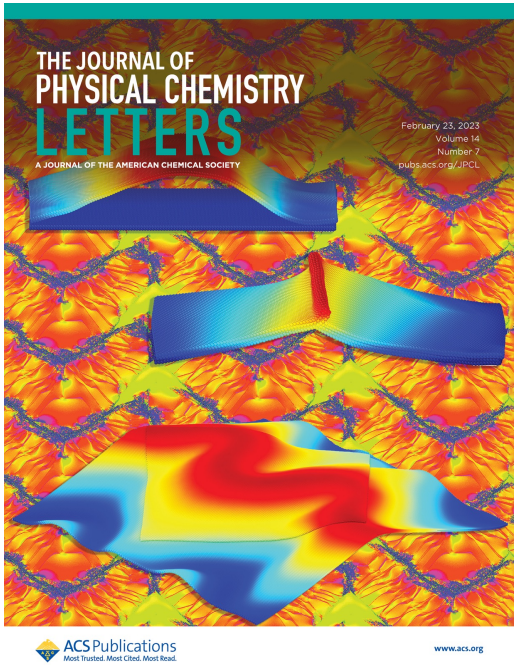
Molecular design (Sep. ’22)



Strain self-assembly of 2D metasurface (Feb. 23, ’23)

Phase-change memory (Nov. 3, ’22)

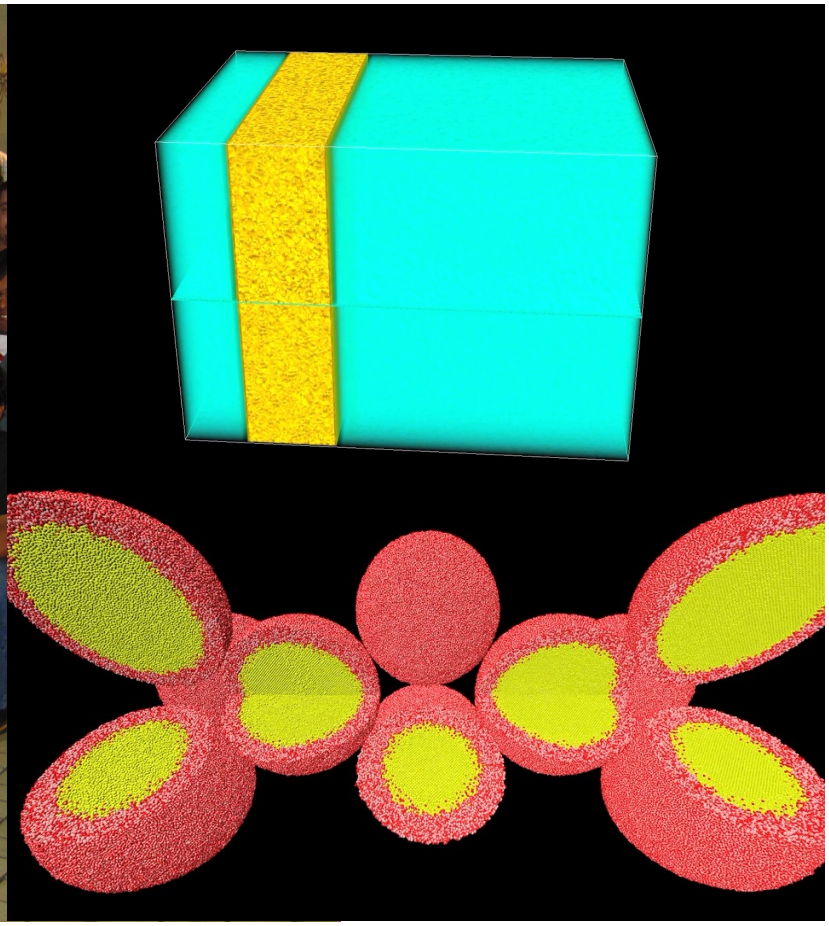




Conclusion

1. Large spatiotemporal-scale quantum, reactive & neural molecular dynamics simulations based on a common algorithmic framework
2. Broad applications of (exa+AI+quantum)4Science

Thank You



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DOE-CMS/Neutron/INCITE/Aurora-ESP,
NSF-FM/CyberTraining, ONR-MURI×2, Sony