Quantum Molecular Dynamics (QMD) Simulations

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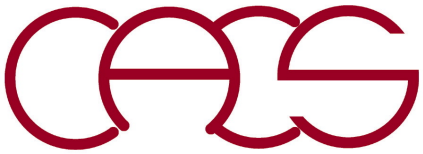
QMD simulation of quantum materials on post-exaflop/s supercomputers is the foundation of future semiconductors, quantum computing, and AI



Changing Computing Landscape for Science

Post-exascale Computing for Science

Compute Cambrian explosion

AI for Science

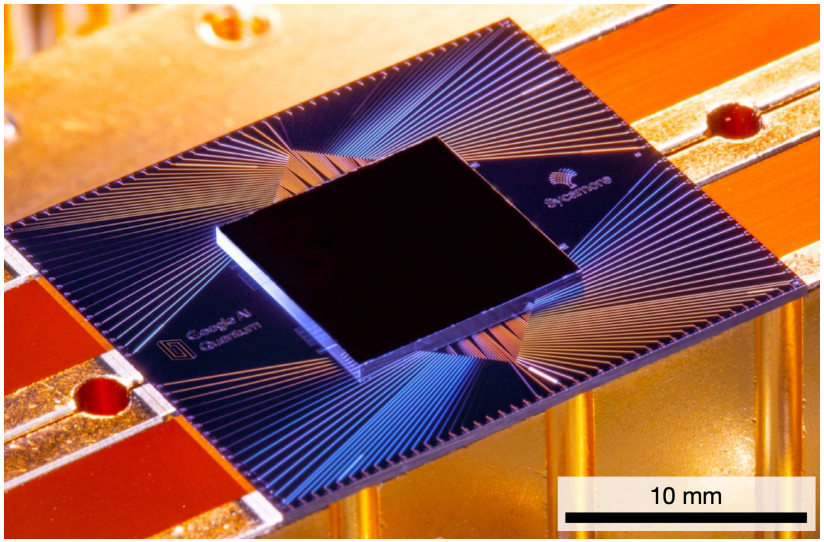
DOE readies multibillion-dollar AI push

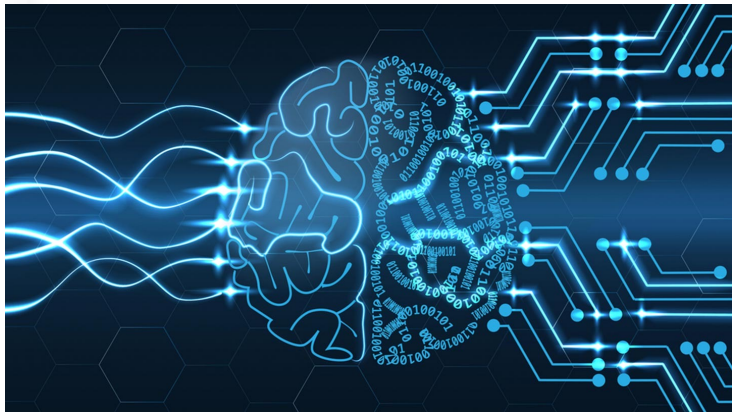
U.S. supercomputing leader is the latest big backer in a globally crowded field

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

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

Quantum Computing for Science





Use all to advance science!

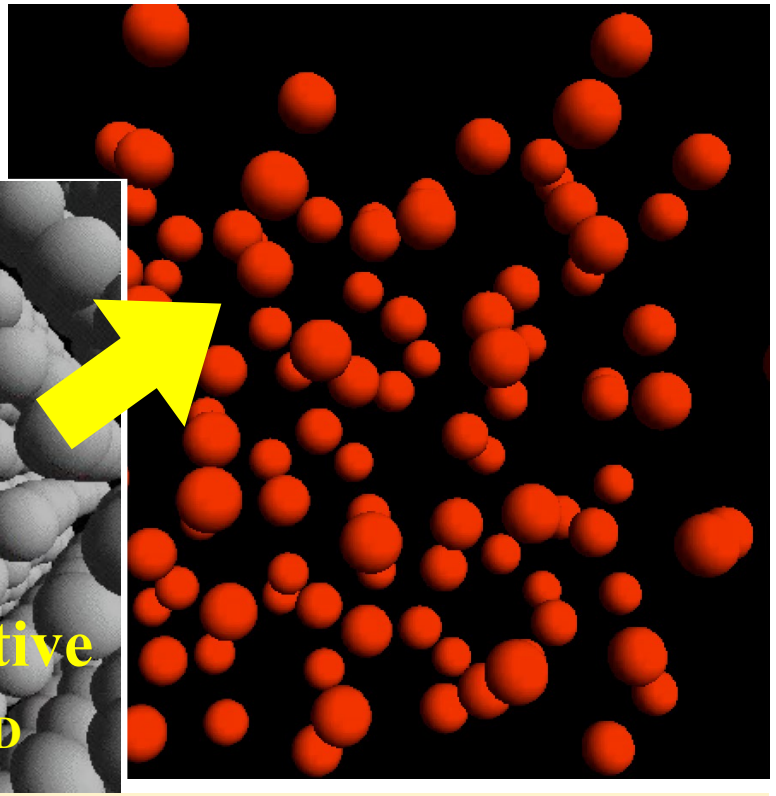
Why post-exa–quantum–AI nexus at USC?

USC Frontiers of Computing

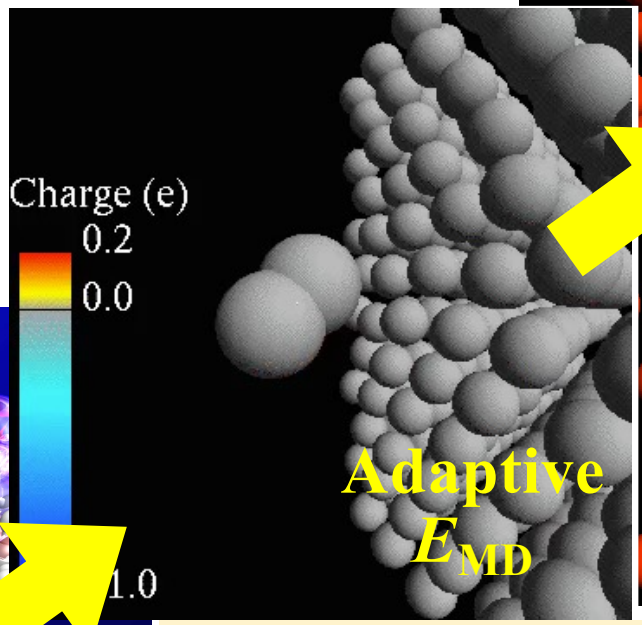
- **New School of Advanced Computing:** https://sac.usc.edu
- **30 senior & 60 junior & mid-level hires**

Molecular Dynamics & Machine Learning

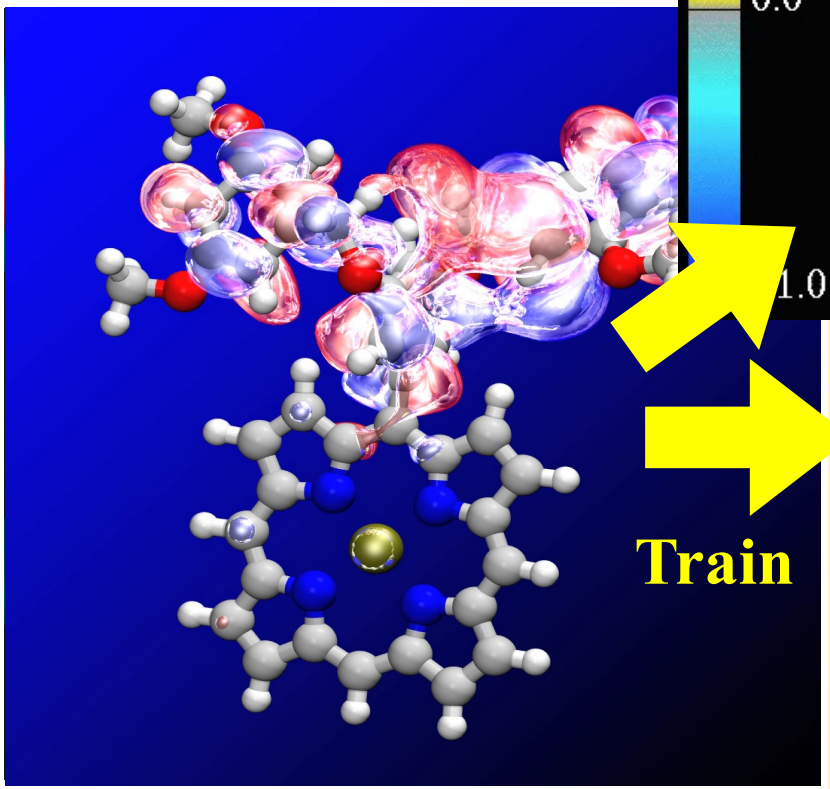
Molecular Dynamics (*MD*)



Reactive MD (*RMD*)

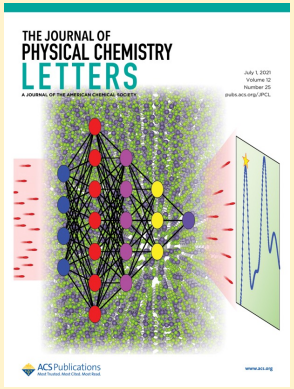


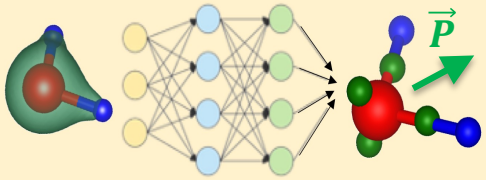
Nonadiabatic quantum MD (*NAQMD*)



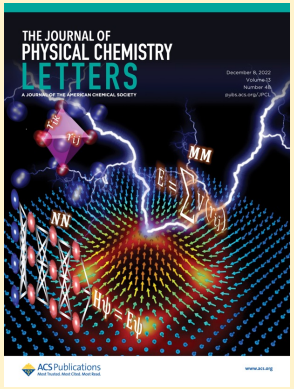
Train

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



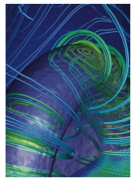
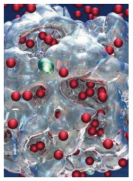
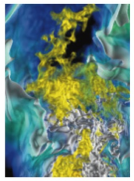
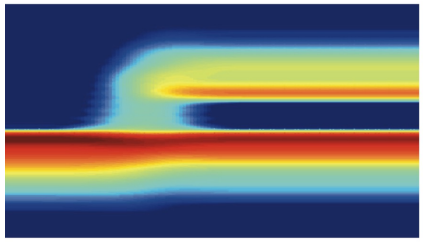


Physical Review Letters
Editor's choice
(May 25, 2021)



Exascale

BES

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)

On-demand hydrogen production from water

10^9-atom RMD

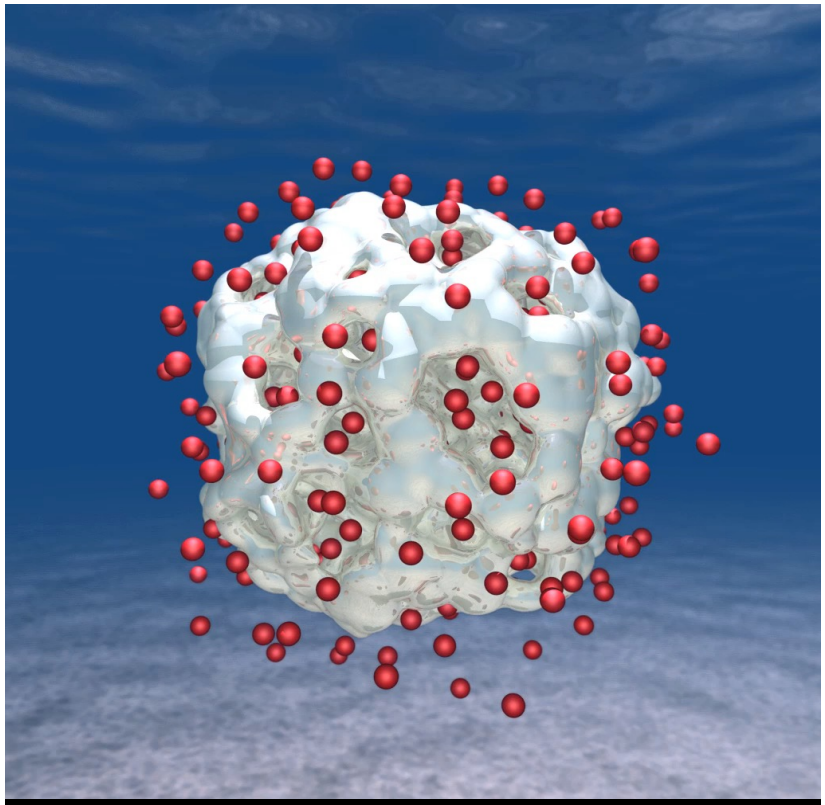
Shekhar *et al.*,
Phys. Rev. Lett.
111, 184503 ('13)

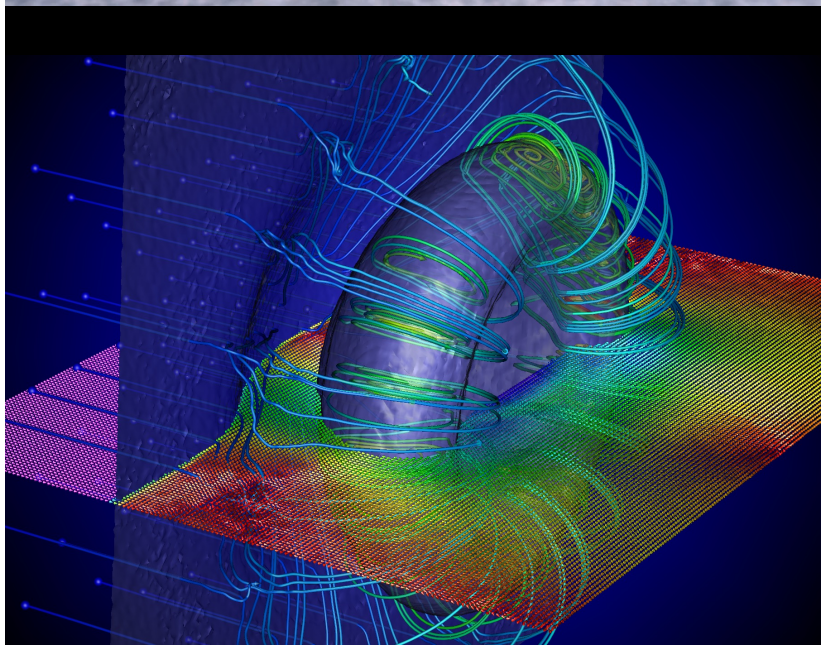
Fluid dynamics atom-by-atom

NOVEMBER 3–5, 2015

ROCKVILLE, MARYLAND

U.S. DEPARTMENT OF ENERGY





Quantum Molecular Dynamics (QMD)

$$M_I \frac{d^2}{dt^2}\mathbf{R}_I = -\frac{\partial}{\partial \mathbf{R}_I} E[\{\mathbf{R}_I\}, \psi(\mathbf{r}_1 \ldots, \mathbf{r}_N)] \; (I = 1, \ldots, N_{\text{atom}})$$

First molecular dynamics using an empirical interatomic interaction

A. Rahman, *Phys. Rev*. **136**, A405 ('64)

$$\psi(\mathbf{r}_1 \ldots, \mathbf{r}_N) \leftarrow \text{argmin}\, E[\{\mathbf{R}_I\}, \psi(\mathbf{r}_1 \ldots, \mathbf{r}_N)]$$

Density functional theory (DFT)

Hohenberg & Kohn, *Phys. Rev*. **136**, B864 ('64)
W. Kohn, *Nobel chemistry prize*, '98

$O(C^N)$ → $O(N^3)$

1 *N*-electron problem intractable → ***N* 1-electron problems tractable**

$\psi(\mathbf{r}_1 \ldots, \mathbf{r}_N)$ $\quad$ $\{\psi_i(\mathbf{r}) | i = 1, \ldots, N\}$

G. Battimelli *et al*., *Computer Meets Theoretical Physics* ('20) pp. 58 & 128

$O(N)$ DFT algorithms

- **Divide-&-conquer DFT** [W. Yang, *Phys. Rev. Lett*. **66**, 1438 ('91); F. Shimojo *et al*., *Comput. Phys. Commun*. **167**, 151 ('05); *Phys Rev. B* **77**, 085103 ('08); *Appl. Phys. Lett*. **95**, 043114 ('09); *J. Chem. Phys*. **140**, 18A529 ('14)]
- **Quantum nearsightedness principle** [W. Kohn, *Phys. Rev. Lett*. **76**, 3168 ('96); E. Prodan & W. Kohn, *P. Nat. Acad. Sci*. **102**, 11635 ('05)]
- **A comprehensive review** [Bowler & Miyazaki, *Rep. Prog. Phys*. **75**, 036503 ('12)]

Born-Oppenheimer Approximation

- **Consider a system of N electrons & N_{atom} nuclei, with the Hamiltonian**

$$\widetilde{H} = \sum_{I=1}^{N_{\text{atom}}} \frac{\mathbf{P}_I^2}{2M_I} + H(\{\mathbf{r}_i\},\{\mathbf{R}_I\})$$

nucleus momentum; electron position; nucleus position

$$= \sum_{I=1}^{N_{\text{atom}}} \left[\frac{\mathbf{P}_I^2}{2M_I} + V_{\text{ext}}(\mathbf{R}_I)\right] + \sum_{i=1}^{N} \left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial \mathbf{r}_i^2} + v_{\text{ext}}(\mathbf{r}_i)\right]$$
$$+ \frac{1}{2}\sum_{i\neq j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_{i,J} \frac{Z_J e^2}{|\mathbf{r}_i - \mathbf{R}_J|} + \frac{1}{2}\sum_{I\neq J} \frac{Z_I Z_J e^2}{|\mathbf{R}_I - \mathbf{R}_J|}$$

nucleus charge

- **Due to the much larger nuclei masses (M_I) compared to the electron mass (m), the quantum-mechanical wave function of the system is separable to those of the electrons & nuclei**
- **At ambient conditions, the electronic wave function remains in its ground state ($|\Psi_0\rangle$) corresponding to the instantaneous nuclei positions ($\{\mathbf{R}_I\}$), with the latter following classical mechanics**

$$M_I \frac{d^2}{dt^2}\mathbf{R}_I = -\frac{\partial}{\partial \mathbf{R}_I}\langle \Psi_0 | H(\{\mathbf{r}_i\},\{\mathbf{R}_I\}) | \Psi_0 \rangle$$

Born-Oppenheimer Approximation

1927 № 20

ANNALEN DER PHYSIK

VIERTE FOLGE. BAND 84

1. *Zur Quantentheorie der Molekeln;*
von M. Born und R. Oppenheimer

Academic Genealogy

Lindsay Bassman Pedigree

Carl Gauss
1777 – 1855
University of Gottingen
Math/Physics 1799

Christopher Gudermann
1798 – 1852
University of Gottingen
Math 1832

Karl Weierstrass
1815 – 1897
University of Bonn
Math

Carl Runge
1856 – 1927
Berlin University
Math/Physics 1880

Max Born
1882 – 1970
University of Göttingen
Physics/Math 1906

J. Robert Oppenheimer
1904 – 1967
University of Göttingen
Physics 1927

David Bohm
1917 – 1992
University of California, Berkeley
Physics 1943

David Pines
1924 – 2018
Princeton University
Physics 1950

Setsuo Ichimaru
1935 -
University of Illinois
Physics 1962

Aiichiro Nakano
1963 –
University of Tokyo
Physics 1989

Lindsay Elizabeth Bassman
1989 –
University of Southern California
Physics 2020

Complexity Reduction: Density Functional Theory

- **P. Hohenberg & W. Kohn, "Inhomogeneous electron gas"**
  Phys. Rev. **136**, B864 ('64)

  The electronic ground state is a functional of the electron density $\rho(\mathbf{r})$

- **W. Kohn & L. Sham, "Self-consistent equations including exchange & correlation effects"** *Phys. Rev.* **140**, A1133 ('65)

  Derived a formally exact self-consistent single-electron equations for a many-electron system

Energy Functional

Exchange-correlation (xc) functional *via* Kohn-Sham decomposition

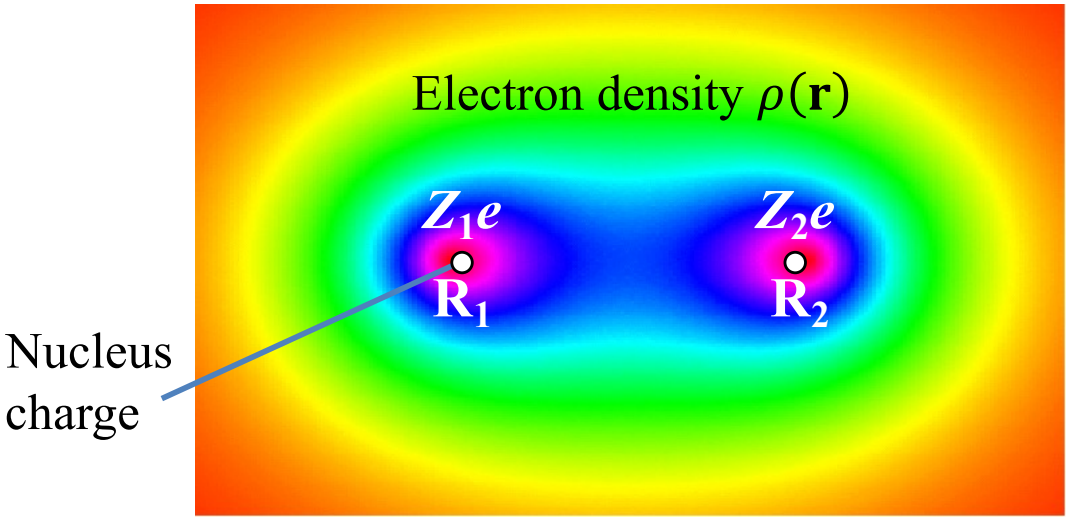
$$E[\rho(\mathbf{r})] = T_s[\rho(\mathbf{r})] + \int d\mathbf{r} v(\mathbf{r})\rho(\mathbf{r}) + \frac{1}{2}\int d\mathbf{r}d\mathbf{r}' \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + E_{xc}[\rho(\mathbf{r})]$$

Kinetic energy of non-interacting electrons

External potential

Hartree energy (mean-field approximation to the electron-electron interaction energy)

Exchange-correlation energy



Kohn-Sham Equation

- **Many-electron problem is equivalent to solving a set of one-electron Schrödinger equations called Kohn-Sham (KS) equations**

$$\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial \mathbf{r}^2} + v_{\mathrm{KS}}(\mathbf{r})\right]\psi_n(\mathbf{r}) = \epsilon_n\,\psi_n(\mathbf{r})$$

KS wave function — KS energy

- **KS potential**

$$v_{\mathrm{KS}} = v(\mathbf{r}) + \int d\mathbf{r}'\,\frac{e^2\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + v_{\mathrm{xc}}(\mathbf{r})$$

exchange-correlation (xc) potential

$$\rho(\mathbf{r}) = \sum_n \Theta(\mu-\varepsilon_n)|\psi_n(\mathbf{r})|^2$$

step function — chemical potential

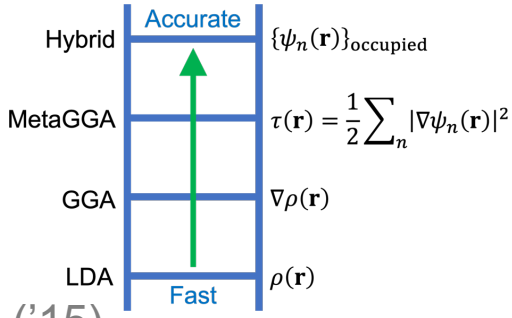
$$v_{\mathrm{xc}}(\mathbf{r}) \equiv \frac{\delta E_{\mathrm{xc}}}{\delta\rho(\mathbf{r})}$$

$$N = \sum_n \Theta(\mu-\varepsilon_n)$$

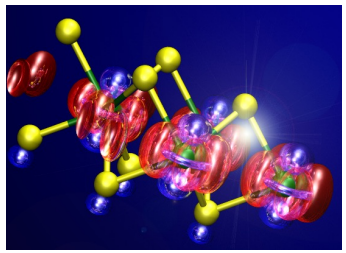
W. Kohn & L. J. Sham, "Self-consistent equations including exchange and correlation effects," *Phys. Rev.* **140**, A1133 ('65)

Abstraction: Exchange-Correlation Functional

- **Universal functional (of density) that describes many-body effects beyond the mean-field approximation**
- **Some commonly used exchange-correlation functionals**
  - > **GGA (generalized gradient approximation)**
    PBE: Perdew, Burke & Ernzerhof, *Phys. Rev. Lett.* **77**, 3865 ('96)
  - > **MetaGGA**
    SCAN: Sun, Ruzsinszky & Perdew, *Phys. Rev. Lett.* **115**, 036402 ('15)
  - > **Hybrid exact-exchange (Hartree-Fock) functionals**
    HSE: Heyd, Scuseria & Ernzerhof, *J. Chem. Phys.* **118**, 8207 ('03)

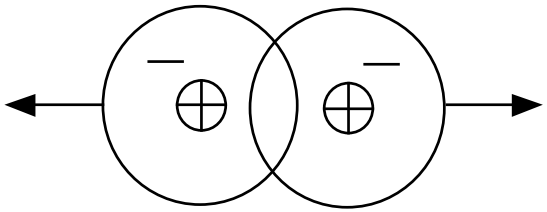


- **Others supported by QXMD code: Select an appropriate functional for the material system & purpose**
  - > **DFT+U method for transition metals**
    $\delta E_{\text{DFT+U}}/\delta n_i = \epsilon_{\text{DFT}} + U(\frac{1}{2} - n_i)$
    Anisimov *et al.*, *Phys. Rev. B* **44**, 943 ('91)



Localized
d-electrons

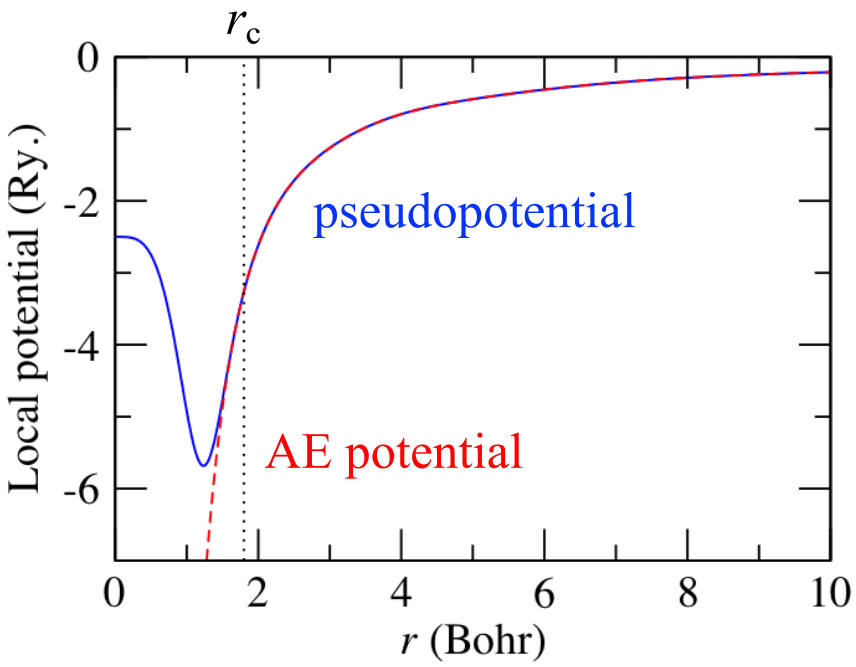
  - > **DFT-D: van der Waals (vDW) functional for molecular crystals & layered materials**
    $E_{\text{disp}} = -s_6 \sum_{i<j} \frac{C_{ij}}{R_{ij}^6} f_{\text{damp}}(R_{ij})$
    Grimme, *J. Chem. Phys.* **132**, 154104 ('10)

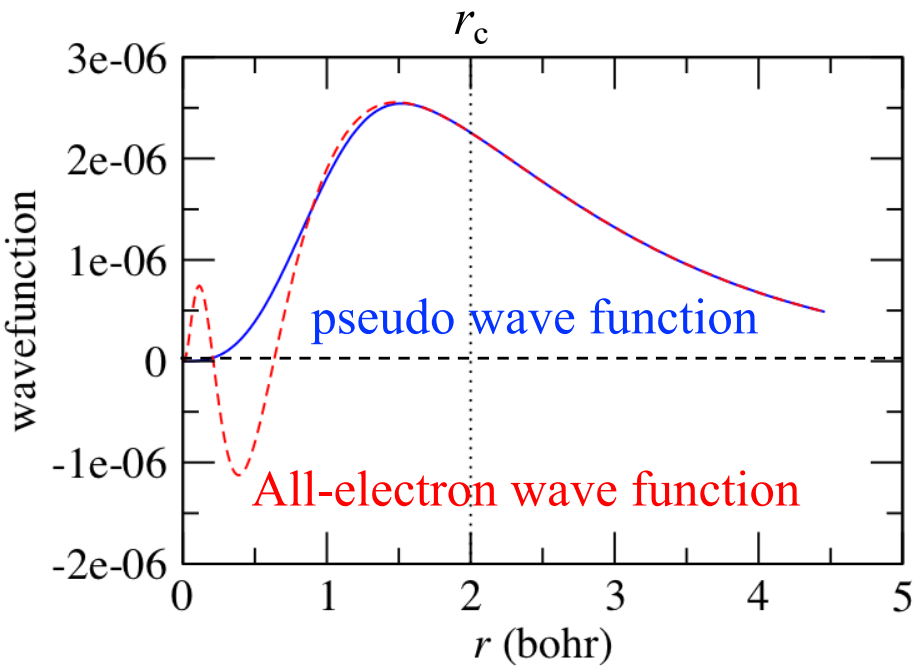


  - > **vdW: Nonlocal correlation functional**
    $E_c^{\text{nl}} = \frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\,\rho(\mathbf{r})\phi(\mathbf{r},\mathbf{r}')\rho(\mathbf{r}')$
    Dion *et al.*, *Phys. Rev. Lett.* **92**, 246401 ('04)

Abstraction: Pseudopotential

- **Consider only (chemically active) valence electrons**
  ***e.g.*, silicon — $1s^2 2s^2 2p^6 3s^2 3p^2$**
- **Pseudopotentials & smooth, nodeless pseudo-wave functions are constructed to agree with the all-electron counterparts beyond a cutoff radius r_c**





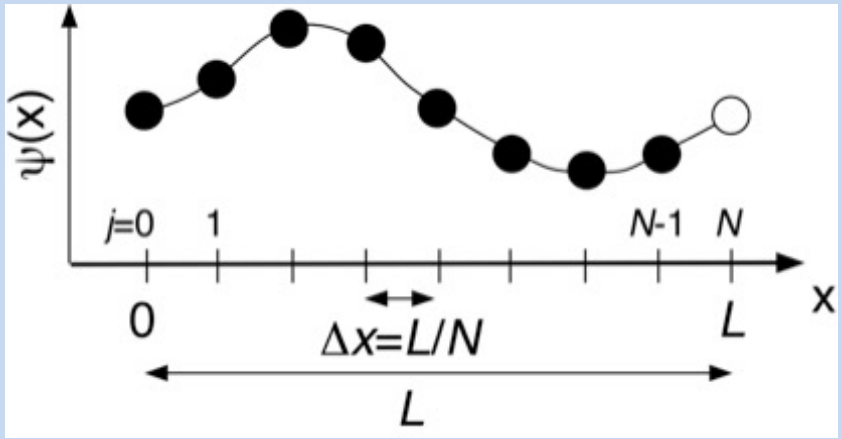
- **Commonly used pseudopotentials**
  - > **Norm-conserving:** Troullier & Martins, *Phys. Rev. B* **41**, 1993 ('91)
  - > **Ultrasoft:** Vanderbilt, *Phys. Rev. B* **41**, 7892 ('90)
  - > **Projector augmented wave (PAW):** Blochl, *Phys. Rev. B* **50**, 17953 ('94)

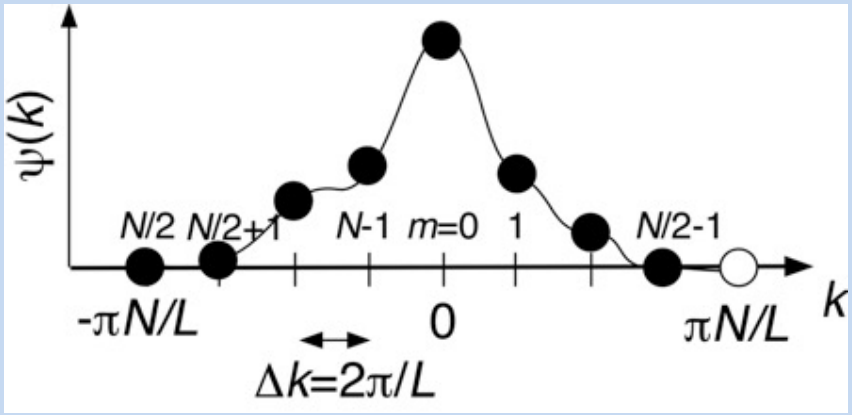
Representation: Plane-Wave Basis

- **Pseudopotentials result in slowly varying wave functions that can be represented on a regular grid, which in turn can be represented as a linear combination of plane waves, *i.e.*, Fourier transform**

$$\psi(\mathbf{r}_j) = \sum_{\mathbf{k}_n} \psi_{\mathbf{k}_n} \exp(i\mathbf{k}_n \bullet \mathbf{r}_j)$$

1D example



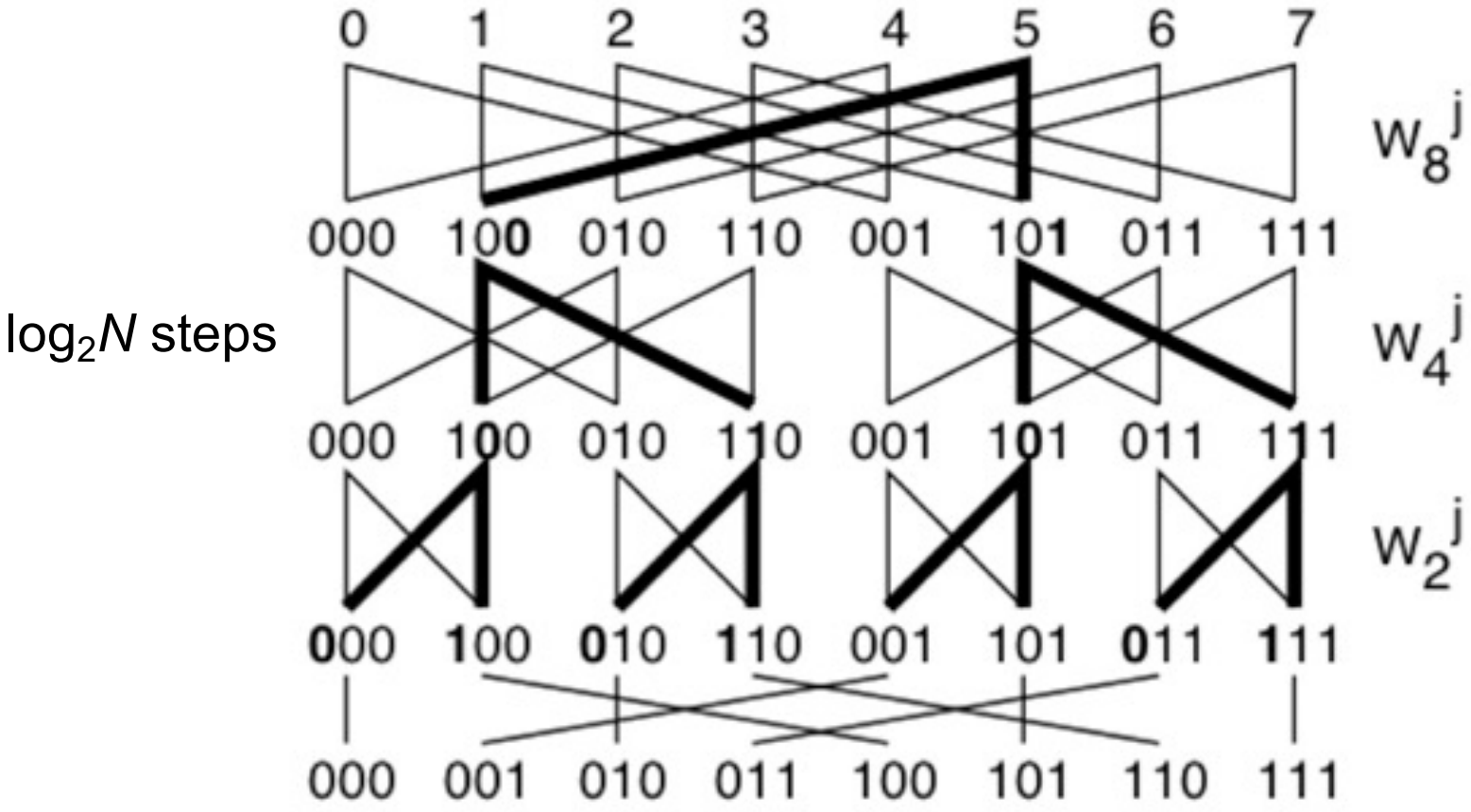


$$x_j = \frac{jL}{N};\ k_n = \frac{2\pi n}{L}$$

Numerics: Fast Fourier Transform

- ***O*(*N*log*N*) fast Fourier-transform (FFT) algorithm is typically used to perform Fourier transform**

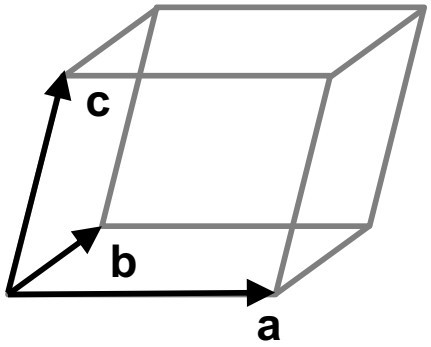
$$\psi(x_j) = \sum_{k_n} \psi_{k_n} \exp(ik_n x_j)$$



Butterfly (hypercube) data-exchange network

Periodic Solid

- **Consider a periodic solid with the unit cell spanned by vectors a, b & c**



- **Fourier transform of a periodic function**

$$u(\mathbf{r}) = \sum_{\mathbf{G}} u_{\mathbf{G}} \exp(i\mathbf{G} \bullet \mathbf{r})$$

$$\mathbf{G} = \frac{2\pi}{\mathbf{a} \bullet (\mathbf{b} \times \mathbf{c})} [l(\mathbf{b} \times \mathbf{c}), m(\mathbf{c} \times \mathbf{a}), n(\mathbf{a} \times \mathbf{b})] \quad (l, m, n \in \mathbb{Z})$$

- **Bloch's theorem**

band index

$$\psi_{n\mathbf{k}}(\mathbf{r}) = \exp(i\mathbf{k} \bullet \mathbf{r})\, u_{n,\mathbf{k}}(\mathbf{r})$$

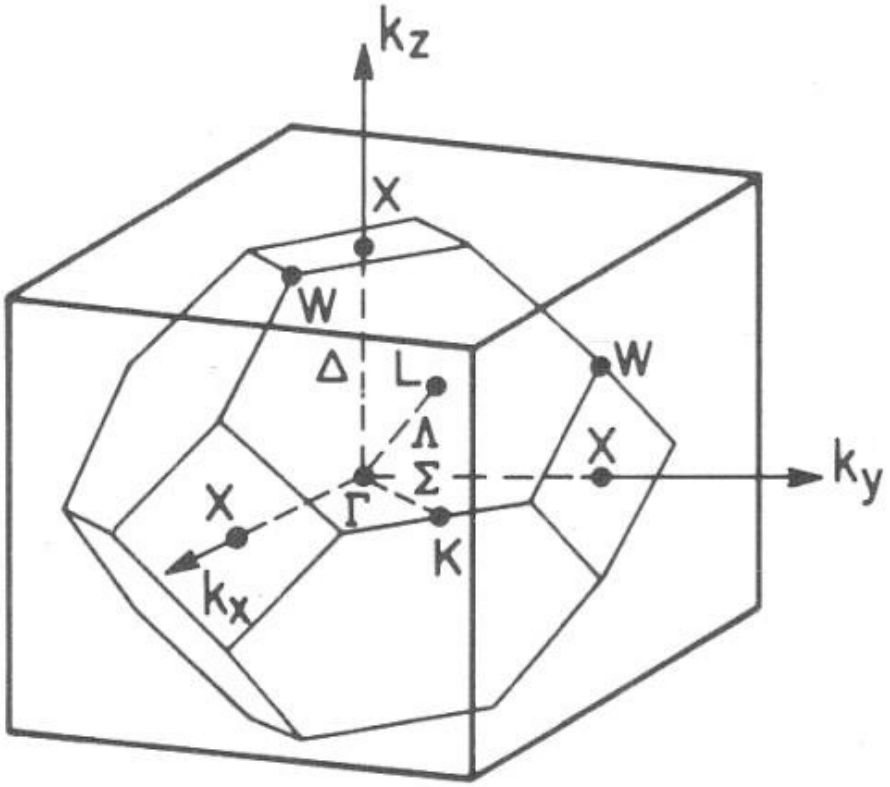
$$= \sum_{\mathbf{G}} u_{n,\mathbf{k}}(\mathbf{G}) \exp(i(\mathbf{k} + \mathbf{G}) \bullet \mathbf{r})$$

$\mathbf{k} \in$ first Brillouin zone in the reciprocal space

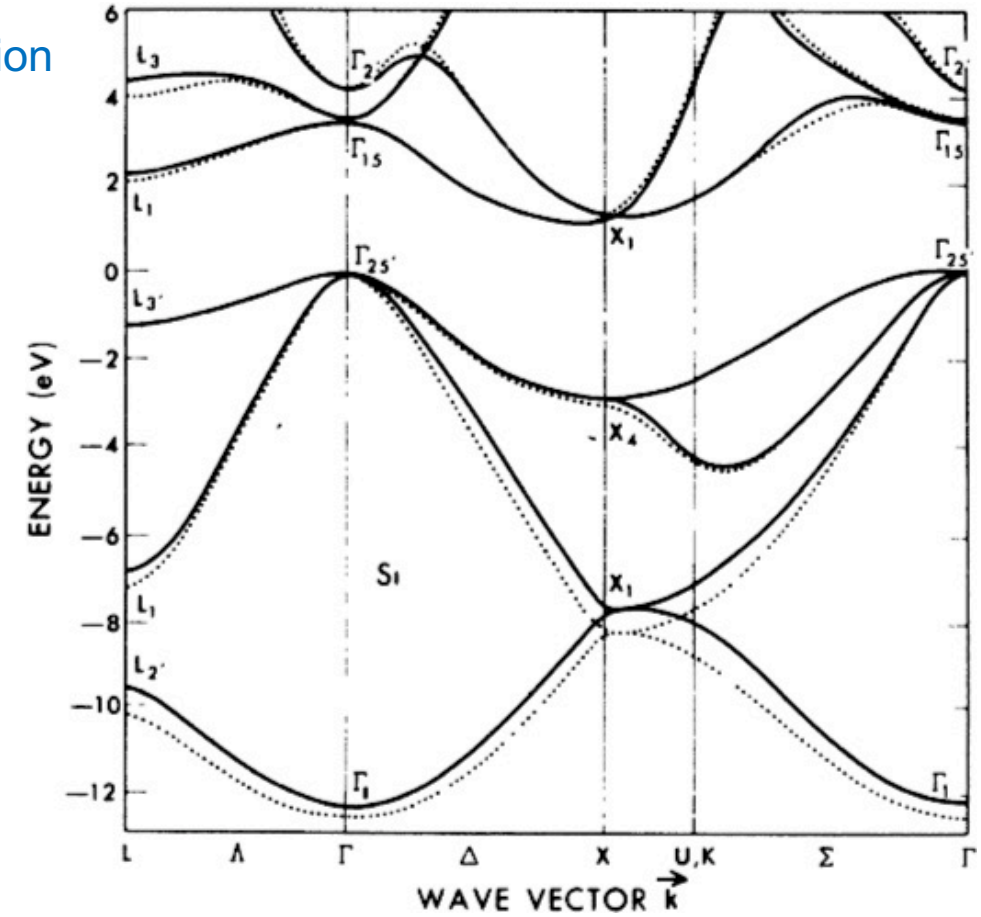
Electronic Bands: Infinite Lattice

- **Bloch theorem:** $\psi_{n\mathbf{k}}(\mathbf{r}) = \exp(i\mathbf{k} \bullet \mathbf{r}) u_{n,\mathbf{k}}(\mathbf{r})$

band index periodic function



Brillouin zone of Si crystal



Kohn-Sham energy

J. R. Chelikowsky & M. L. Cohen, *Phys. Rev. B* **10**, 5095 ('74)

Self-Consistent Field Iteration

$$\left(-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial \mathbf{r}^2} + \hat{V}_{\text{ion}} + \hat{V}_{\text{H,xc}}[\rho(\mathbf{r})]\right)\psi_n(\mathbf{r}) = \epsilon_n \psi_n(\mathbf{r})$$

Given $\rho(\mathbf{r})$, iteratively obtain $\{\psi_n, \epsilon_n\}$, *e.g.*, by preconditioned conjugate gradient

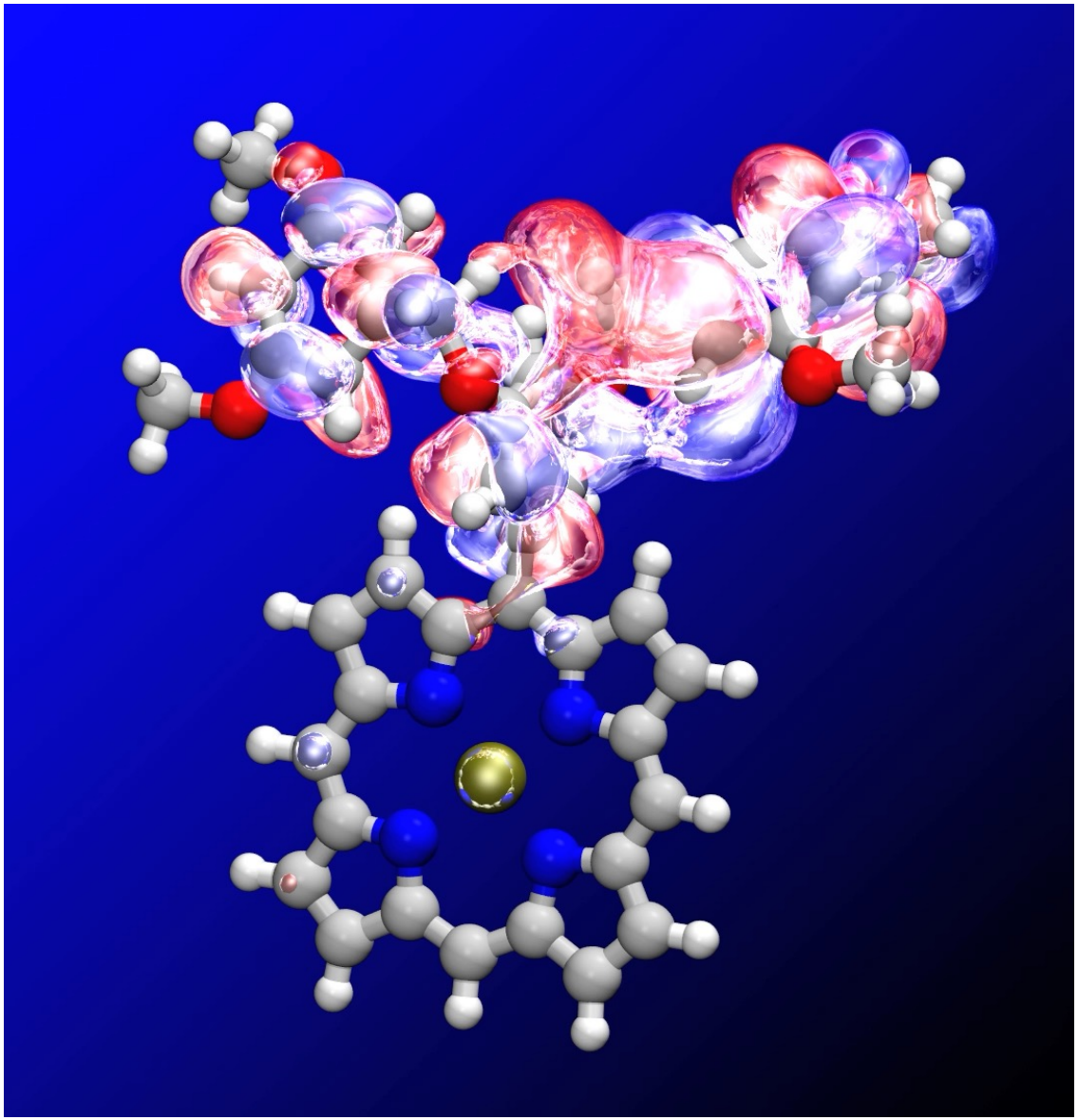
Given $\{\psi_n, \epsilon_n\}$, determine μ and compute $\rho(\mathbf{r})$

$$\rho(\mathbf{r}) = \sum_n |\psi_n(\mathbf{r})|^2 \, \Theta(\mu - \epsilon_n)$$

Chemical potential

$$N = \int d\mathbf{r}\rho(\mathbf{r})$$

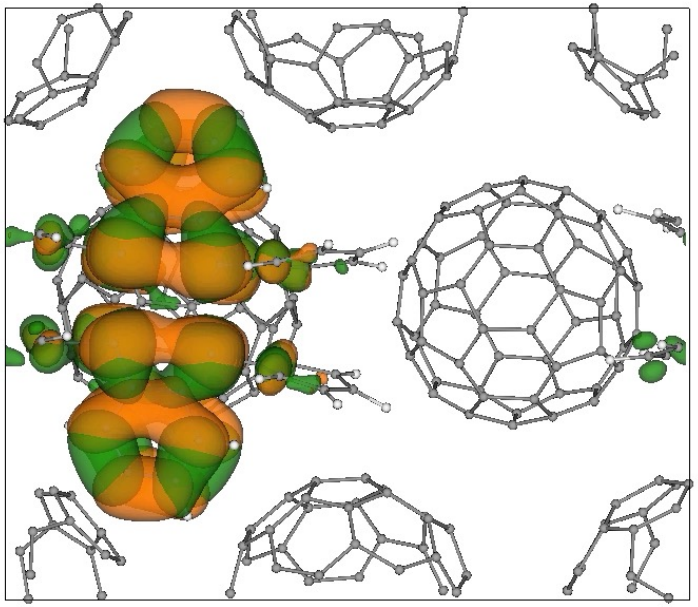
Nonadiabatic Quantum Molecular Dynamics



Appl. Phys. Lett. **98**, 113302 (’11); *ibid*. **100**, 203306 (’12); *ibid*. **102**, 173301 (’13); *Comput. Phys. Commun*. **184**, 1 (’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); *Sci Adv*. **8**, eabk2625 (‘22); *ibid*. **10**, eadp1890 (’24)

Zn porphyrin

Rubrene/C_{60}



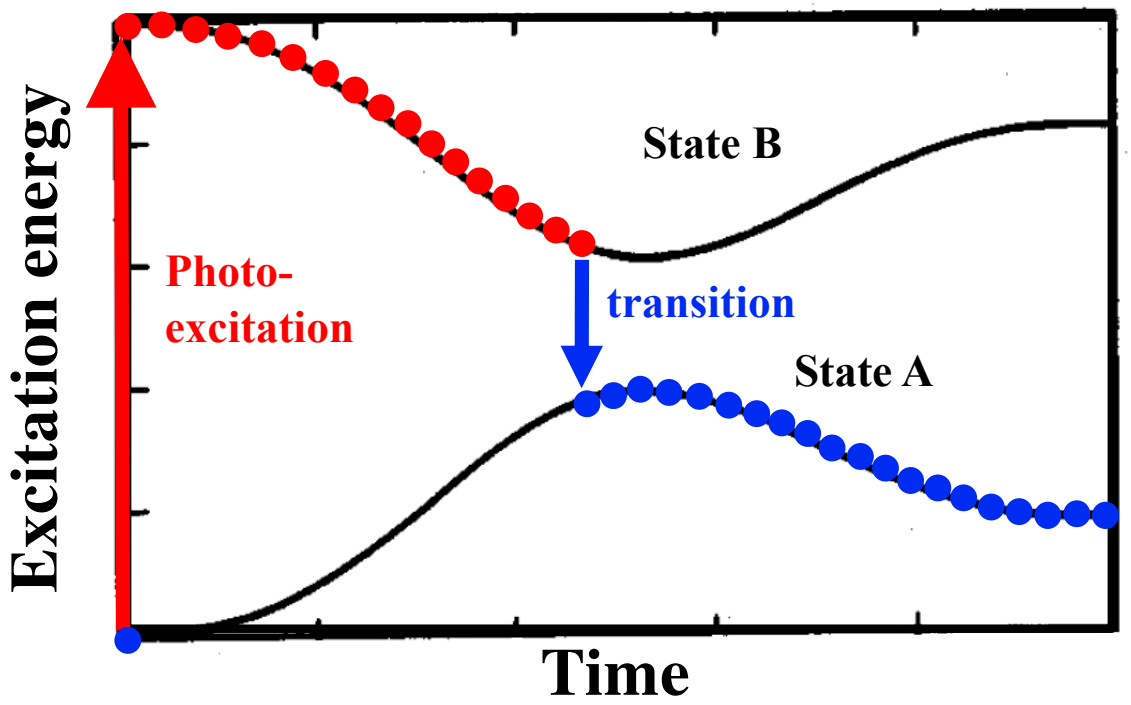
quasi-electron; quasi-hole

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

Surface-Hopping NAQMD

- **Incorporate electron transitions with the time-dependent density-functional theory (TDDFT) & surface-hopping method**

Tully, *J. Chem. Phys*. **93**, 1061 ('90), *ibid*. **129**, 044104 ('08) ; Duncan *et al*., *J. Am. Chem. Soc*. **129**, 8528 ('07)



- **Electronic transitions from the current state to another occur stochastically based on the switching probability obtained by solving TDDFT equations**

K-th excitation frequency

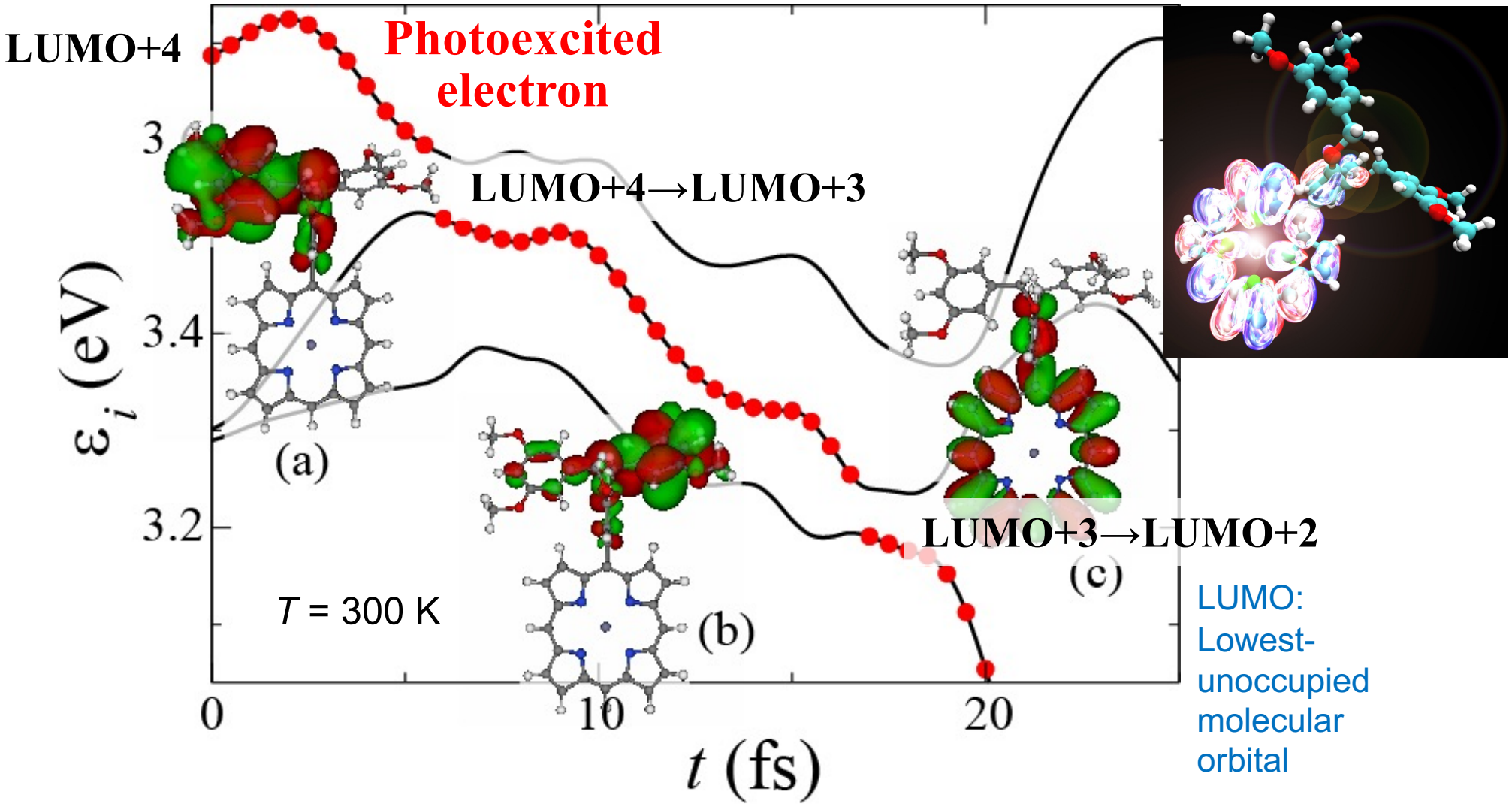
$$\Psi(\mathbf{r},t) = \sum_J C_J^{(I)}(t)\Phi_J(\mathbf{r};\mathbf{R}(t)) \qquad C_I^{(I)}(0) = \delta_{I,J}$$

$$\frac{d}{dt}C_J^{(I)}(t) = -\sum_k C_k^{(I)}(t)\left(i\omega_K\delta_{JK} + \left\langle \Phi_J \left| \frac{\partial}{\partial t} \right| \Phi_K \right\rangle\right)$$

J-th adiabatic excited state

Electronic transition assisted by nuclei motion

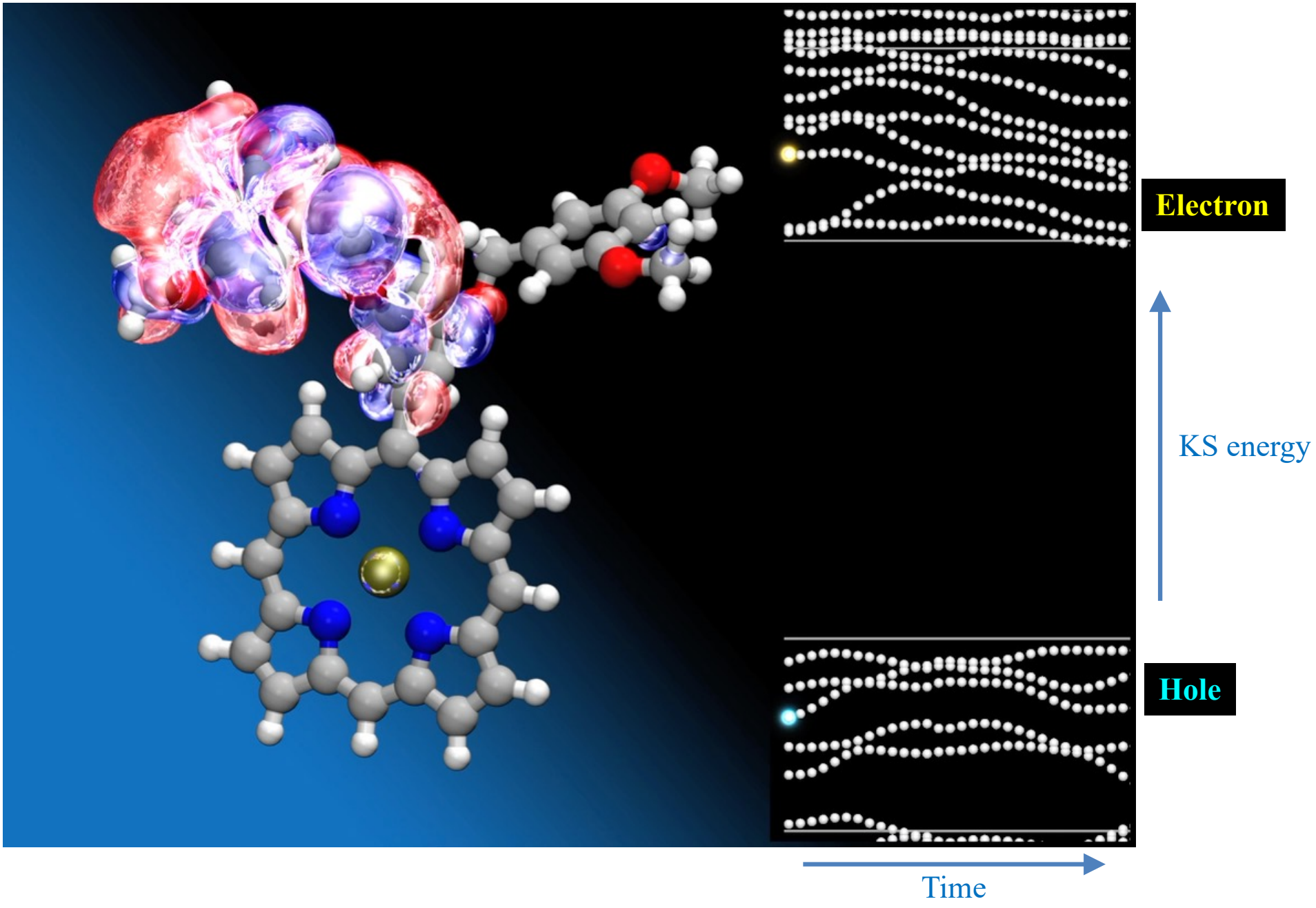
Example: Electron Transfer in a Dendrimer



LUMO: Lowest-unoccupied molecular orbital

- **The photoexcited electron at the peripheral antenna is transferred to the core due to the energy-crossing & overlapping of orbitals assisted by thermal molecular motions**

Surface-Hopping in Action



Excitonic Effects: LR-TDDFT

- **Excited electron-hole pairs within the linear-response time-dependent density functional theory (LR-TDDFT)** [Casida, '95]

$$\delta V(t) = \delta v_{kl\tau}(t)\hat{a}^{+}_{k\tau}\hat{a}_{l\tau} \longrightarrow \delta P_{ij\sigma}(t) = \delta\langle\Phi(t)|\hat{a}^{+}_{i\sigma}\hat{a}_{j\sigma}|\Phi(t)\rangle$$

electron hole

$$\chi_{ij\sigma,kl\tau}(t-t') = \delta P_{ij\sigma}(t)/\delta v_{kl\tau}(t')$$

- **Excitation energies from the poles of the response function $\chi_{ij\tau,kl\sigma}(\omega)$**

$2N_{\text{unoccupied}}N_{\text{occupied}} \times 2N_{\text{unoccupied}}N_{\text{occupied}}$ matrix eigenequation

$$\begin{pmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B}^{*} & \mathbf{A}^{*} \end{pmatrix}\begin{pmatrix} \mathbf{X}_I \\ \mathbf{Y}_I \end{pmatrix} = \hbar\omega_I \begin{pmatrix} \mathbf{1} & \mathbf{0} \\ \mathbf{0} & -\mathbf{1} \end{pmatrix}\begin{pmatrix} \mathbf{X}_I \\ \mathbf{Y}_I \end{pmatrix}$$

I-th excitation energy

Kohn-Sham energy

$$A_{ia\sigma,jb\tau} = \delta_{\sigma,\tau}\delta_{i,j}\delta_{a,b}(\varepsilon_{a\sigma} - \varepsilon_{i\sigma}) + K_{ia\sigma,jb\tau} \qquad B_{ia\sigma,jb\tau} = K_{ia\sigma,bj\tau}$$

$$K_{ia\sigma,i'a'\sigma'} = \iint \psi^{*}_{i\sigma}(\mathbf{r})\psi_{a\sigma}(\mathbf{r})\left(\frac{e^2}{|\mathbf{r}-\mathbf{r}'|} + \frac{\delta^2 E_{\text{xc}}}{\delta\rho_{\sigma}(\mathbf{r})\delta\rho_{\sigma'}(\mathbf{r}')}\right)\psi_{i'\sigma'}(\mathbf{r}')\psi^{*}_{a'\sigma'}(\mathbf{r}')\,d\mathbf{r}\,d\mathbf{r}'$$

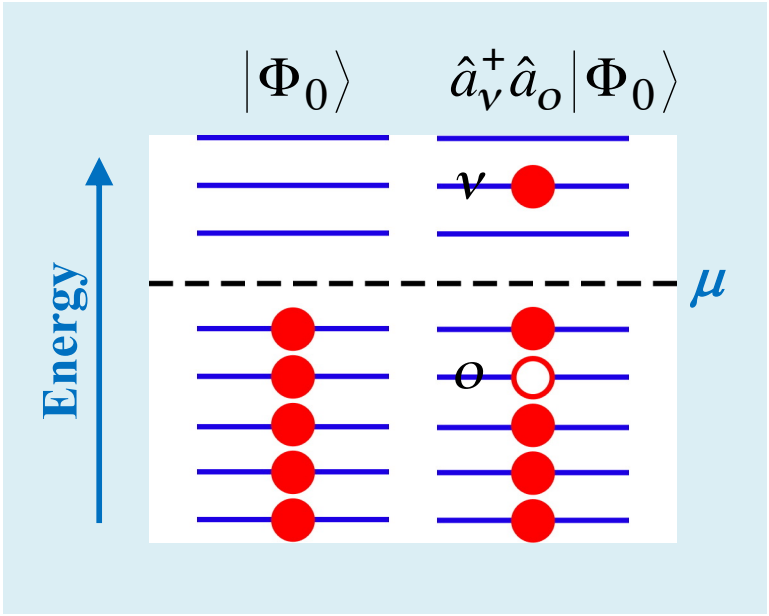
Coulomb & exchange-correlation interaction matrix elements

Electronic Excited States

- ***I*-th excited state**

$$\left|\Phi_I(\mathbf{r};\mathbf{R})\right\rangle = \sum_{i\in\{\text{occupied}\}} \sum_{a\in\{\text{unoccupied}\}} \sum_{\sigma} \sqrt{\frac{\varepsilon_{a\sigma}-\varepsilon_{i\sigma}}{\hbar\omega_I}} \left(X_{I,ia\sigma}+Y_{I,ia\sigma}\right) \hat{a}^{+}_{a\sigma}\hat{a}_{i\sigma} \left|\Phi_0(\mathbf{r};\mathbf{R})\right\rangle$$

electron-hole pair ground state



QXMD Code

- **Quantum molecular dynamics (QMD) code developed by Prof. Fuyuki Shimojo at Kumamoto University in Japan**
- **Various eXtensions co-developed with USC-CACS: Nonadiabatic QMD, linear-scaling divide-&-conquer, parallelization, *etc*.**
- **Unique features:**
  - > **Interatomic forces with electronic excitation to study photo-excited lattice dynamics**
    Shimojo *et al*., *Comput. Phys. Commun*. **184**, 1 ('13)
  - > **Range-separated hybrid exact-exchange functional for exciton binding**
    Tawada *et al*., *J. Chem. Phys*. **120**, 8425 ('04)
  - > **Lean divide-&-conquer density functional theory (LDF-DFT) with small *O*(*N*) prefactor**
    Shimojo *et al*., *J. Chem. Phys*. **140**, 18A529 ('14)
  - > **Omni-directional multiscale shock technique (OD-MSST)**
    Shimamura *et al*., *Appl. Phys. Lett*. **107**, 231903 ('15); **108**, 071901 ('16)
  - > **Berry-phase computation of bulk polarization**
- **Other features:**
  - > **Various functionals: spin-polarized, GGA+U, DFT+D, nonlocal correlation**
  - > **Nudged elastic band (NEB) method for energy-barrier calculation**

GitHub repository:
https://github.com/USCCACS/QXMD_DEV

Software download site:
https://cybermagics.netlify.app/software

Current & Future Supercomputing

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

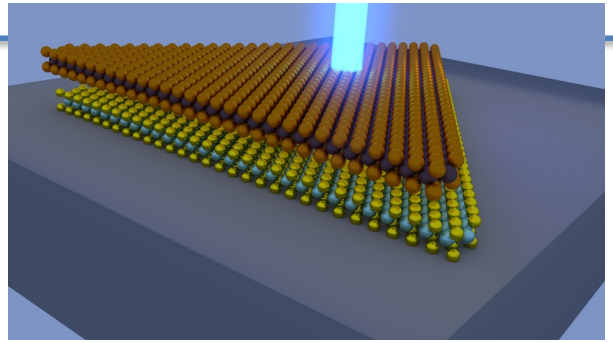
U.S. DEPARTMENT OF ENERGY
INCITE
LEADERSHIP COMPUTING
Innovative & Novel Computational Impact on Theory & Experiment

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

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

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

AURORA | EARLY SCIENCE PROGRAM



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

1.01 exaflop/s
Intel Aurora

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

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

But...

TOP 500 The List.

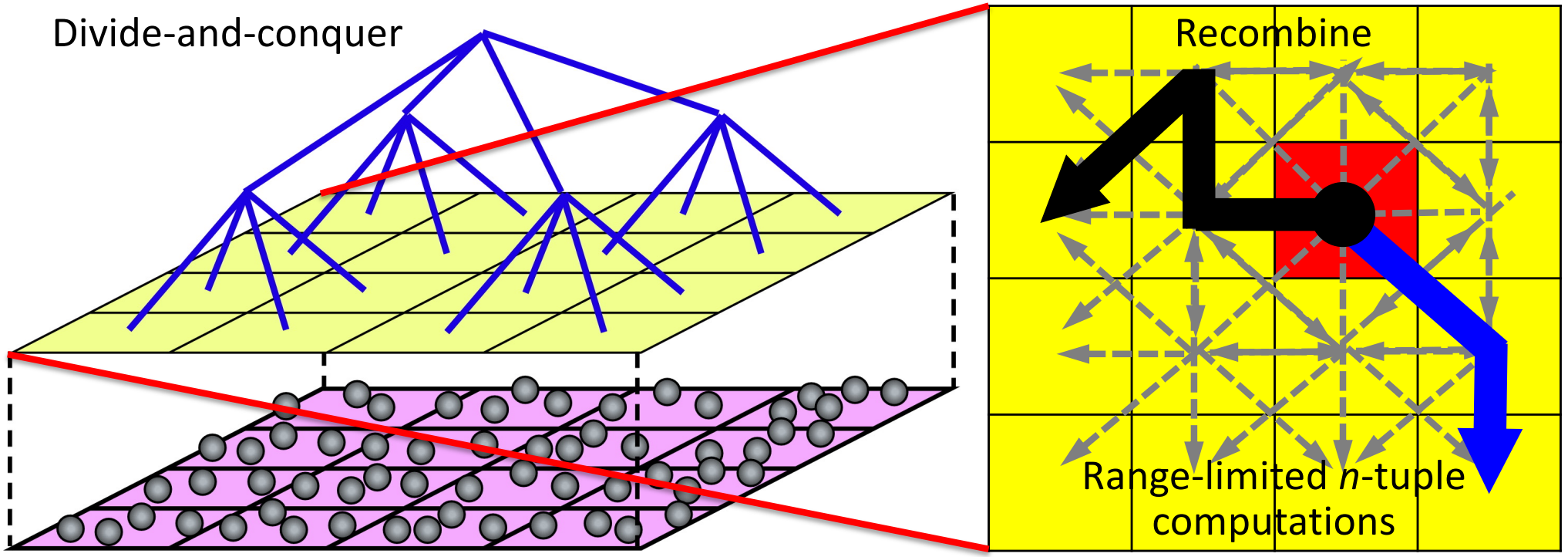
Intel Dumps Knights Hill, Future of Xeon Phi Product Line Uncertain

Michael Feldman | November 15, 2017 04:34 CET

https://www.top500.org/news/

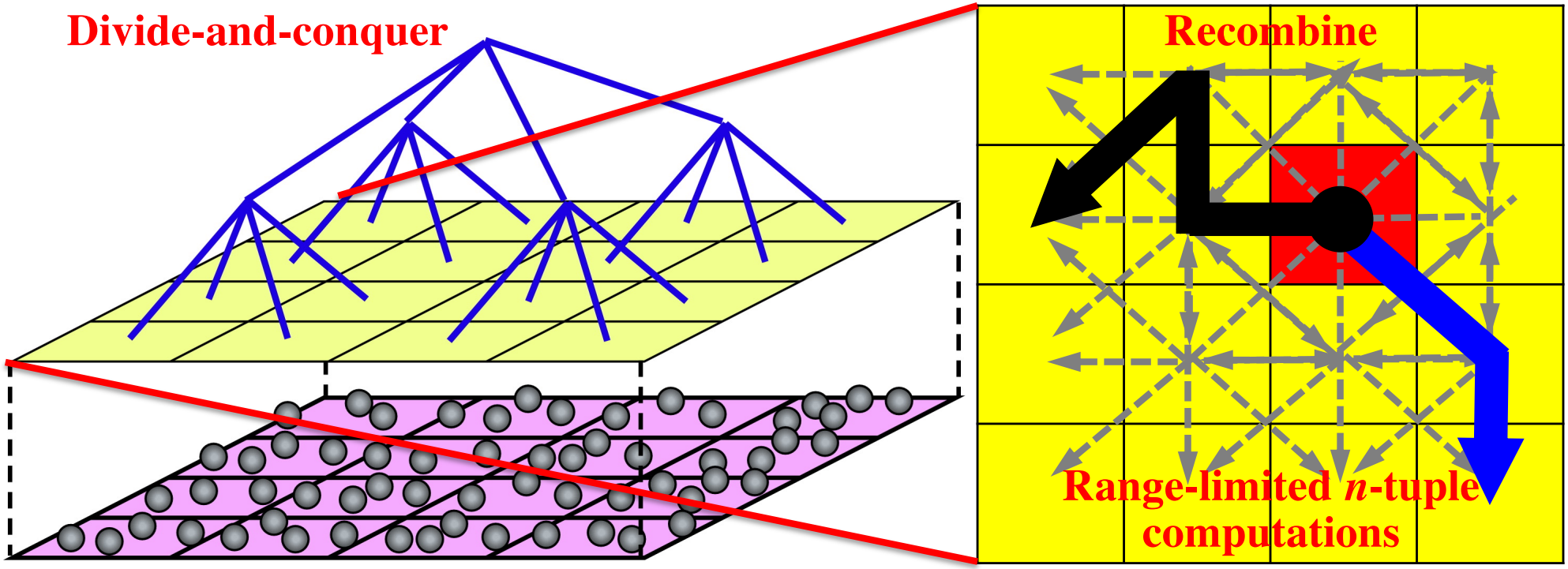
- **Need *metascalable* (or "design once, scale on new architectures") parallel applications**
- **Proposed *divide-conquer-recombine***

F. Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14);
K. Nomura *et al.*, *ACM/IEEE SC14* ('14)



M. Kunaseth *et al.*, *ACM/IEEE SC13* ('13)

Divide-Conquer-Recombine (DCR) Engines



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

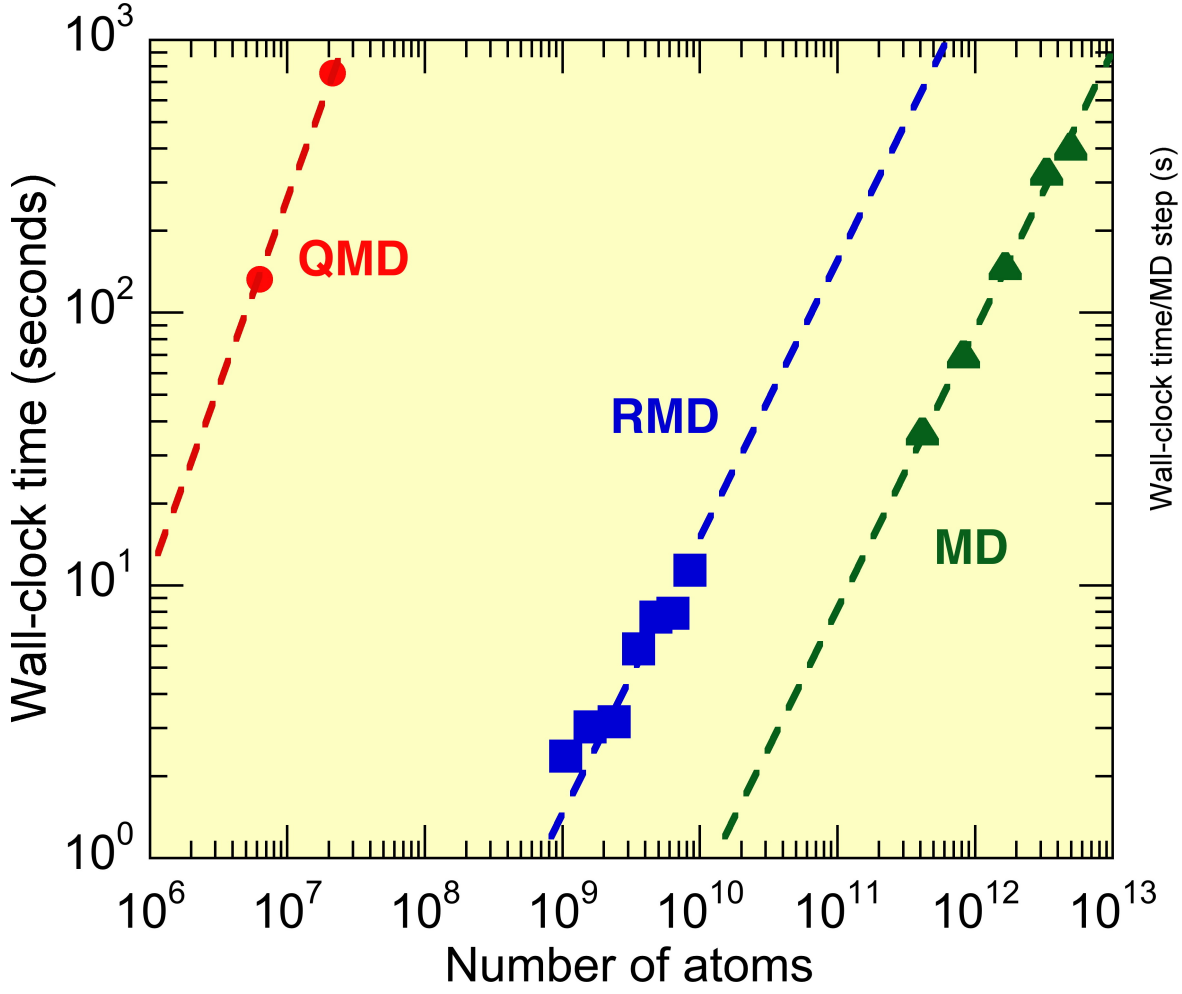
- **Quantum MD: Lean divide-&-conquer density functional theory (LDC-DFT) algorithm minimizes the prefactor of $O(N)$ computational cost**

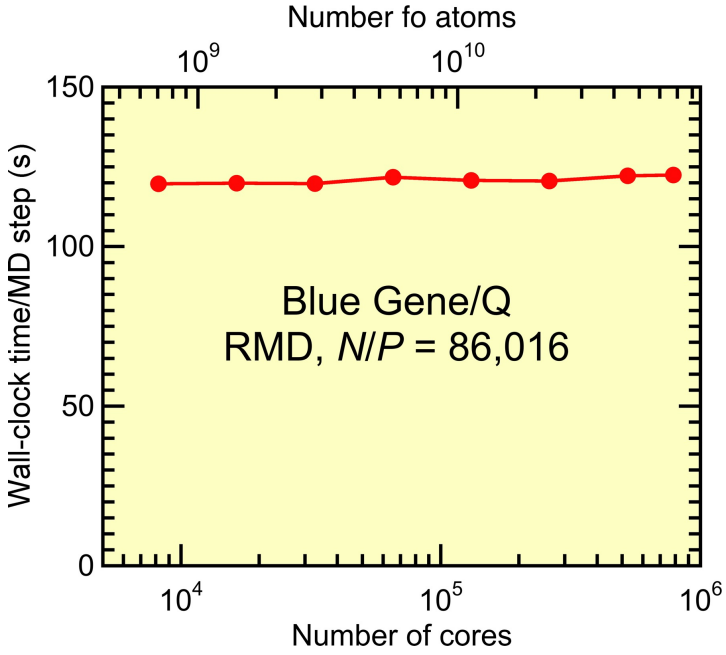
  F. Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14); K. Nomura *et al.*, *IEEE/ACM SC14*

- **Reactive MD: Extended-Lagrangian reactive molecular dynamics (XRMD) algorithm eliminates the speed-limiting charge iteration**

  K. Nomura *et al.*, *Comput. Phys. Commun.* **192**, 91 ('15)

Scalable Simulation Algorithm Suite





QMD (quantum molecular dynamics): DC-DFT

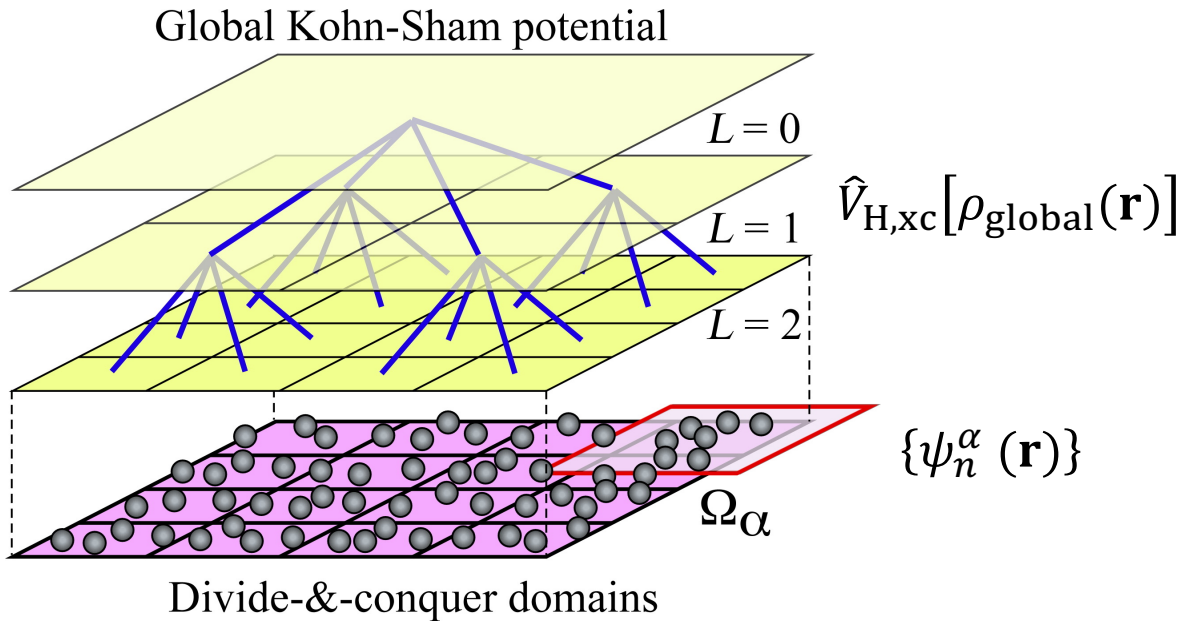
RMD (reactive molecular dynamics): F-ReaxFF

MD (molecular dynamics): MRMD

- **4.9 trillion-atom space-time multiresolution MD (MRMD) of SiO_2**
- **67.6 billion-atom fast reactive force-field (F-ReaxFF) RMD of RDX**
- **39.8 trillion grid points (50.3 million-atom) DC-DFT QMD of SiC**

parallel efficiency 0.984 on 786,432 Blue Gene/Q cores

Divide-&-Conquer Density Functional Theory



- **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

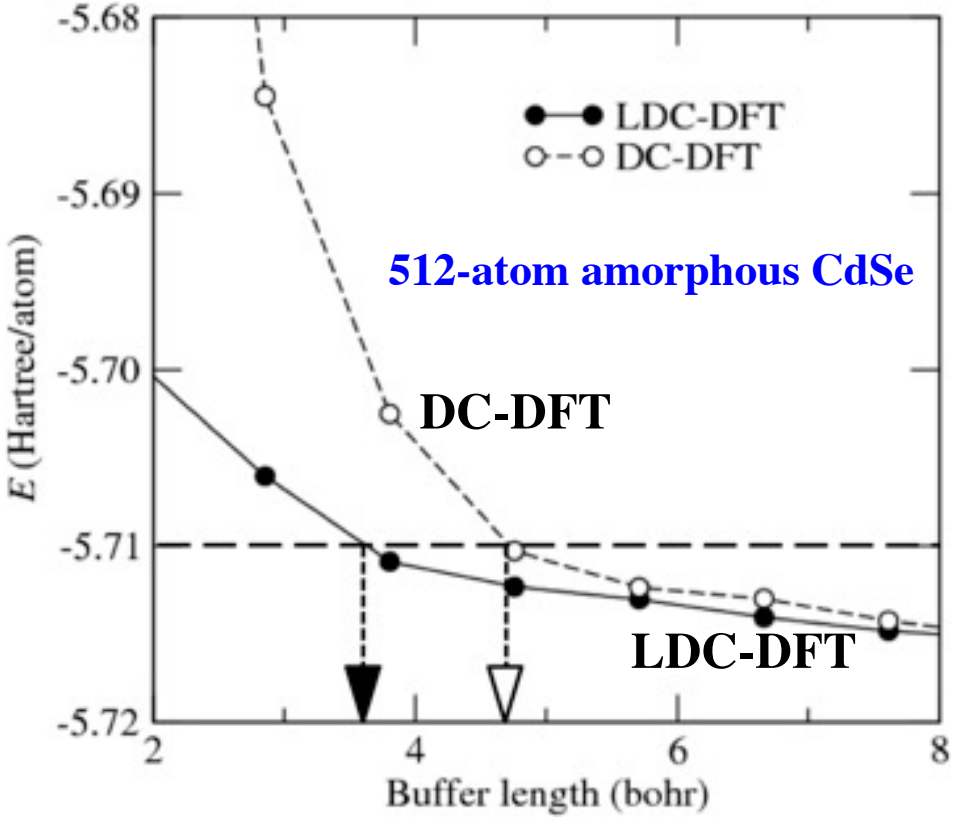
$$N = \int d\mathbf{r}\, \rho_{\mathrm{global}}(\mathbf{r})$$

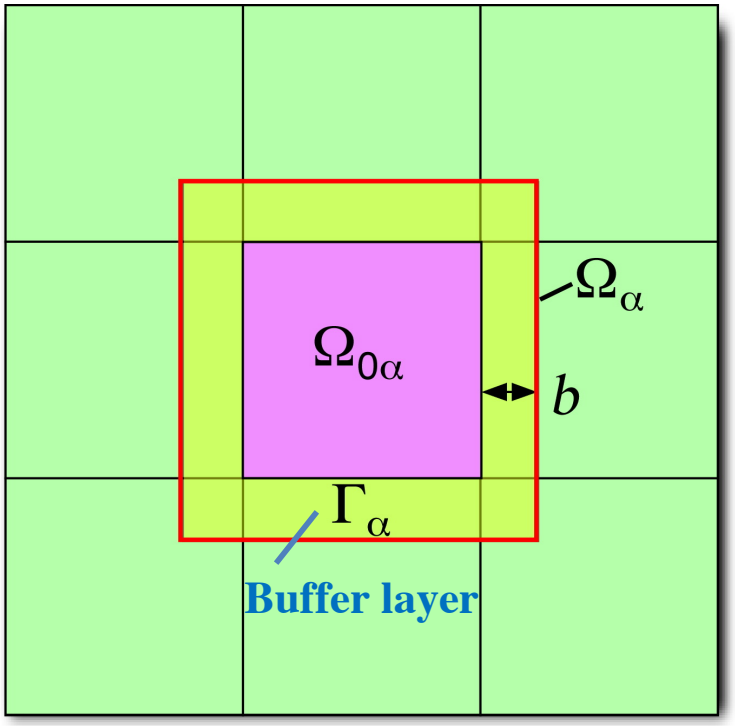
Lean Divide-&-Conquer (LDC) DFT

- **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**



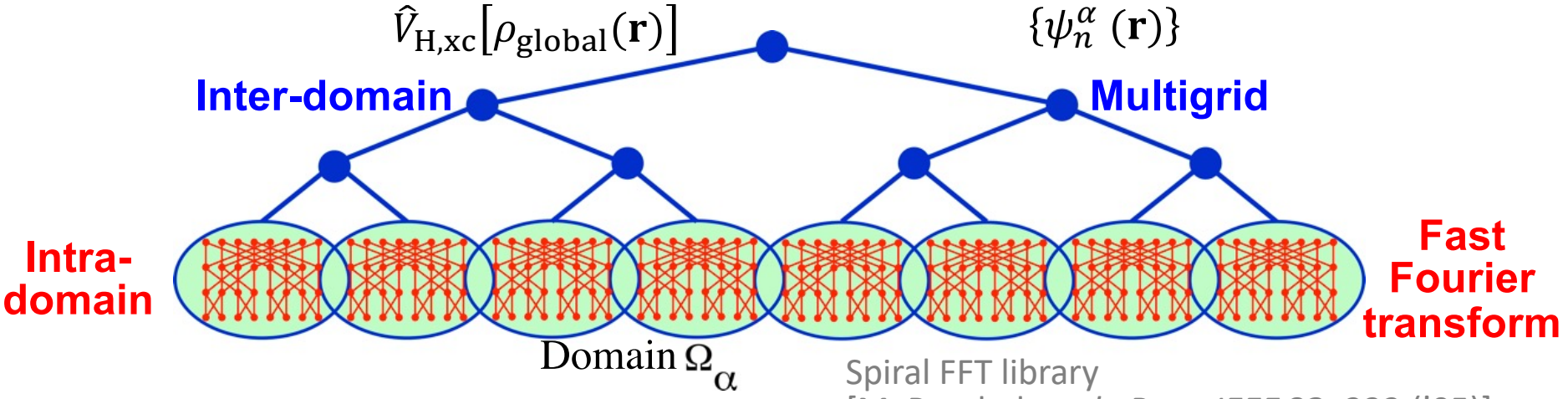


- **Factor 2.03 (for ν = 2) ~ 2.89 (for ν = 3) reduction of the computational cost with an error tolerance of 5×10^{-3} a.u. (per-domain complexity: n^ν)**

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

Hierarchical Computing

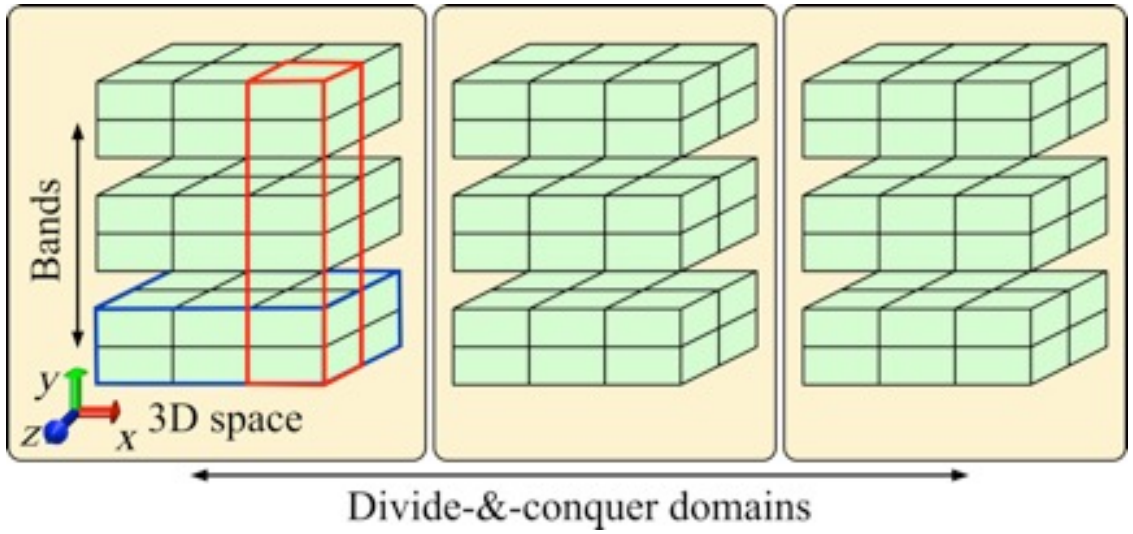
- **Globally scalable (real-space multigrid) + locally fast (plane wave) electronic solver**



Spiral FFT library
[M. Puschel *et al.*, *Proc. IEEE* **93**, 232 ('05)]

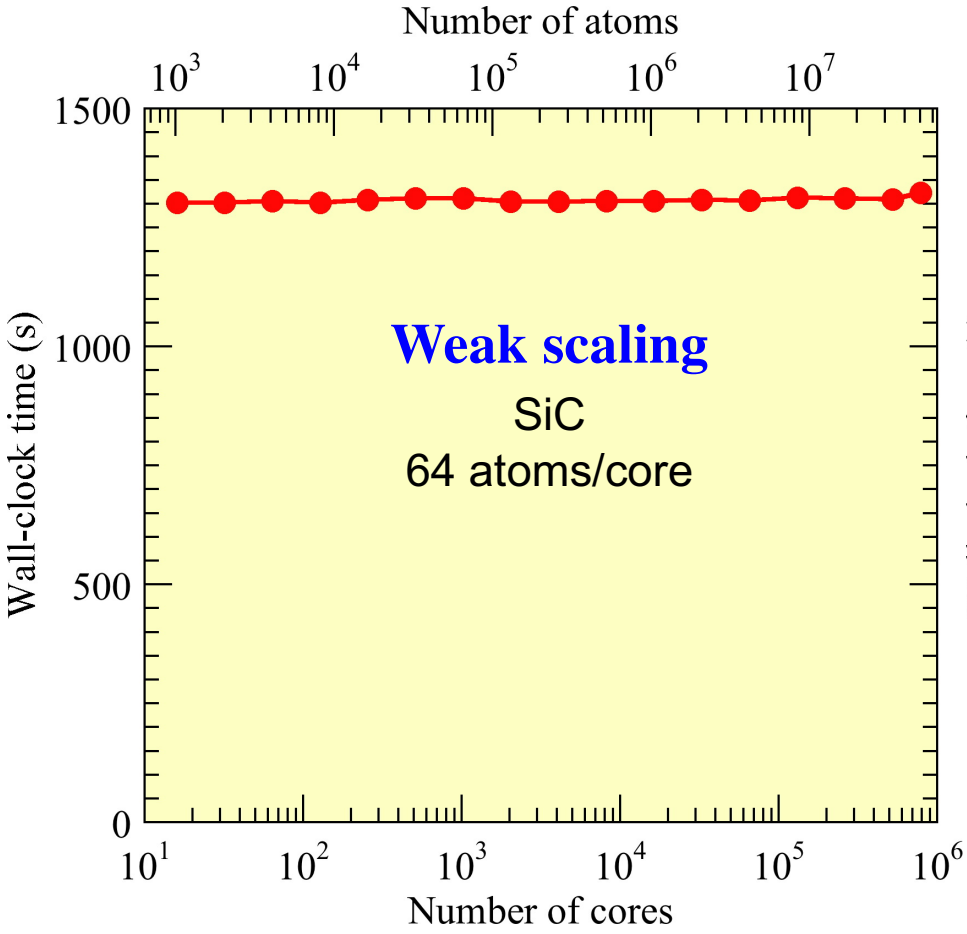
cf. globally- sparse-yet-locally-dense eigensolver [J. H. Lam *et al.*, *Nature Commun*. **15**, 3479 ('24)]

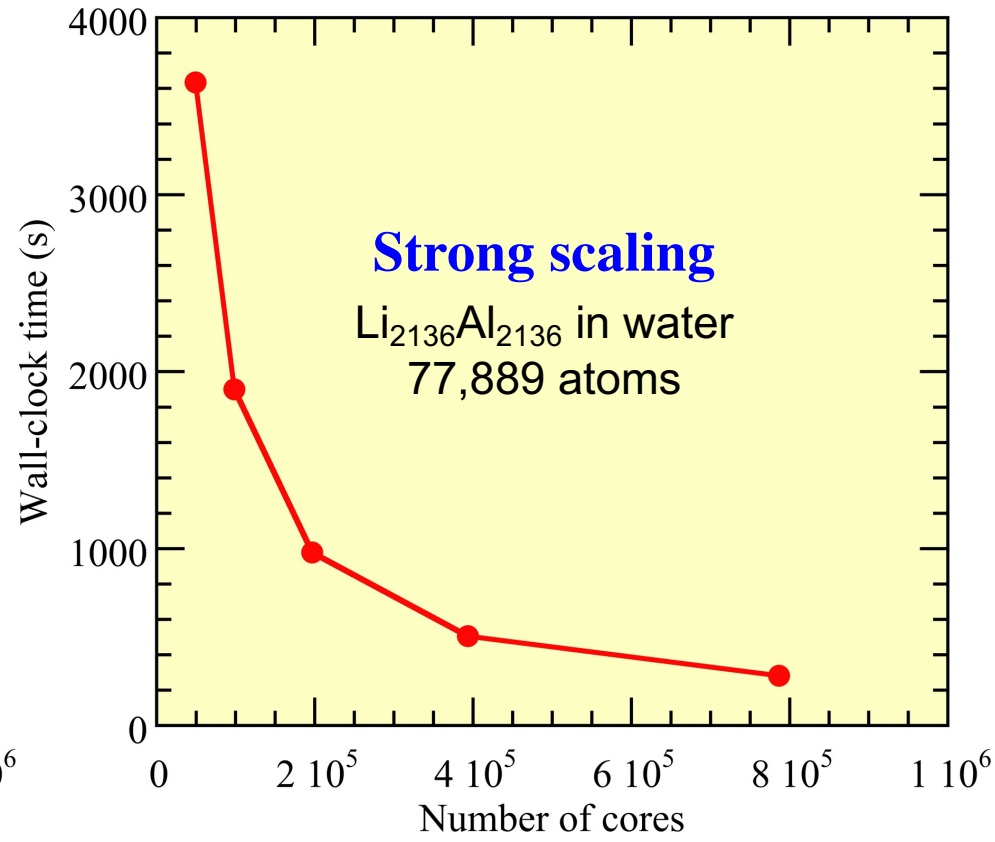
- **Hierarchical band (*i.e.*, Kohn-Sham orbital) + space + domain (BSD) decomposition**



Parallel Performance

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





- **62-fold reduction of time-to-solution** [441 s/SCF-step for 50.3M atoms] **from the previous state-of-the-art** [55 s/SCF-step for 102K atoms, Osei-Kuffuor *et al.*, *PRL* '14]

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

Floating Point Performance

- **Transform from band-by-band to all-band computations to utilize a matrix-matrix subroutine (DGEMM) in the level 3 basic linear algebra subprograms (BLAS3) library**
- **Algebraic transformation of computations**

Example: Nonlocal pseudopotential operation

D. Vanderbilt, *Phys. Rev. B* **41**, 7892 ('90)

$$\hat{v}_{\mathrm{nl}}|\psi_n^\alpha\rangle = \sum_I^{N_{\mathrm{atom}}} \sum_{ij}^{L_{\max}} |\beta_{i,I}\rangle D_{ij,I} \langle\beta_{j,I}|\psi_n^\alpha\rangle \quad (n = 1, \ldots, N_{\mathrm{band}})$$

$$\mathbf{\Psi} = \left[|\psi_1^\alpha\rangle, \ldots, |\psi_{N_{\mathrm{band}}}^\alpha\rangle\right] \widetilde{\mathbf{B}}(i) = \left[|\beta_{i,1}\rangle, \ldots, |\beta_{i,N_{\mathrm{atom}}}\rangle\right] \left[\widetilde{\mathbf{D}}(i,j)\right]_{I,J} = D_{ij,I}\delta_{IJ}$$

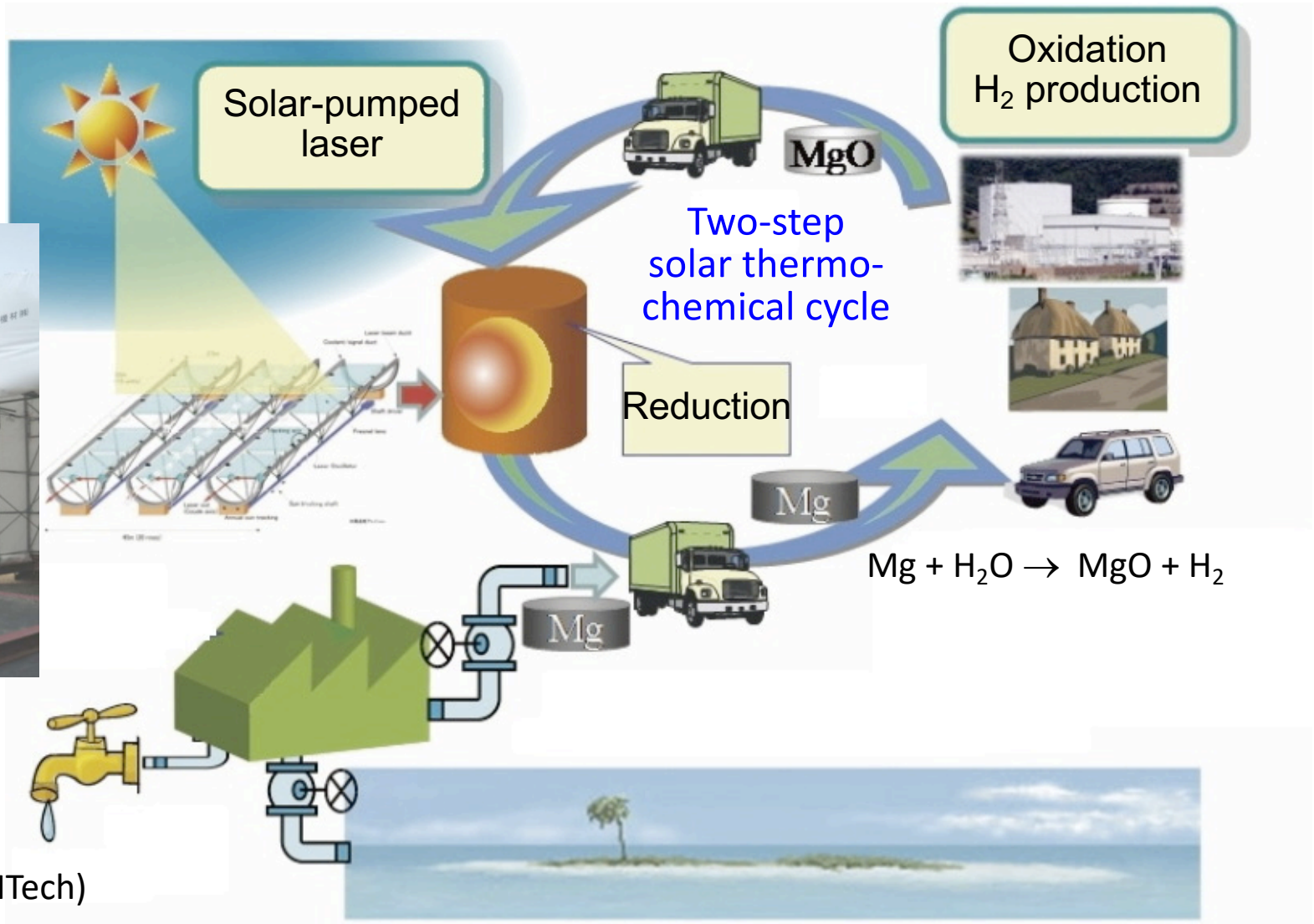
$$\hat{v}_{\mathrm{nl}}\mathbf{\Psi} = \sum_{i,j}^{L} \widetilde{\mathbf{B}}(i)\widetilde{\mathbf{D}}(i,j)\widetilde{\mathbf{B}}(j)^T$$

- **50.5% of the theoretical peak FLOP/s performance on 786,432 Blue Gene/Q cores (entire Mira at the Argonne Leadership Computing Facility)**
- **55% of the theoretical peak FLOP/s on Intel Xeon E5-2665**

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

S. C. Tiwari *et al.*, *HPC Asia* ('20) best paper

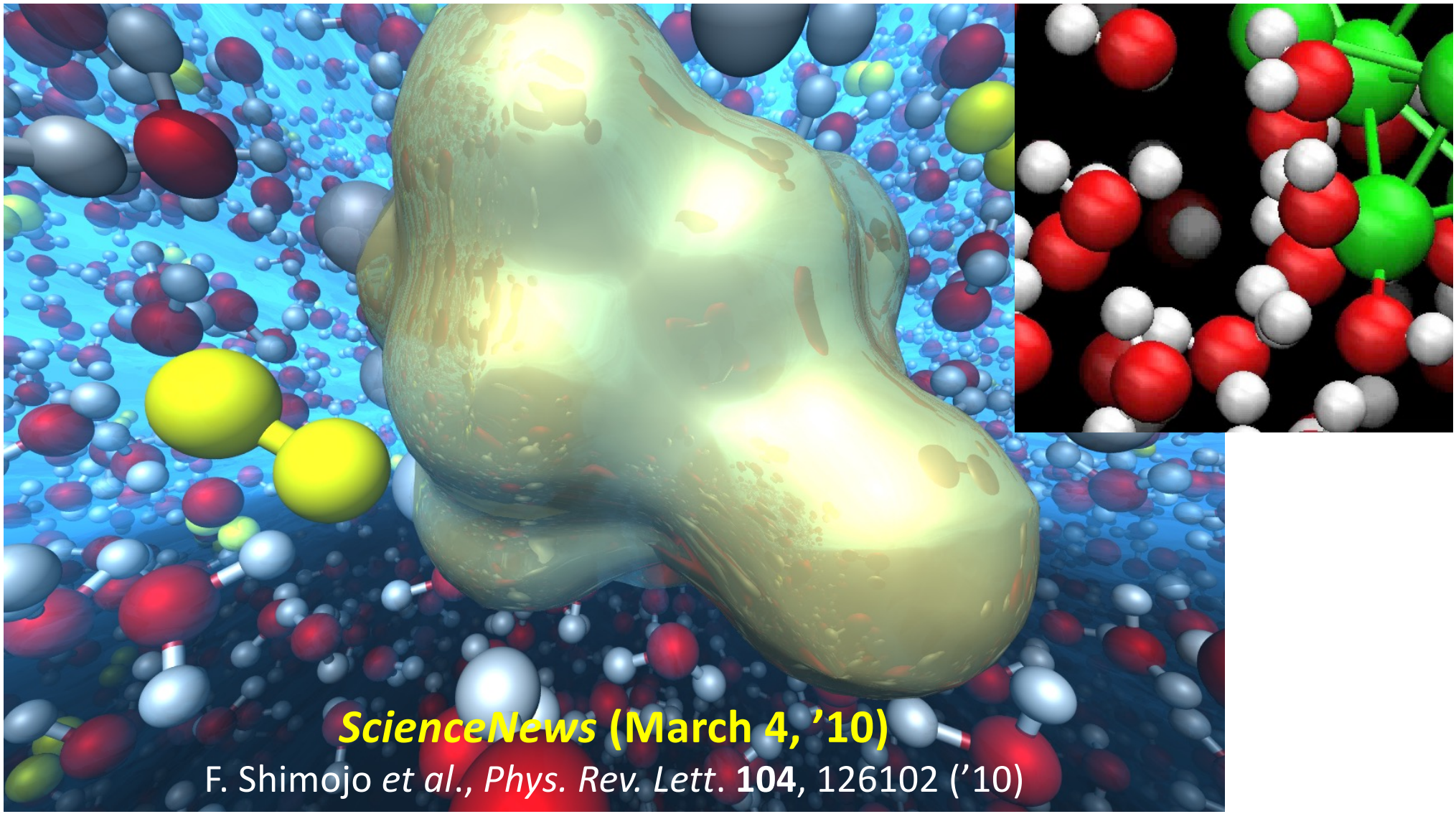
Renewal Energy Cycle by Metal Carriers



T. Yabe (TITech)

- **Problem: Accelerated hydrogen-production reaction kinetics for metal (Mg, Al, Zn, Fe) + water?**

Nanotechnology Solution



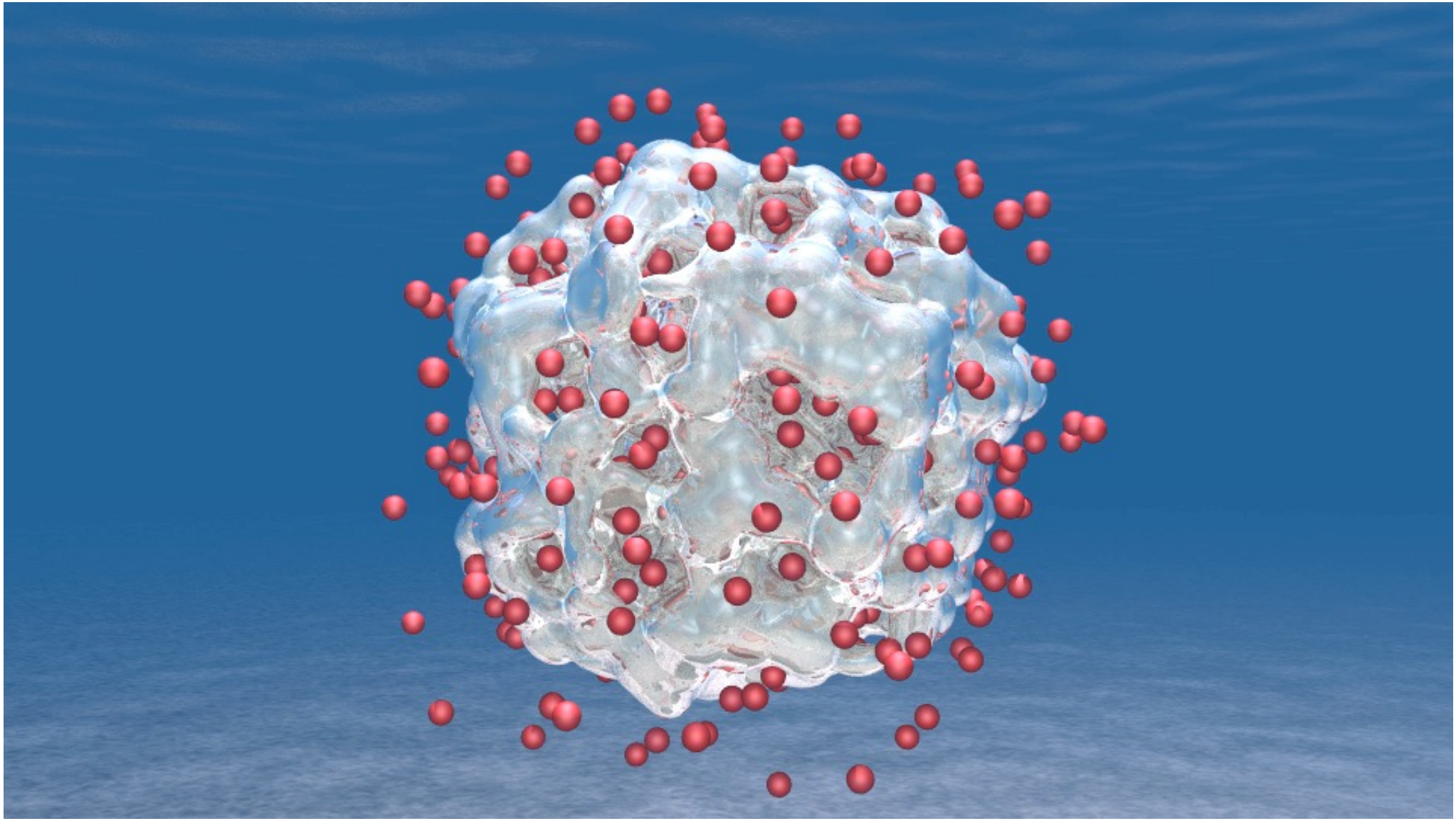
- **QMD simulation shows rapid H_2 production from water by a superatom* (Al_{17}), but the technology is not scalable to larger particle sizes**

*Roach, Castleman, Khanna *et al.*, *Science* **323**, 492 ('09)

H_2 Production from Water Using LiAl Particles

16,661-atom QMD simulation of $Li_{441}Al_{441}$ in water on 786,432 IBM Blue Gene/Q cores

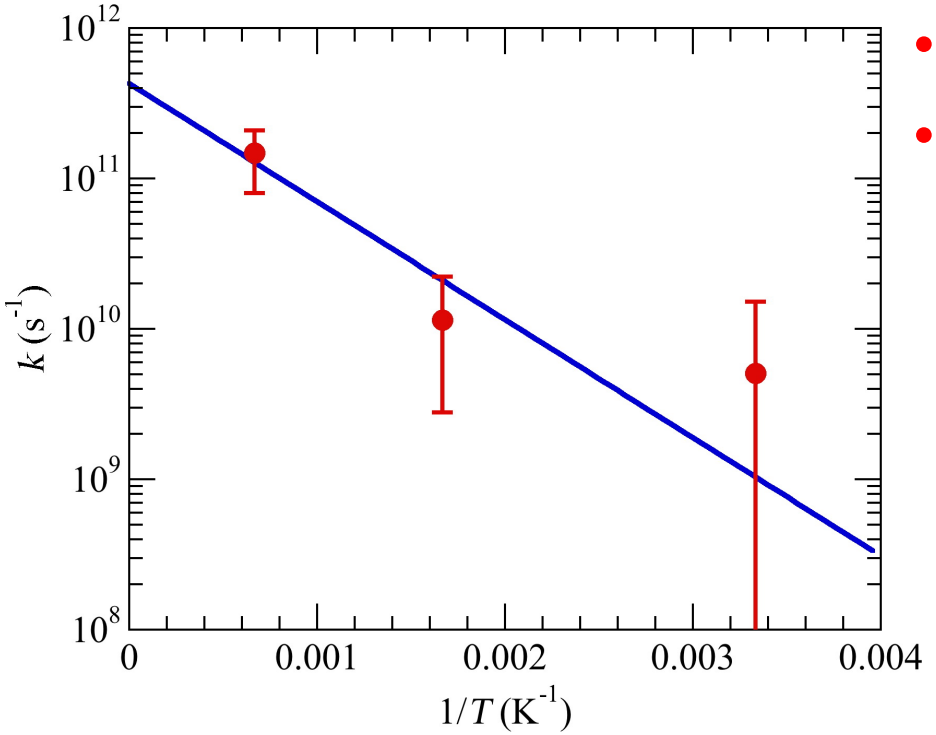
K. Shimamura *et al.*, *Nano Lett.* **14**, 4090 ('14)



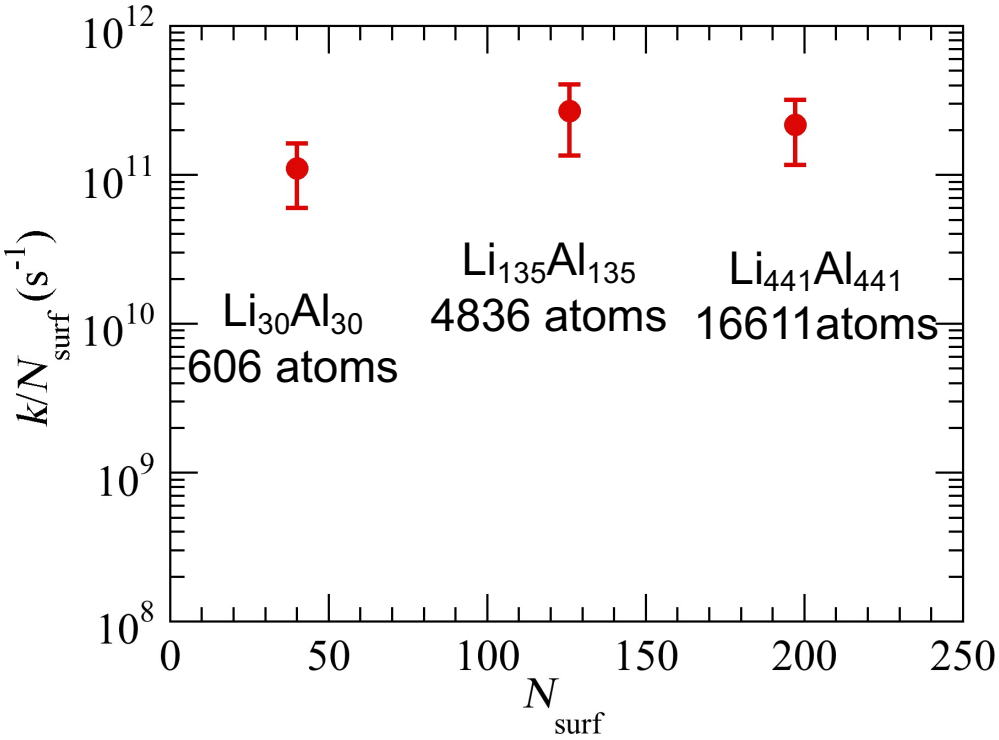
21,140 time steps (129,208 self-consistent-field iterations)

Rapid & Scalable H_2 Production

- **Orders-of-magnitude faster H_2 production from water than with pure Al**



- **Activation barrier = 0.068 eV**
- **Reaction rate = 1.04×10^{9} (s^{-1}) per LiAl pair at 300 K**

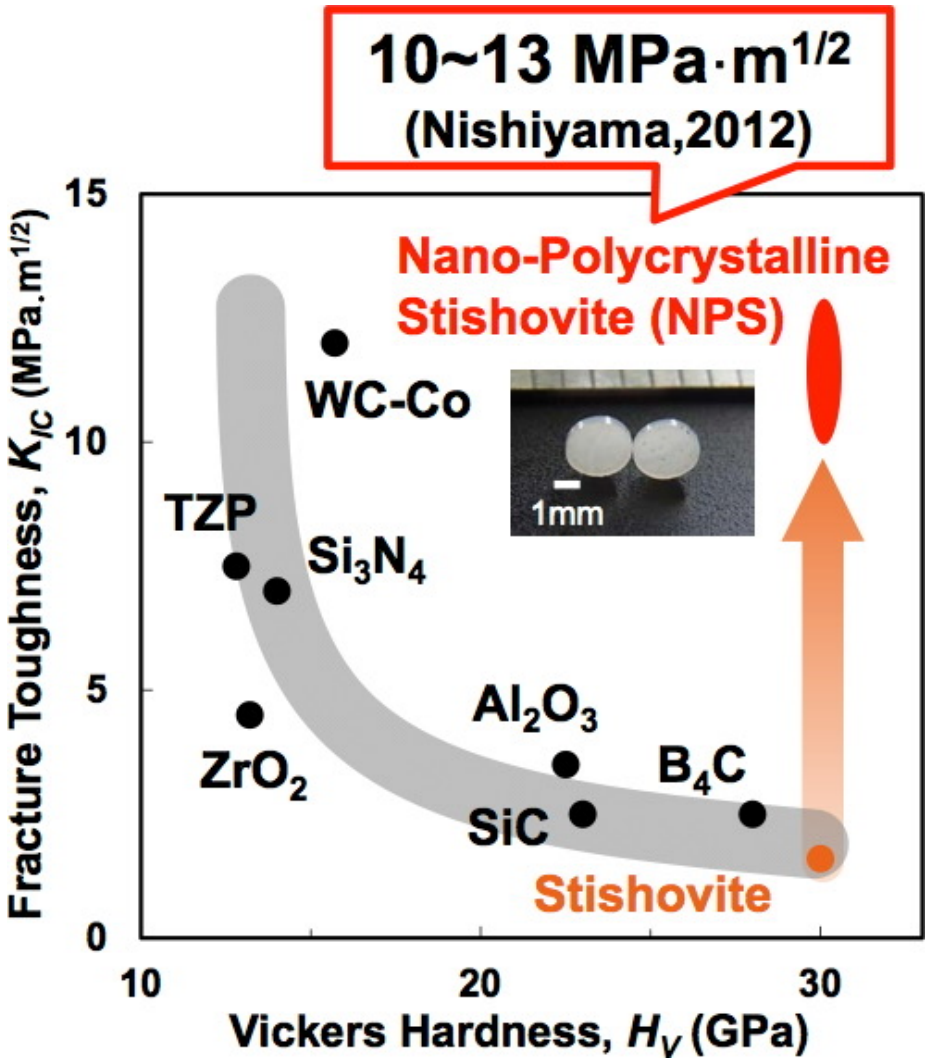


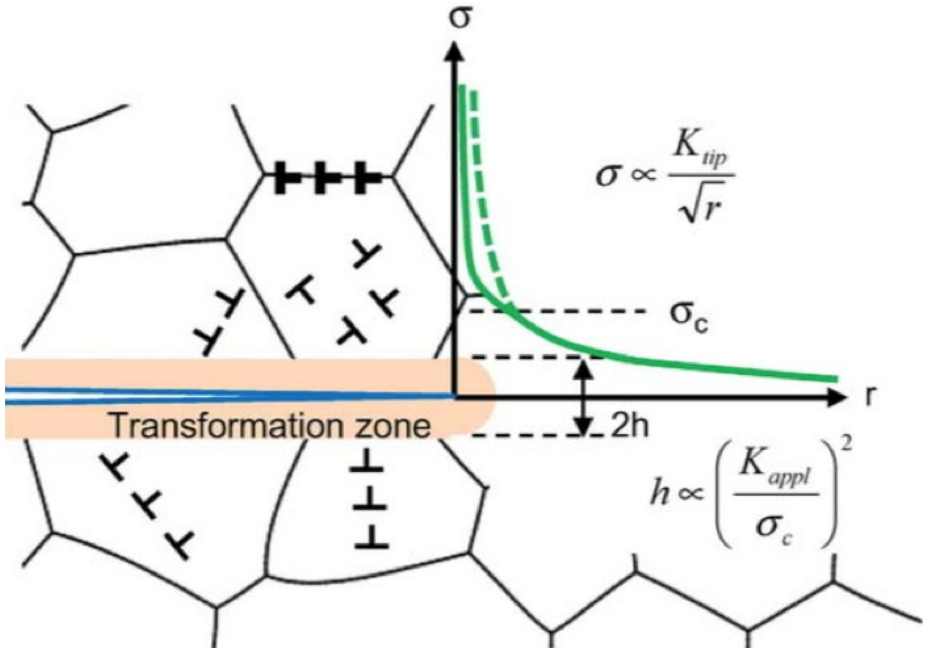
- **Reaction rate does not decrease for larger particles → industrial scalability**

K. Shimamura *et al.*, *Nano Lett.* **14**, 4090 ('14); K. Nomura *et al.*, *IEEE/ACM SC14* ('14)

Crack Self-Healing Stishovite

- **Superhard, ultratough nano-polycrystalline stishovite (NPS) synthesized**
  N. Nishiyama *et al.*, *Scripta Mater.* **67**, 955 ('12); *Sci. Rep*. **4**, 6588 ('14)
- **Made of Earth-abundant silica glass, NPS provides sustainable supply of high-performance ceramics**



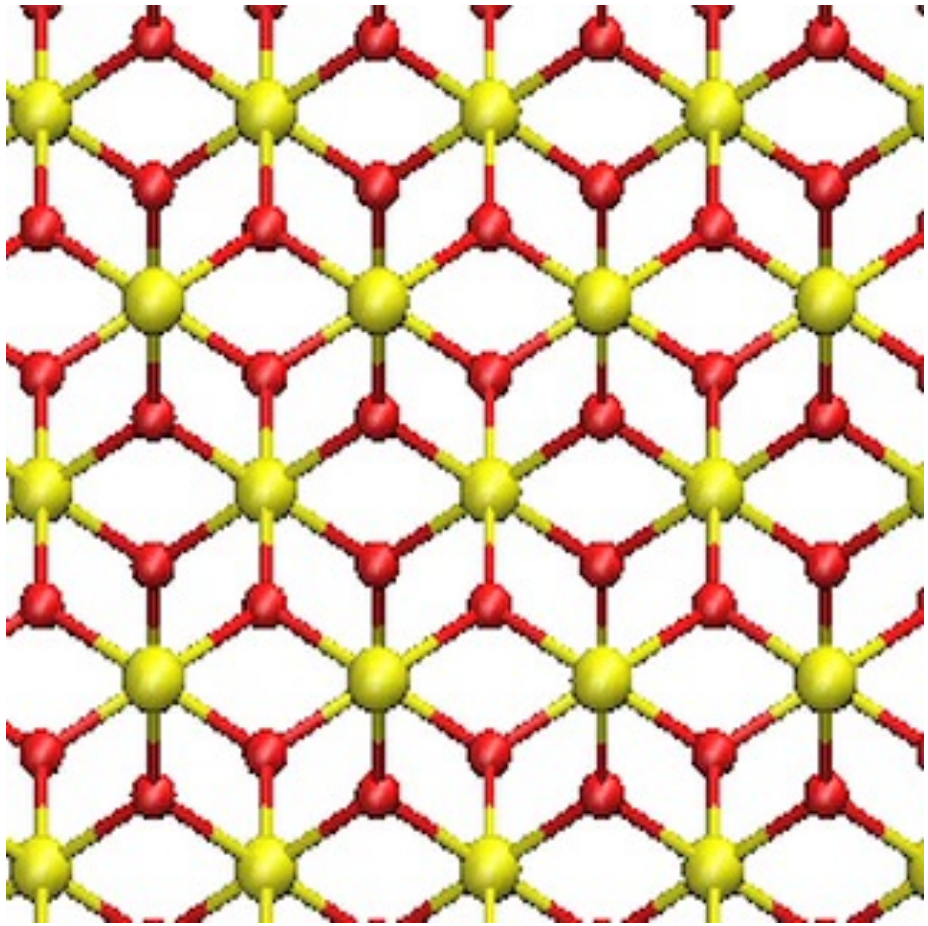


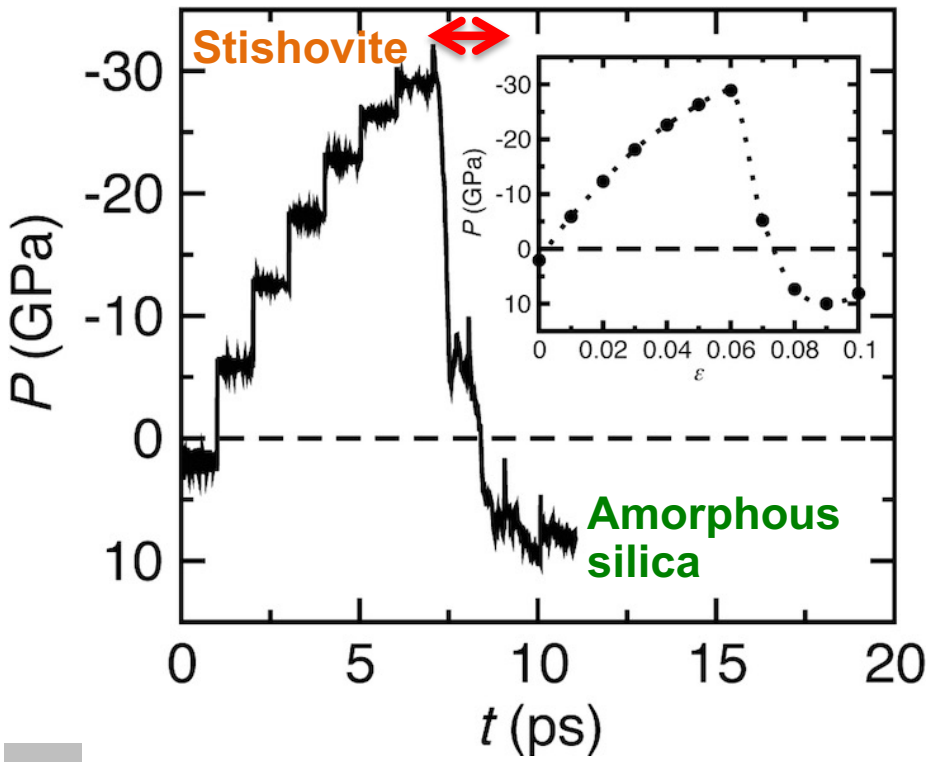
K. Yoshida *et al.*, *Sci. Rep*. **5**, 10993 ('15); *Acta Mater*. **124**, 316 ('17)

- **Toughening mechanism hypothesized to be amorphization under tension**
- **To catch up with a fast moving crack, amorphization needs rapid, but no theoretical nor experimental evidence**

Rapid Tensile Amorphization

- **QMD simulation reveals rapid amorphization of stishovite within picoseconds under tension ~ 30 GPa**





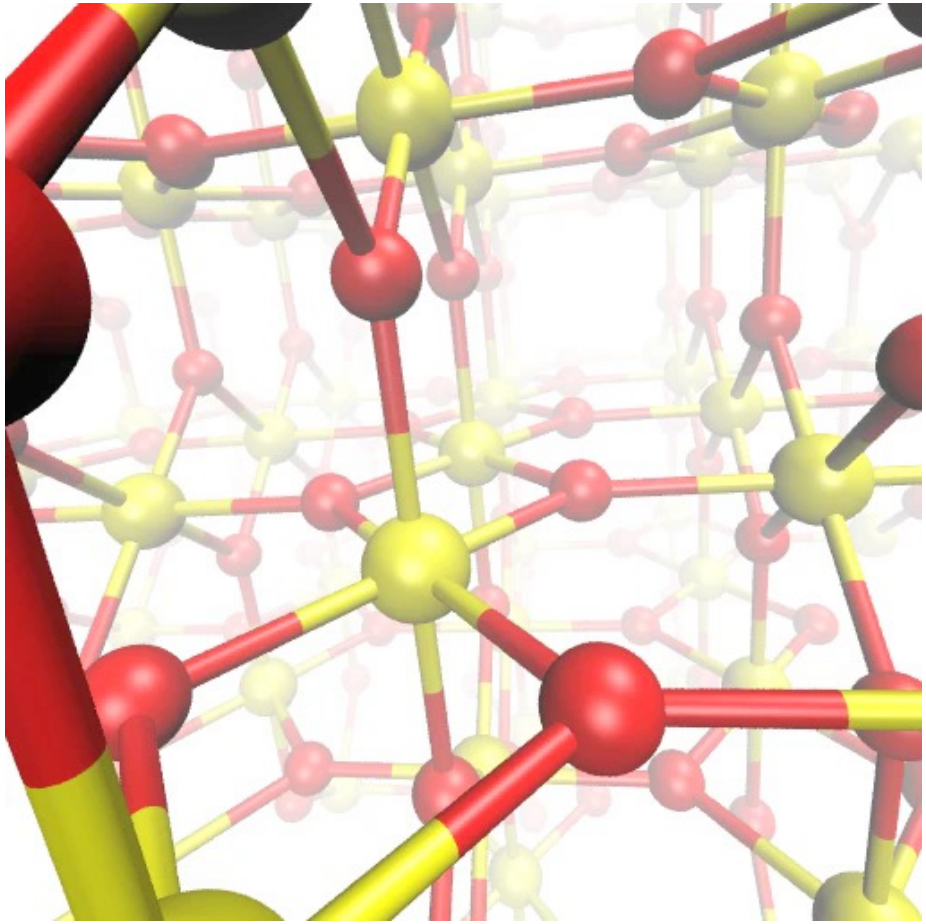
$Volume_{amorphous\ silica} \sim 2 \times Volume_{stishovite}$

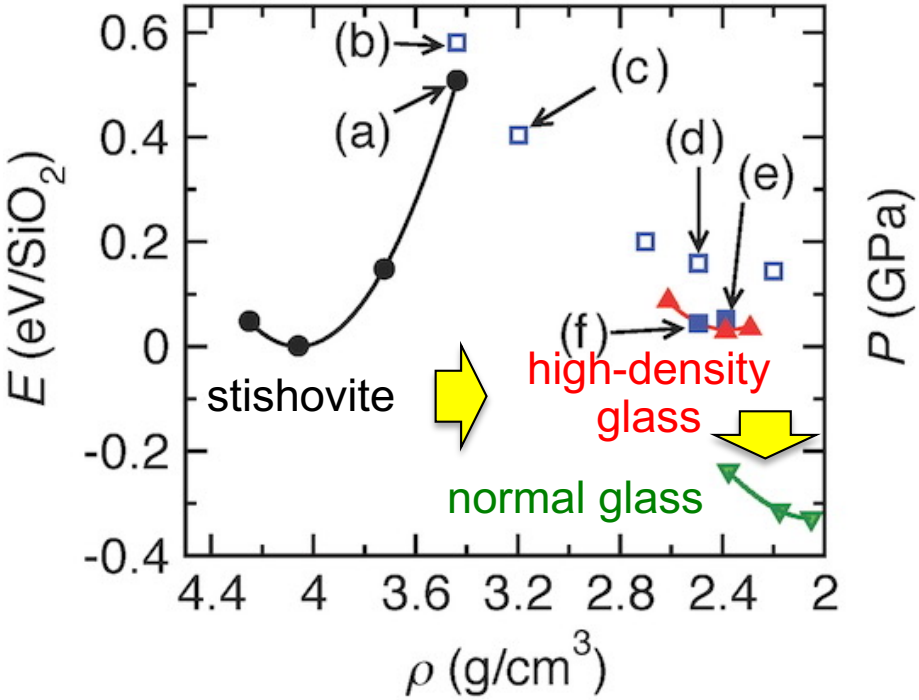
- **The rapid & expansive amorphization can catch up with, screen & self-heal a fast-moving crack**

M. Misawa *et al.*, *Science Advances* **3**, e1602339 ('17)

Rapid Amorphization Mechanism

- **Found a displacive amorphization mechanism that only involves short-distance collective motions of atoms, thereby facilitating the rapid transformation**

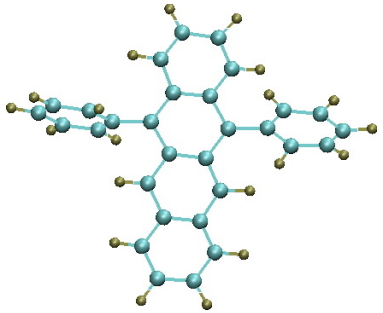




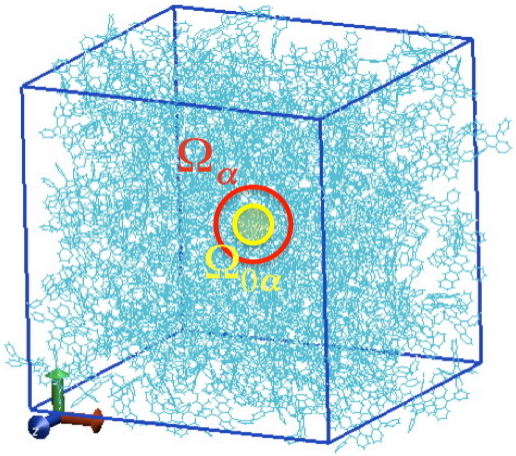
- **Two-step amorphization pathway from stishovite to glass involves an intermediate state akin to an experimentally suggested "high-density glass polymorph"**

Singlet Fission in Amorphous DPT

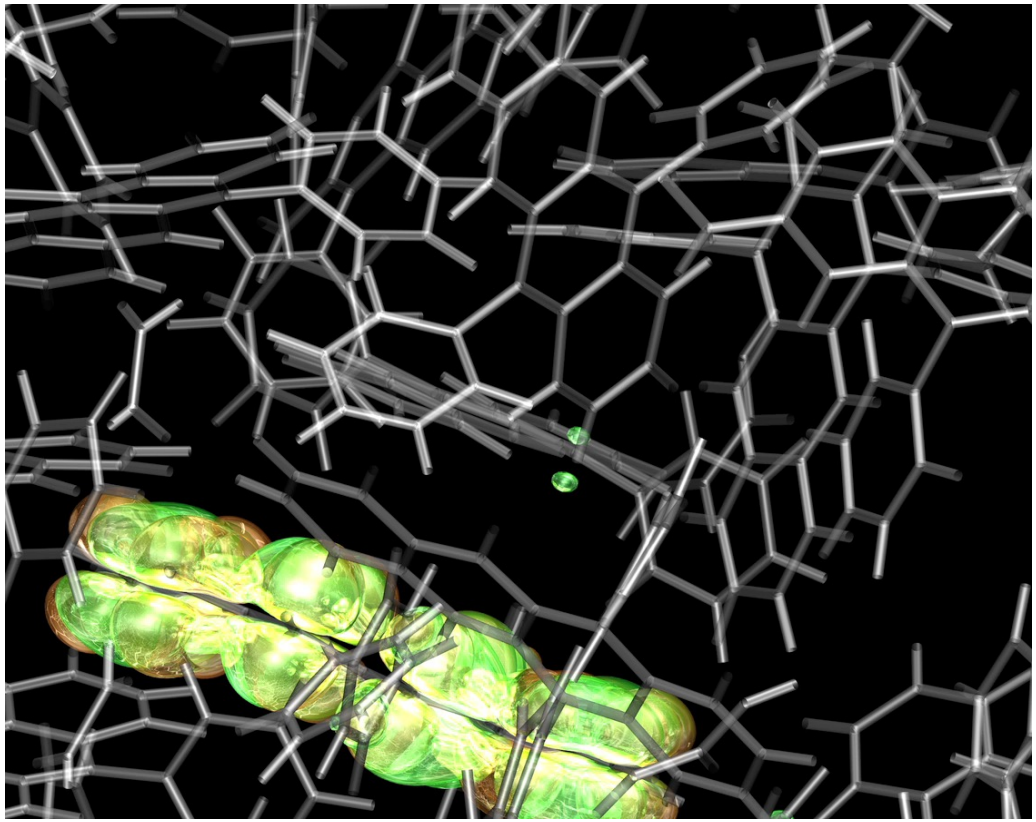
- **Photo-current doubling by splitting a singlet exciton into 2 triplet excitons**
- **Singlet fission in mass-produced disordered organic solid → efficient low-cost solar cells**
- **Experimental breakthrough: SF found in amorphous diphenyl tetracene (DPT)**



DPT molecule



Amorphous DPT



Quasi-electron

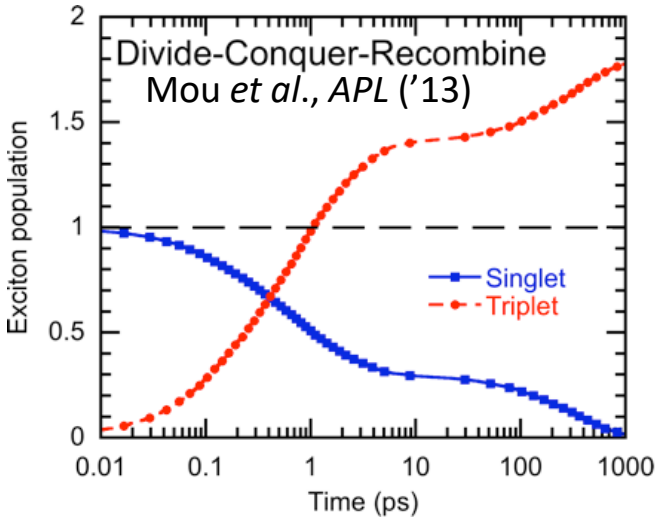
Quasi-hole

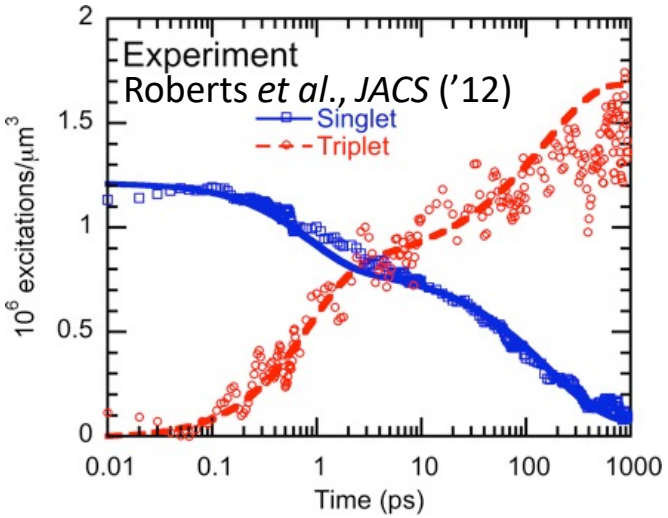
W. Mou *et al.*, *Appl. Phys. Lett.* **102**, 173301 ('13)

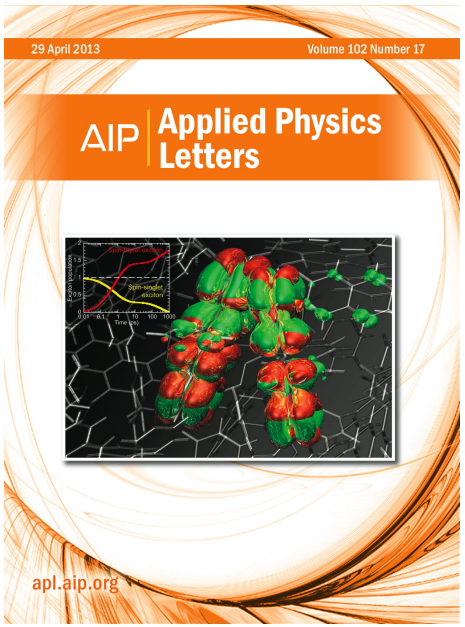
- **Divide-conquer-recombine nonadiabatic QMD (phonon-assisted exciton dynamics) + time-dependent perturbation theory (singlet-fission rate) + kinetic Monte Carlo calculations of exciton population dynamics in 6,400-atom amorphous DPT**

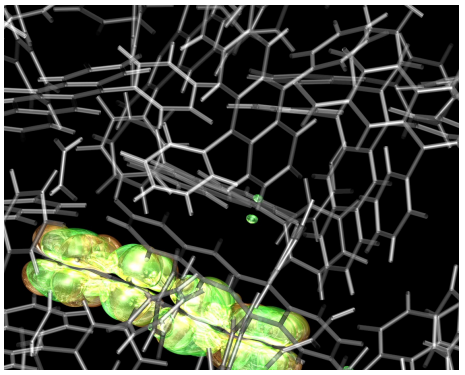
Singlet-Fission Hot Spot

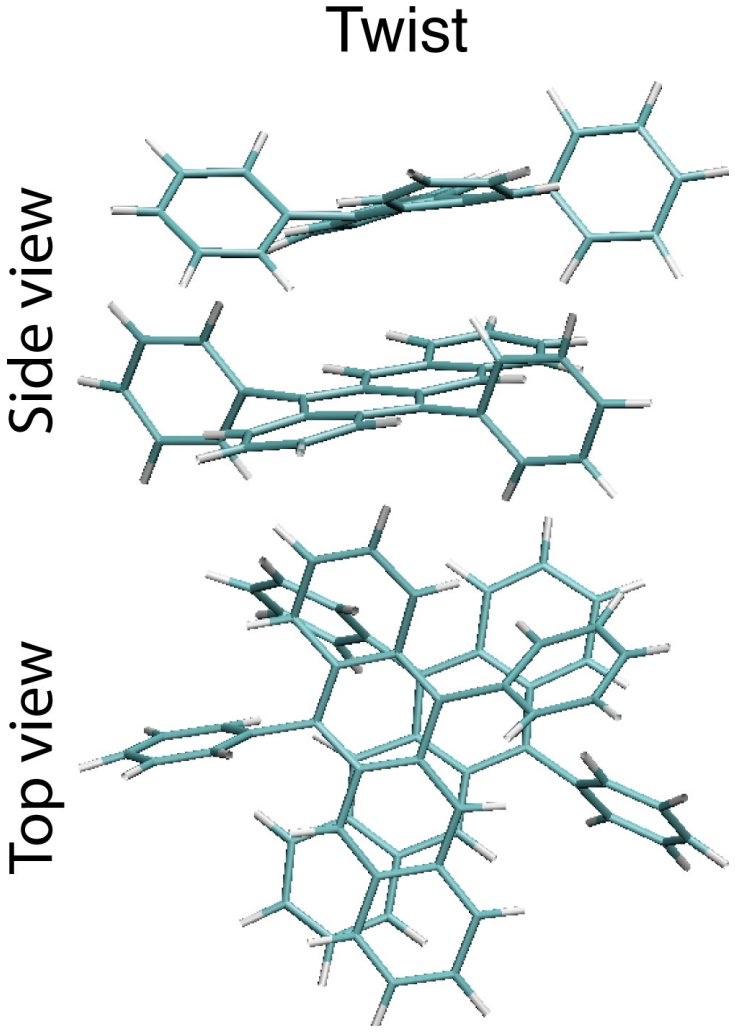
- **Nonadiabatic quantum molecular dynamics simulations not only reproduced experimentally measured exciton population dynamics but also revealed unknown molecular geometry of singlet fission hot spots**





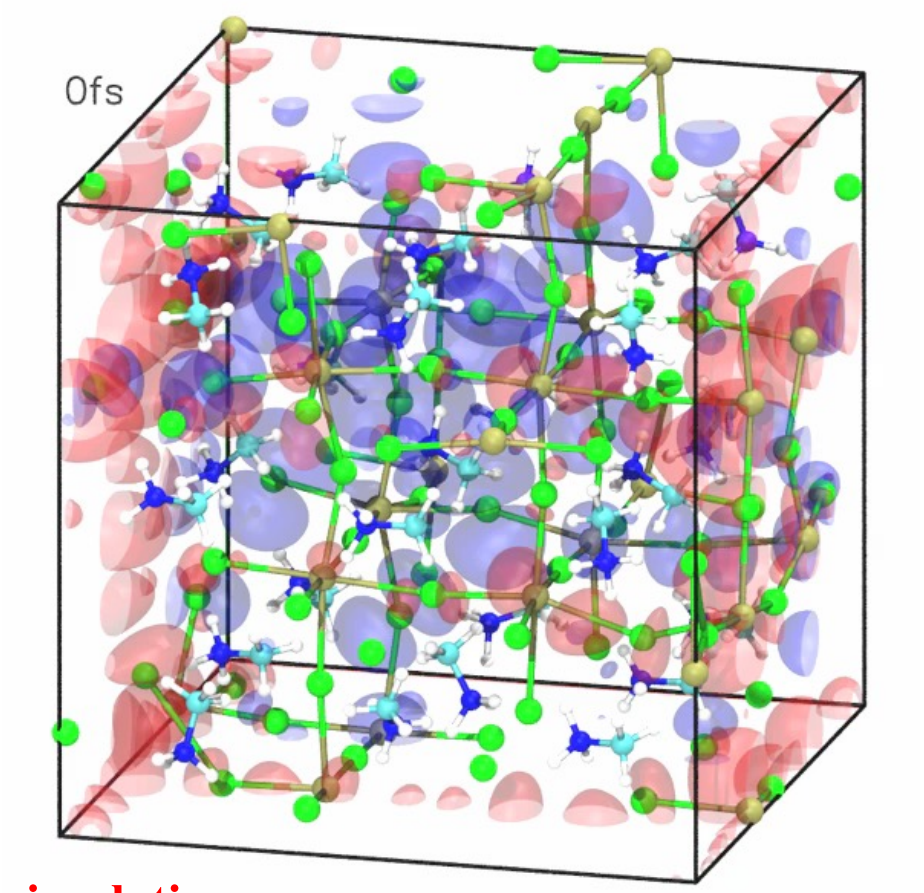






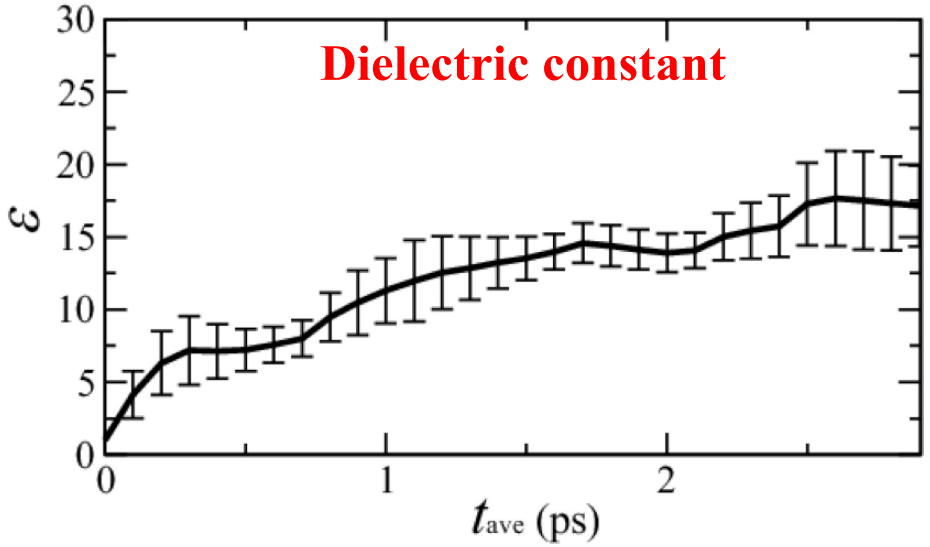
Photoexcited Carriers in $MAPbI_3$

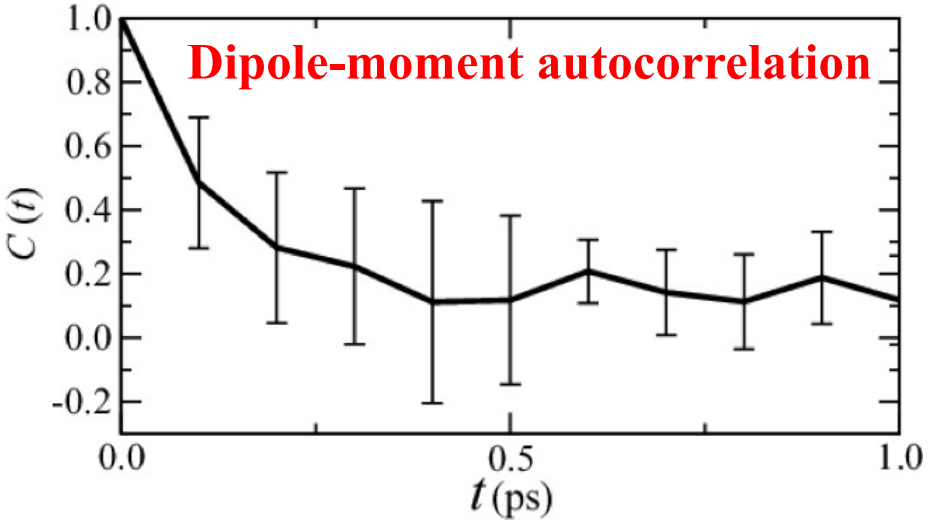
- **Organometal halide perovskites (*e.g.* methylammonium lead iodide, $CH_3NH_3PbI_3$ or $MAPbI_3$) for solar cells with high power conversion efficiency > 20%**
  [Stranks & Snaith, *Nat. Nanotechnol.* **10**, 391 ('15)]



- **Nonabiadiabatic QMD simulation**
  Pb & I sublattices act as disjunct pathways for rapid & balanced transport of free electrons & holes — electron (63% Pb-6p) & hole (90% I-5p);
  diffusion coefficients $D_e = (1.16\pm0.31)\times10^{-2}$ cm^2/s & $D_h = (1.01\pm0.42)\times10^{-2}$ cm^2/s
  Expt: $D_e = (1.7\pm1.1)\times10^{-2}$ cm^2/s & $D_h = (1.1\pm0.7)\times10^{-2}$ cm^2/s [Stranks *et al.*, *Science* **342**, 341 ('13)]

Screening Role of Methylammonium Sublattice





$$\varepsilon = 1 + \frac{4\pi}{3k_B TV}\left(\langle \mathbf{M}^2 \rangle - \langle \mathbf{M} \rangle^2\right)$$

time average

dipole moment

$cf.\ \varepsilon_{\text{expt}}\,(10^{12}\ \text{Hz}) = 7\text{-}10$

Lin *et al.*, *Nat. Photonics* **9**, 106 ('15)

$$C(t) = \frac{\langle \mathbf{M}(t + t_0) \cdot \mathbf{M}(t_0) \rangle}{\langle \mathbf{M}(t_0) \cdot \mathbf{M}(t_0) \rangle}$$

Rapid response time ~ 1 ps

$cf.\ \tau_{\text{expt}} = 2$ ps

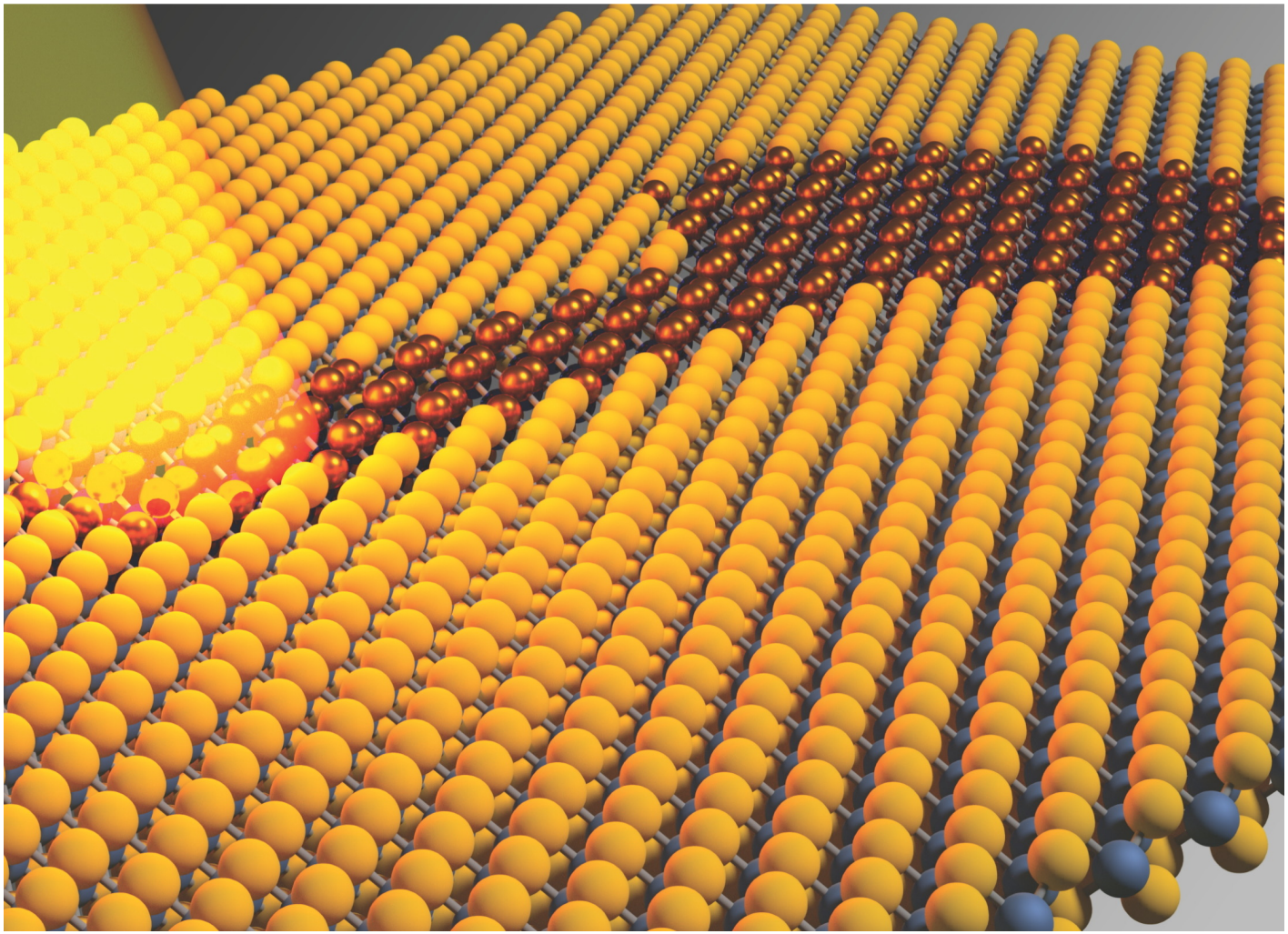
Deschler *et al.*, *JPCL* **5**, 1421 ('15)

- **Large dielectric constant of MA sublattice causes small exciton binding energy, 0.012 ± 0.009 eV (experimental upper bound = 0.05 eV** [D'Innocenzo *et al.*, *Nat. Commun.* **5**, 3586 ('14)]**)**
- **MA sublattice quickly screens out electrostatic electron-hole attraction to unbind an exciton & generate free carriers within 1 ps** [*cf.* Zhu *et al.*, *Science* **353**, 1409 ('16)]

Hakamata *et al.*, *Sci. Rep.* **5**, 19599 ('16)

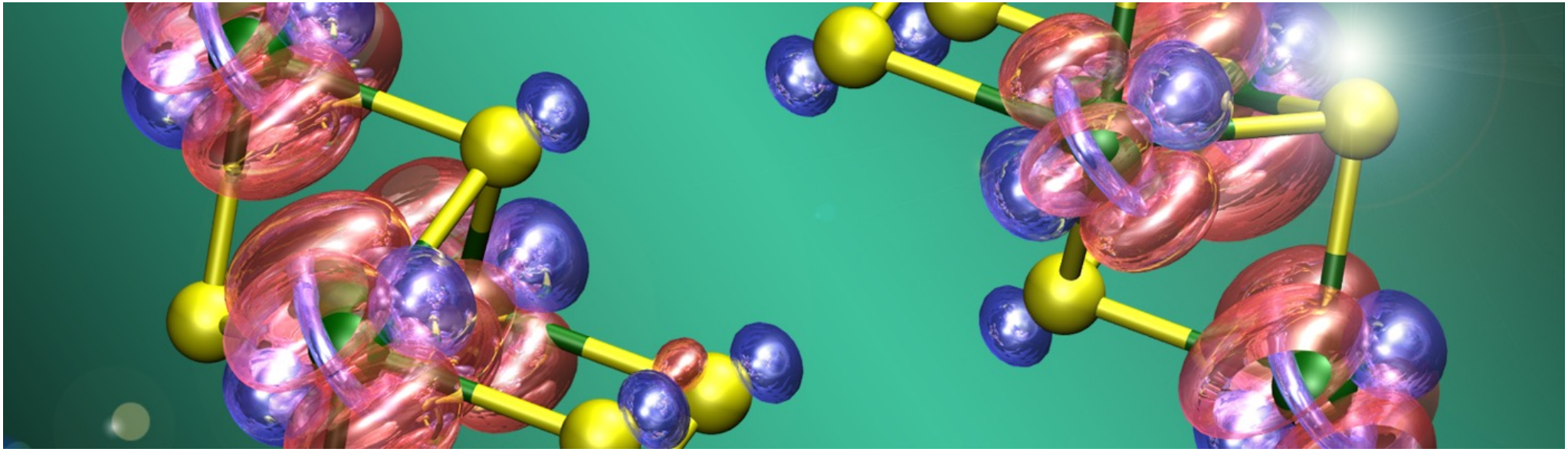
Ultrafast Control of Quantum Materials

Goal: Use ultrafast laser pulses to transform material structures & properties (*e.g.*, semiconductor-to-metal) on demand

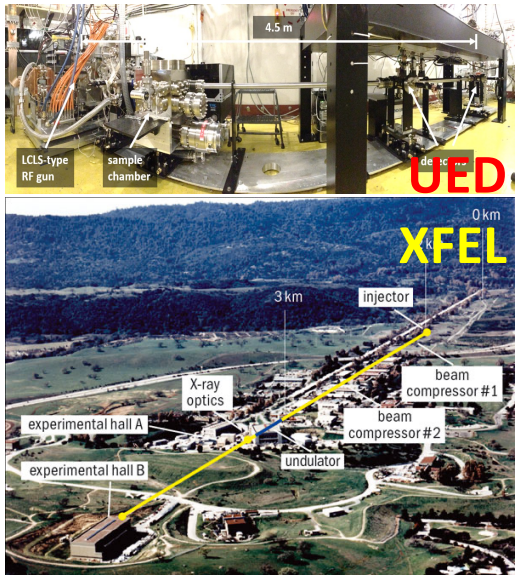


A. Krishnamoorthy *et al.*, *Nanoscale* **10**, 2742 (’18); journal cover

Simulation-Experiment Synergy



- **In ultrafast 'electron & X-ray cameras,' laser light hitting a material is almost completely converted into nuclear motions — key to switching material properties on & off at will for future electronics applications.**
- **High-end nonadiabatic quantum molecular dynamics simulations reproduce the ultrafast energy conversion at exactly the same space & time scales, and explain it as a consequence of photo-induced phonon softening.**

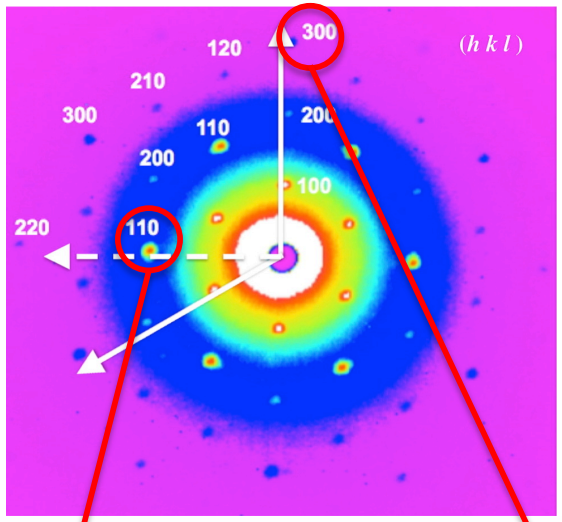


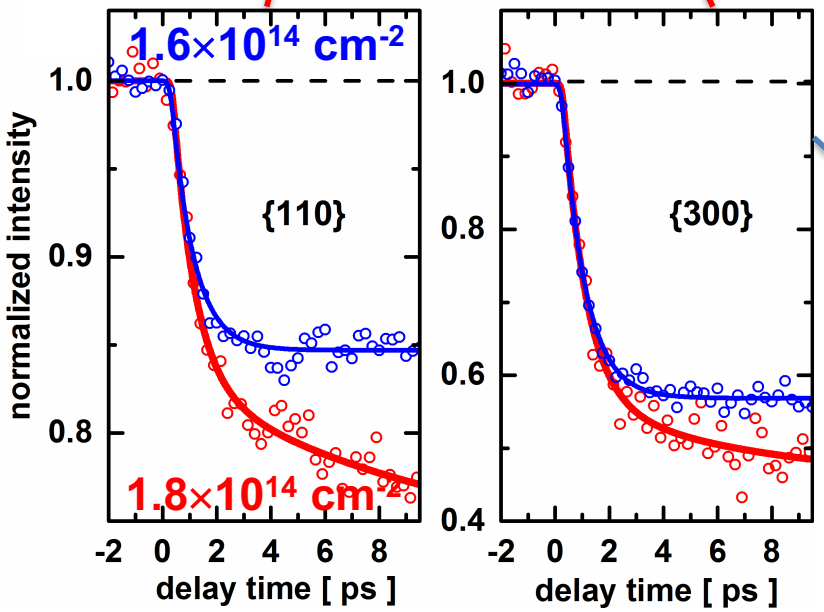
Ultrafast electron diffraction: M.F. Lin *et al*., *Nature Commun*. **8**, 1745 ('17)

X-ray free-electron laser: I. Tung *et al*., *Nature Photon.* **13**, 425 ('19)

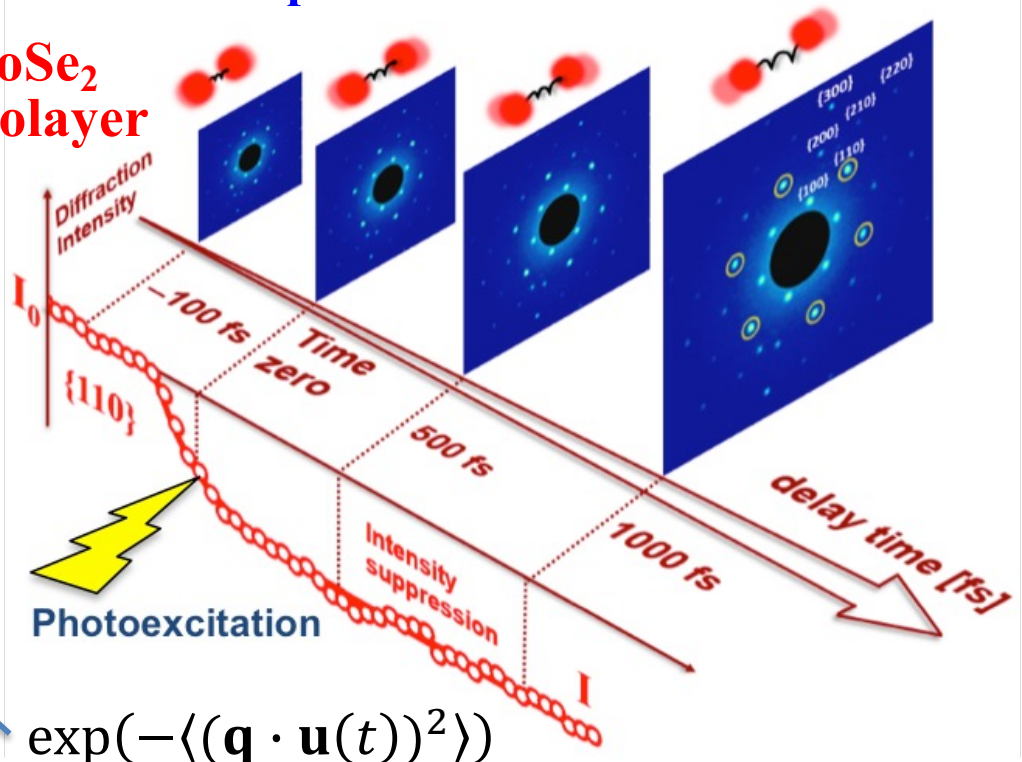
Ultrafast Coupled Electron-Lattice Dynamics

- **Ultrafast electron diffraction experiment shows nearly perfect energy conversion from electronic excitation to lattice motions within ps**





$MoSe_2$ monolayer



$$\exp(-\langle(\mathbf{q}\cdot\mathbf{u}(t))^2\rangle)$$

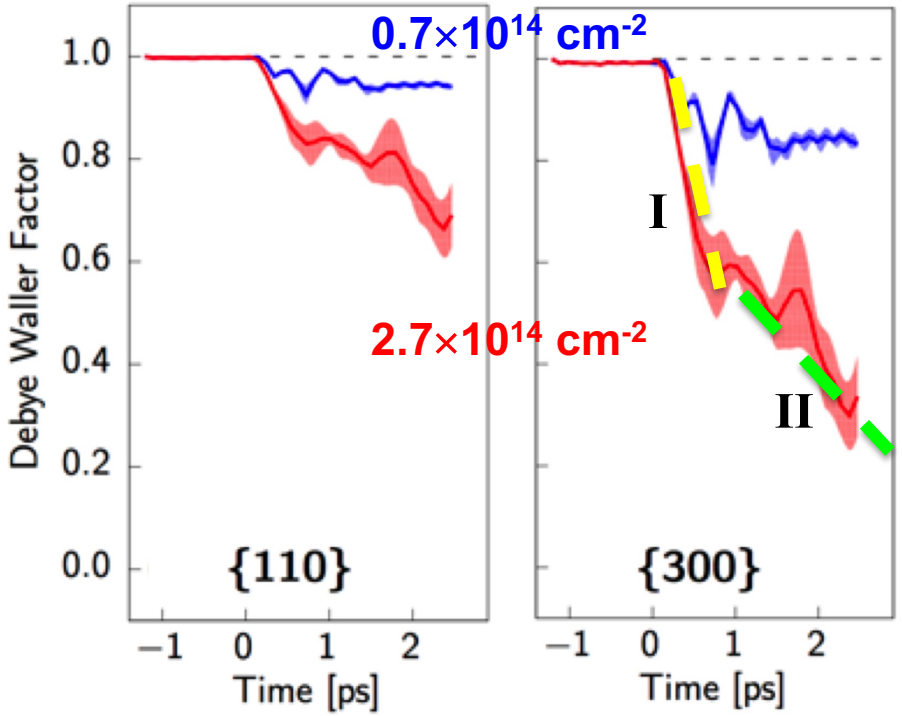
atomic displacement

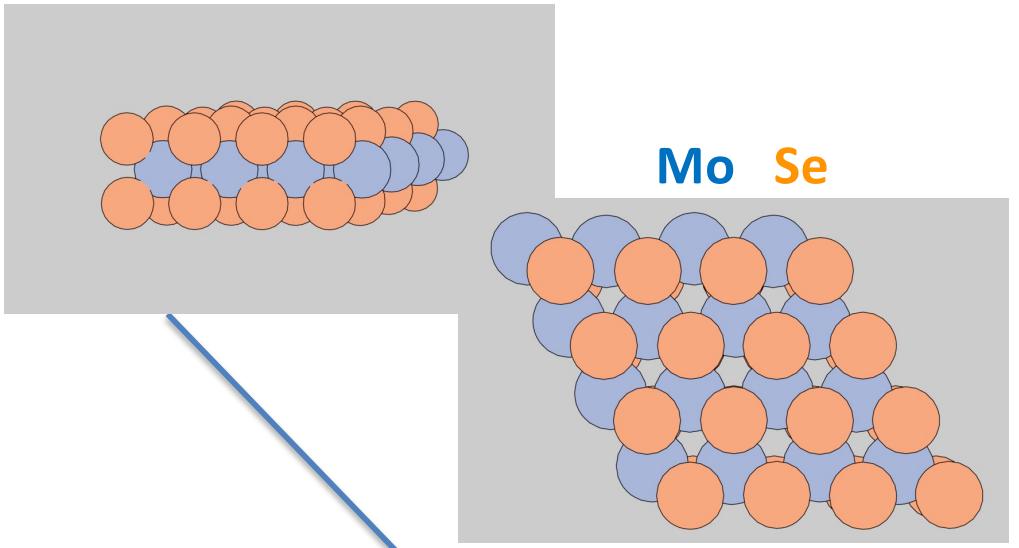
- **Dynamics of Debye-Waller factor reveals rapid disordering for both {300} & {110} peaks**
- **Transition from mono- to bi-exponential decay at higher electron-hole density**

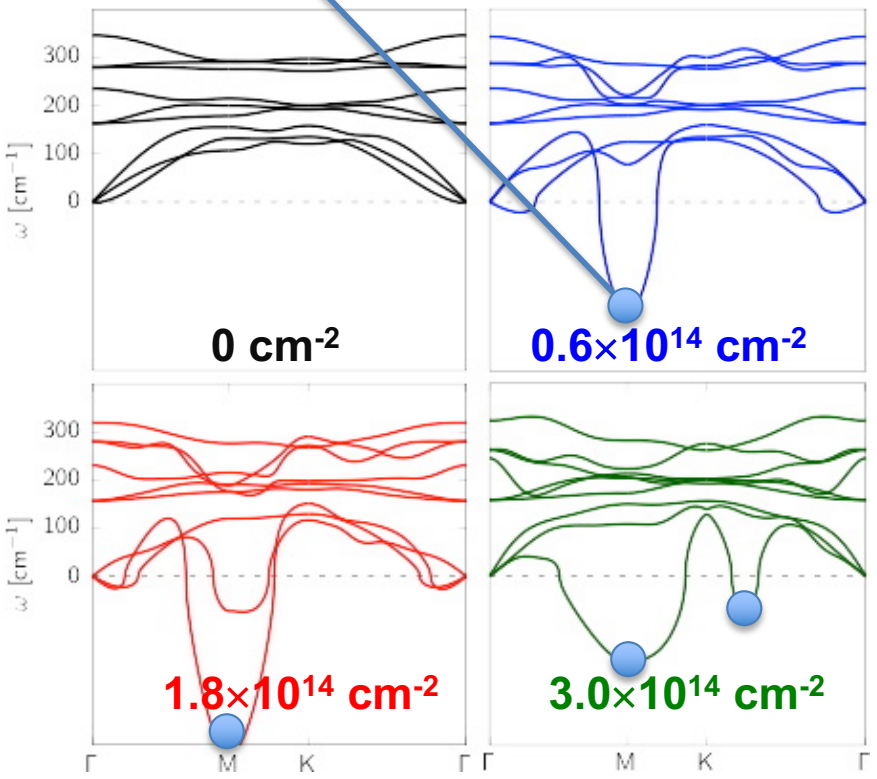
M.F. Lin *et al.*, *Nature Commun.* **8**, 1745 ('17)

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**





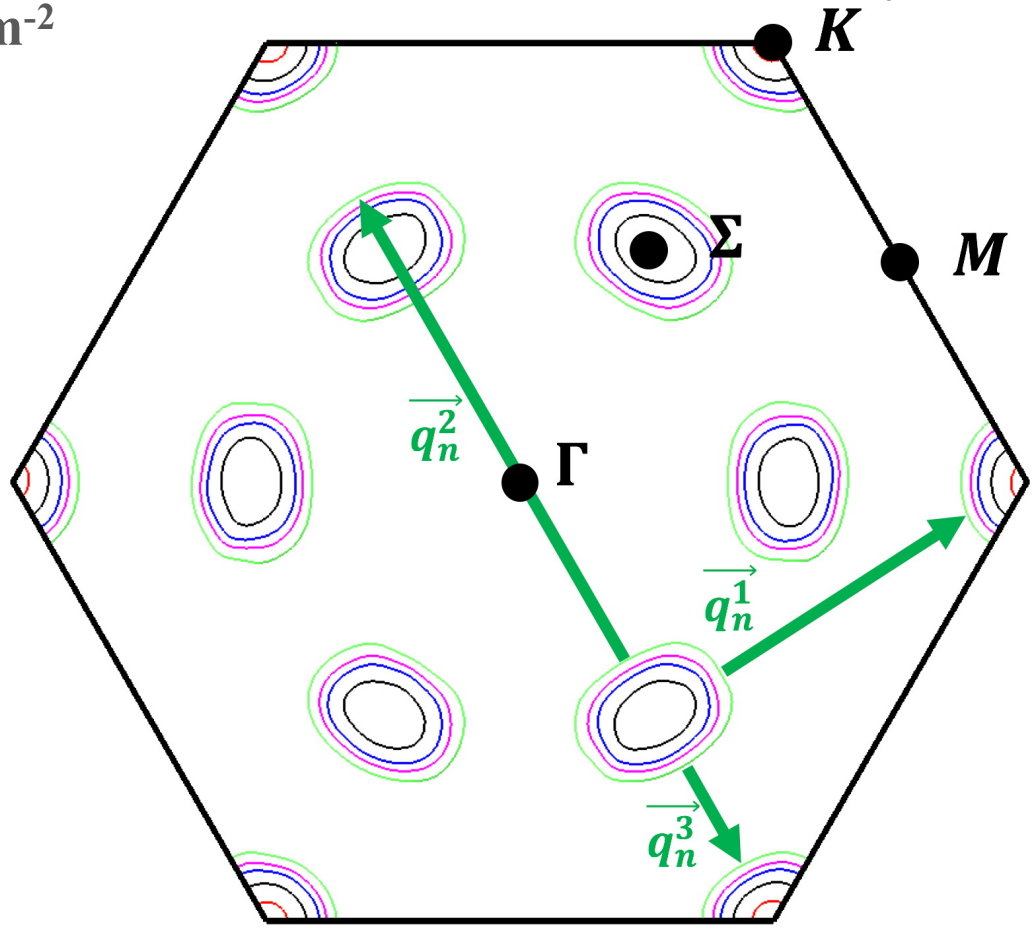


- **Rapid lattice dynamics is explained by the softening of M-point (1/2 0 0) phonon**
- **Bi-exponential transition is explained by the softening of additional phonon modes at higher electron-hole densities**

Lin *et al.*, *Nature Commun.* **8**, 1745 ('17)
Bassman *et al.*, *Nano Lett.* **18**, 4653 ('18)

Electronic Origin of Phonon Softening

- Electronic Fermi surface for the electron-hole density n(e-h) ranging from 0.2 to 2×10^{14} cm^{-2}

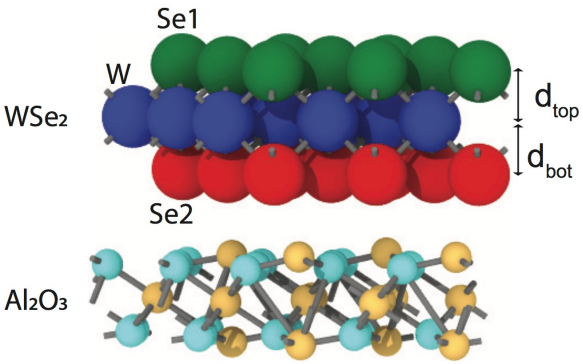


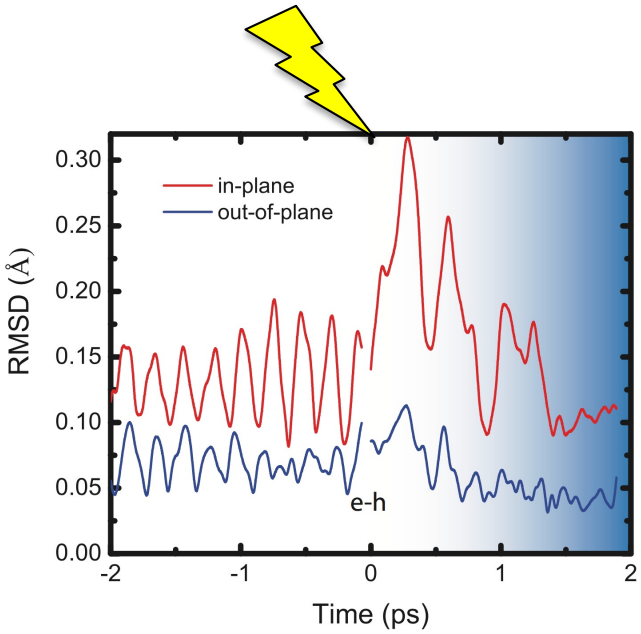
n(e-h) = 0.22, 1, 2, 3, 4 $\times10^{14}$ cm^{-2}

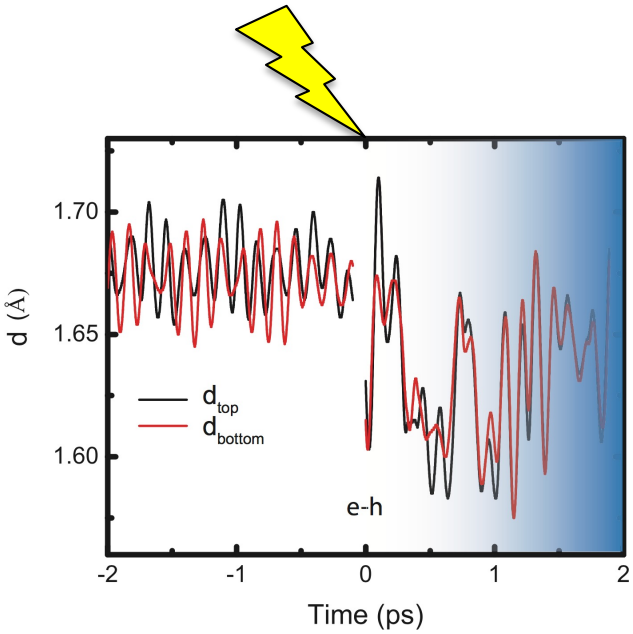
- While the Fermi surface is localized at K-points at minimal excitation (red), it occupies Σ-pockets at larger n(e-h) (black & blue), enabling electron scattering by emitting $\vec{q}_n^1$ (M), $\vec{q}_n^2$ (Σ) and $\vec{q}_n^3$ (K) phonons

WSe_2 Monolayer on Al_2O_3 Substrate

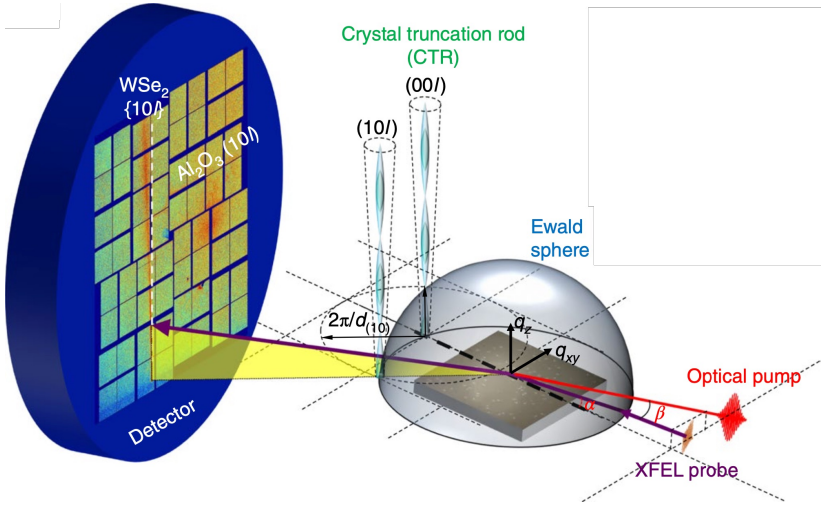
- **NAQMD simulation to study photoexcitation dynamics of WSe_2 monolayer on Al_2O_3 substrate**







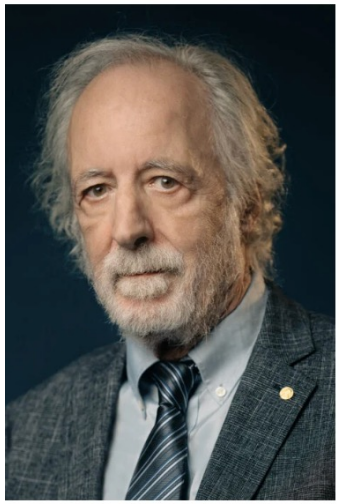
- **Enhanced in-plane atomic displacements upon photoexcitation**
- **Photo-induced intralayer contraction of W-Se distances**
- **Good agreement with femtosecond surface X-ray scattering experiments at Stanford SLAC/LCLS**



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

Attosecond Light-Matter Interaction

The Nobel Prize in Physics 2023



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Pierre Agostini

Prize share: 1/3

© Nobel Prize Outreach. Photo: Clément Morin

Ferenc Krausz

Prize share: 1/3

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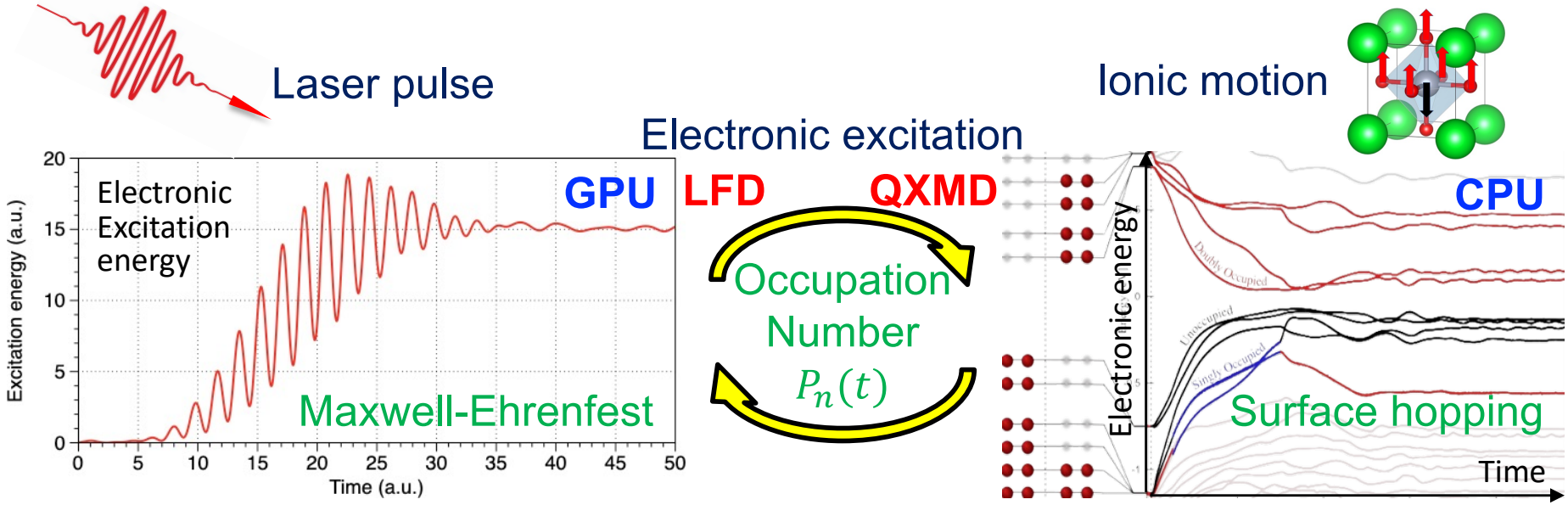
Anne L'Huillier

Prize share: 1/3

The Nobel Prize in Physics 2023 was awarded to Pierre Agostini, Ferenc Krausz and Anne L'Huillier "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

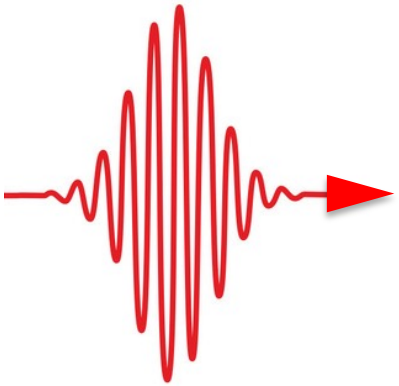
Nonadiabatic Quantum MD: DC-MESH

- **DC-MESH (divide-&-conquer Maxwell + Ehrenfest + surface-hopping): *O*(*N*) algorithm to simulate photo-induced quantum materials dynamics**
- **LFD (local field dynamics): Maxwell equations for light & real-time time-dependent density functional theory equations for electrons to describe light-matter interaction**
- **QXMD (quantum molecular dynamics with excitation): Nonadiabatic coupling of excited electrons & ionic motions based on surface-hopping approach**
- **"Shadow" LFD (GPU)-QXMD (CPU) handshaking *via* electronic occupation numbers with minimal CPU-GPU data transfer**
- **GSLD: Globally sparse (interdomain Hartree coupling *via* multigrid) & locally dense (intradomain nonlocal exchange-correlation computation *via* BLAS) solver**

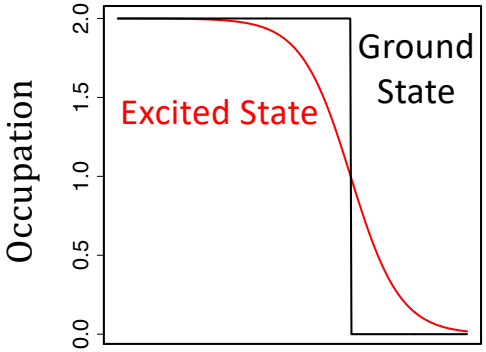


Razakh *et al.*, *PDSEC* (IEEE, '24)

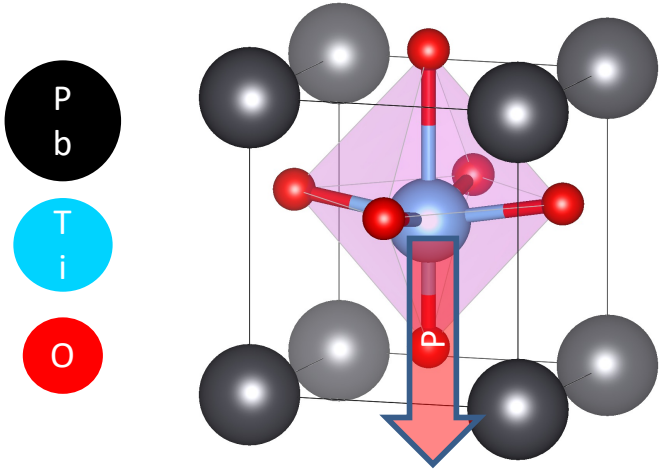
Multiscaling from DC-MESH to XS-NNQMD



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**

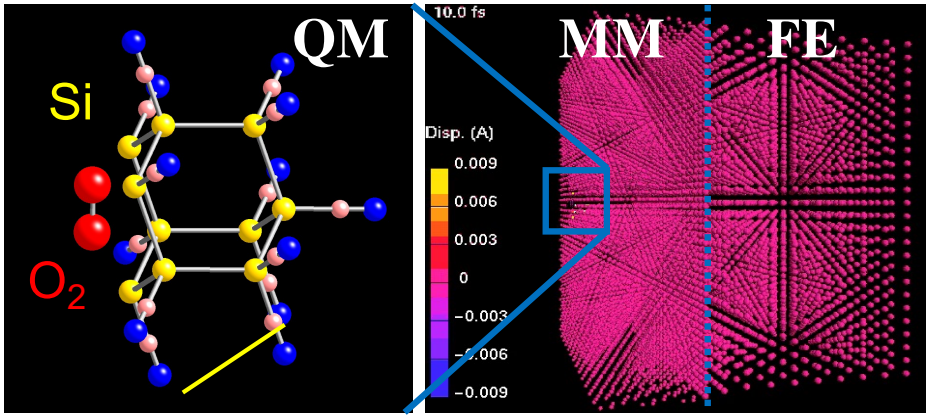
Excited energy landscape

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)

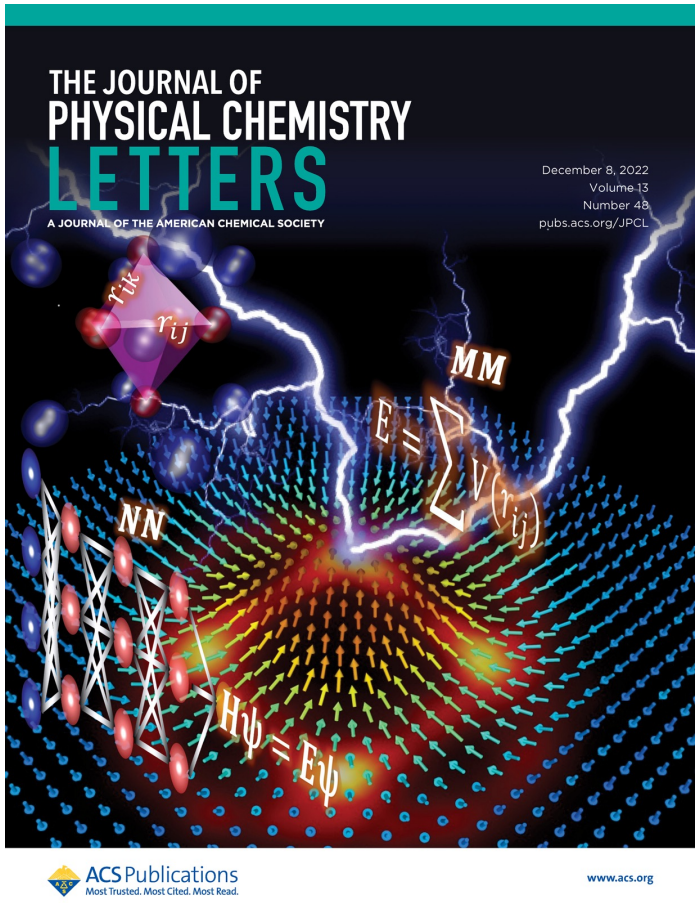
  Warshel, *Angew. Chem.* **53**, 10020 ('14)



QM/MM/FE (finite-element method)

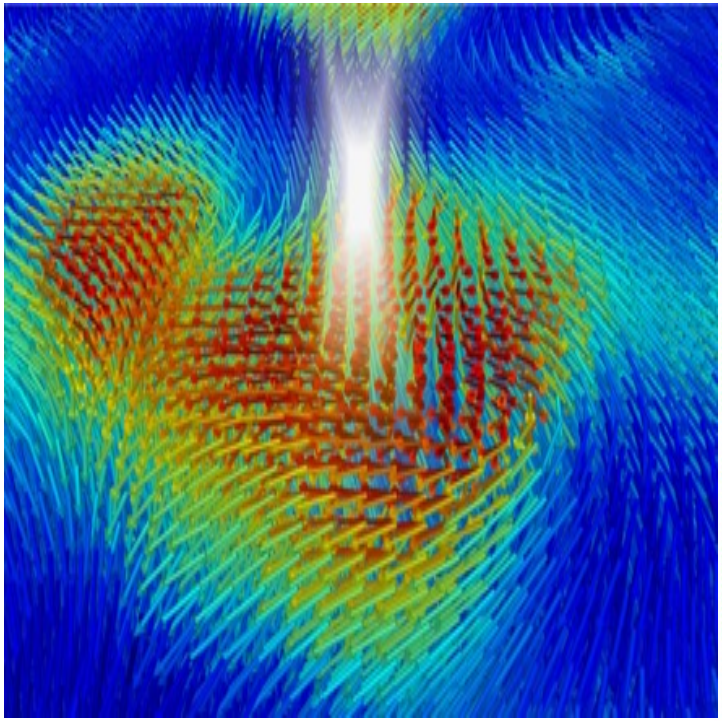
Ogata *et al*, *Comput. Phys. Commun.* **138**, 143 ('01)

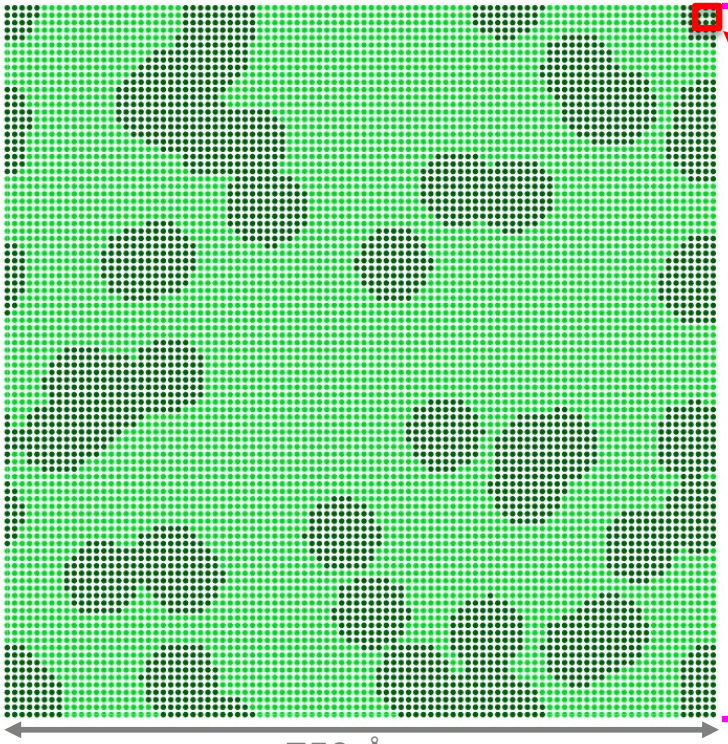
- **NN/MM:** NNQMD for ferroelectric ($PbTiO_3$: PTO) embedded in MM for paraelectric ($SrTiO_3$: STO) to apply appropriate strain boundary condition



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

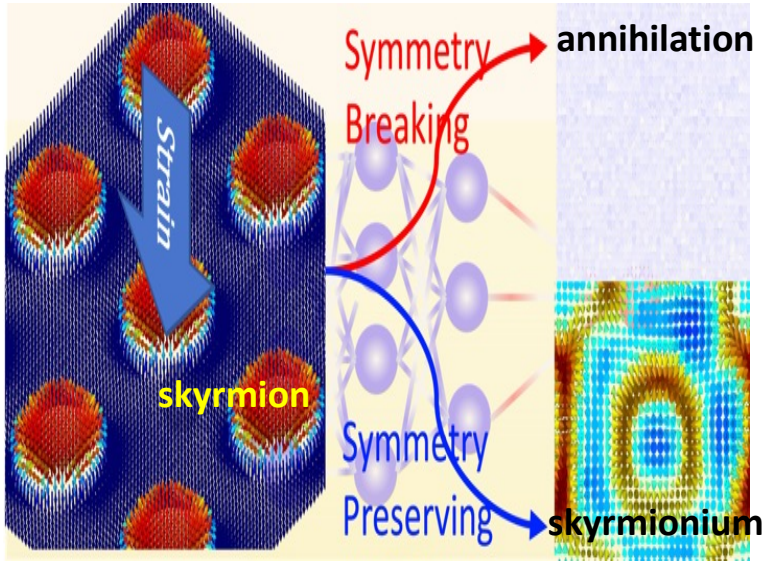
Application: Ferroelectric Opto-Topotronics





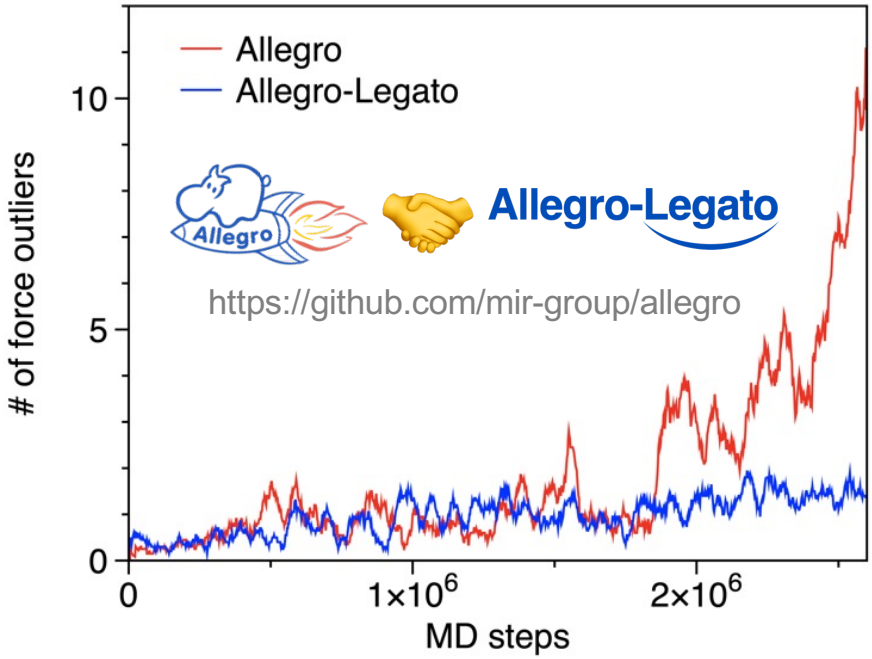
- **Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications**
- **Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)**
- **Symmetry-controlled skyrmion-to-skyrmionium***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

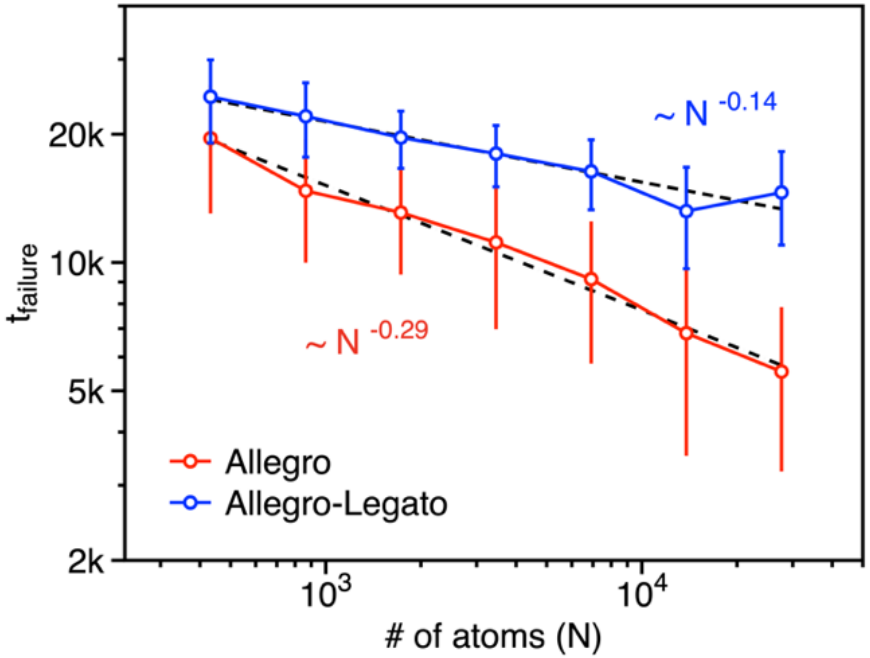
Linker *et al., Science Adv.* **8**, eabk2625 ('22); *JPCL* **13**, 11335 ('22); *Nano Lett.* **23**, 7456 ('23)



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**

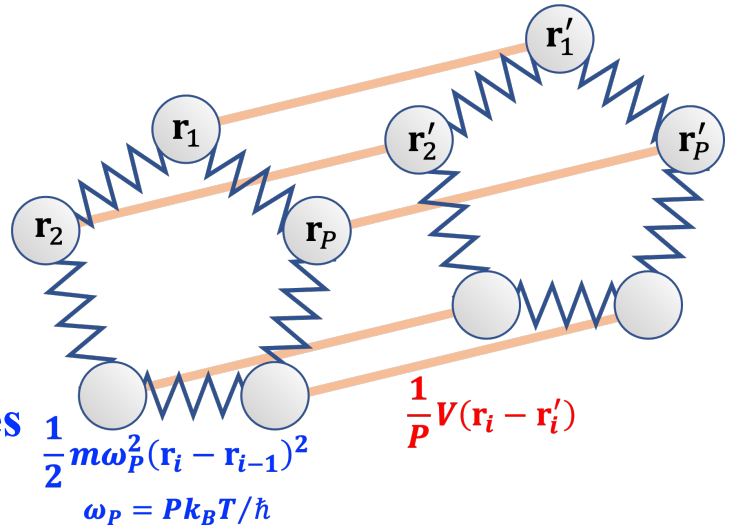


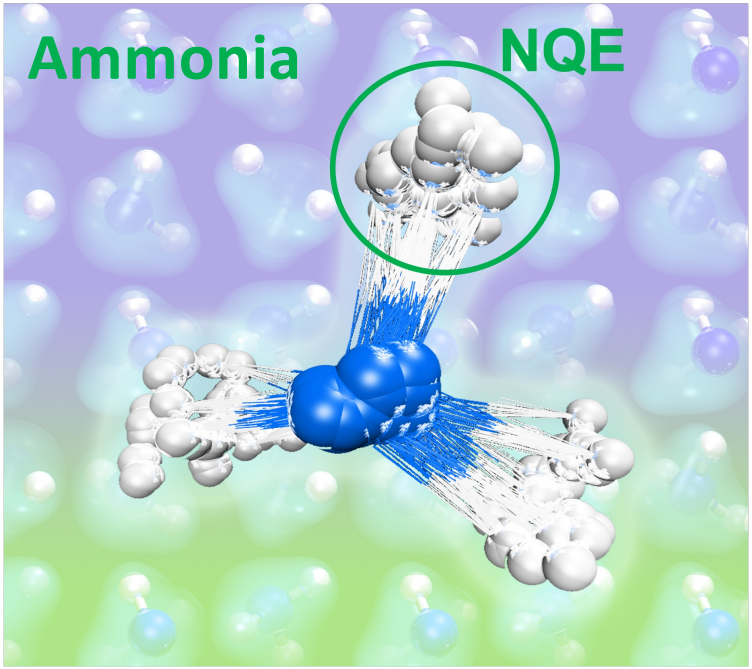


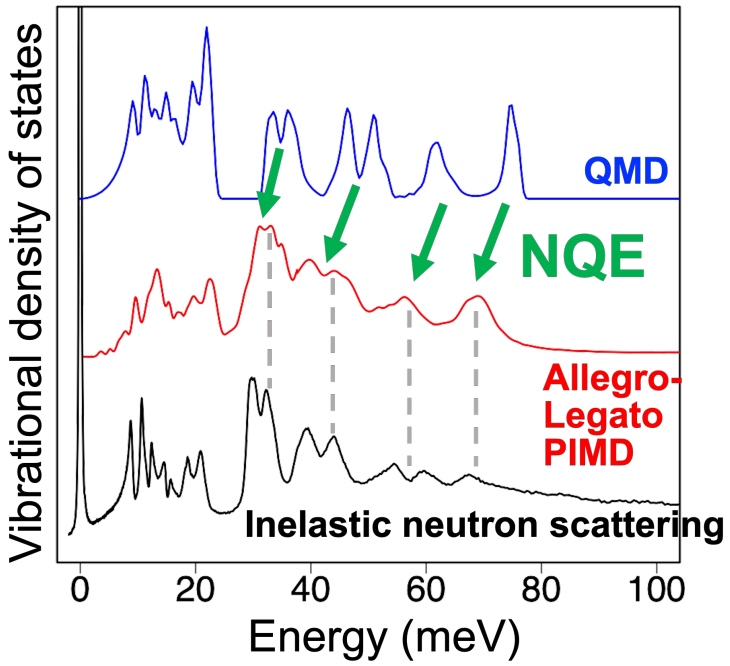
H. Ibayashi *et al.*, *ISC23—LNCS* **13948**, 223 ('23); *arXiv*: 2303.08169

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**







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

AIQ-XMaS Software Suite

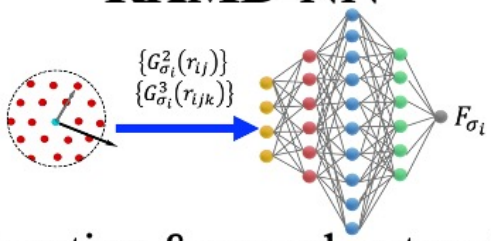
AI & Quantum-Computing Enabled Exa Quantum Materials Simulator

QXMD

Nonadiabatic quantum molecular dynamics

Aurora ESP engine

RXMD-NN



Reactive & neural-network molecular dynamics

GEARS

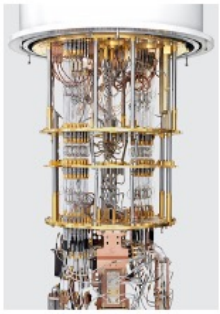
Augmented-reality user interface

EZFF

```
EZFF/
├── LICENSE
├── MANIFEST.in
├── README.md
├── docs
├── examples
├── ezff
│   ├── errors.py
│   ├── ffio.py
│   ├── interfaces
│   │   ├── gulp.py
│   │   ├── qchem.py
│   │   ├── vasp.py
│   │   ├── lammps.py
│   │   └── rxmd.py
│   └── utils
│       ├── convert_units.py
│       └── reaxff.py
├── setup.py
└── tests
```

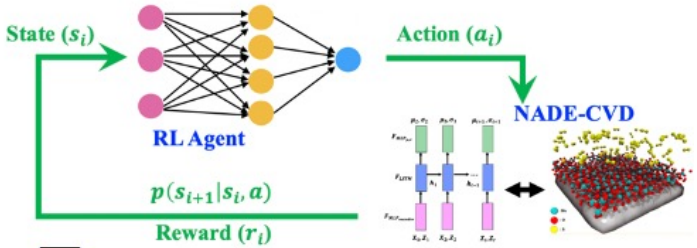
Easy force-field parameterization & uncertain quantification

MISTIQS



Quantum many-body dynamics on quantum computers

MAITAI



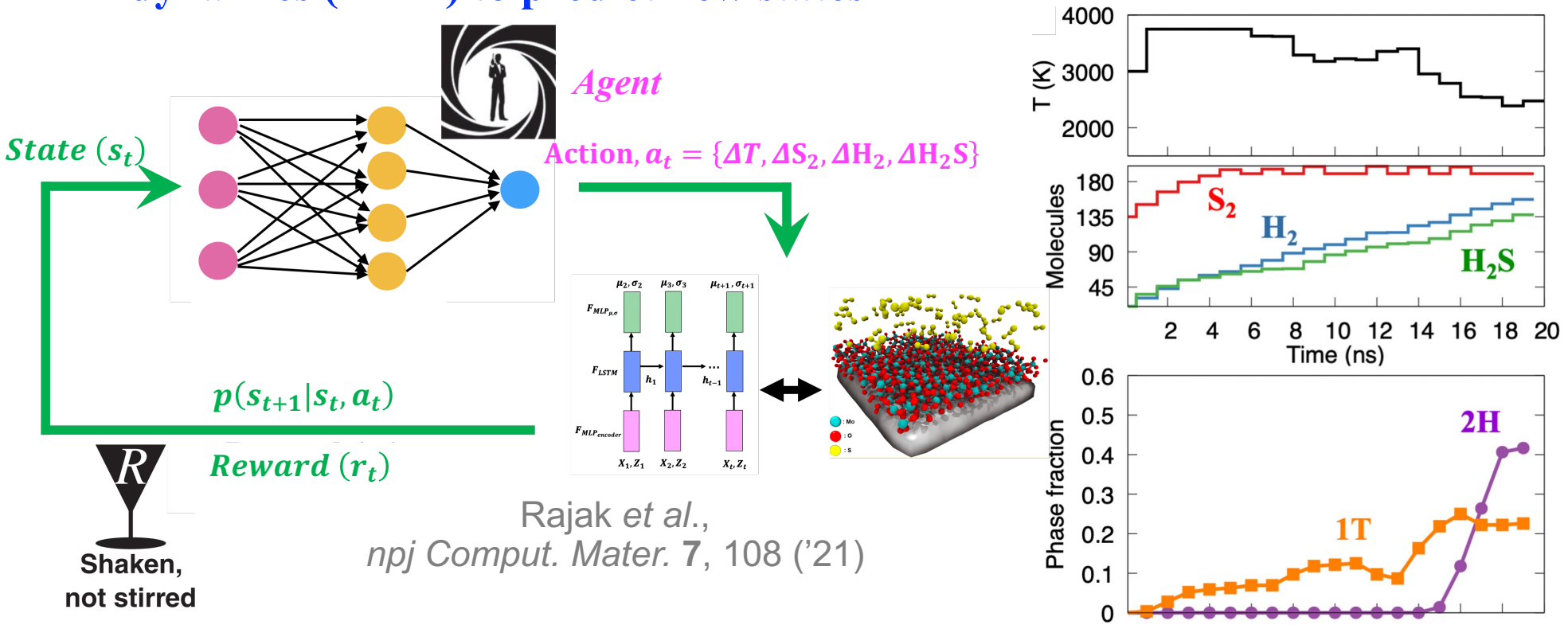
AI tools for materials design

You will obtain hands-on training on QXMD

https://cybermagics.netlify.app/software

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)

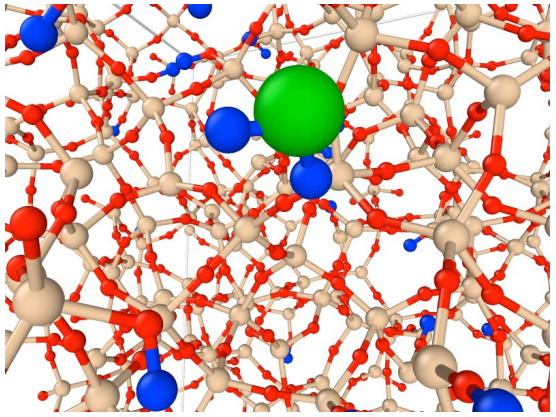
cf. Sgroi *et al.*, *Phys. Rev. Lett.* ***126***, 020601 ('21)

RL to Enable Long-Time Dynamics

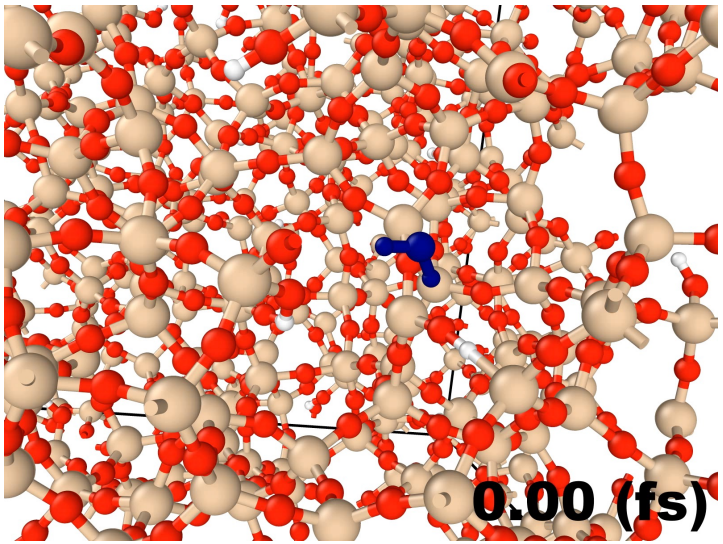
- **Phase 1—explore (agent parallelism): Multiple reinforcement learning (RL) agents autonomously discover *long low-activation-barrier pathways***
  Mnih, *Nature* **518**, 529 ('15); Hessel, *AAAI* **32**, 11796 ('18)
- **Phase 2—refine (time parallelism): Concurrent nudged-elastic-band (NEB) refinements of multiple *minimum-energy path (MEP) segments***
- **Estimate time based on transition-state theory**

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

Agent guiding O in H_2O

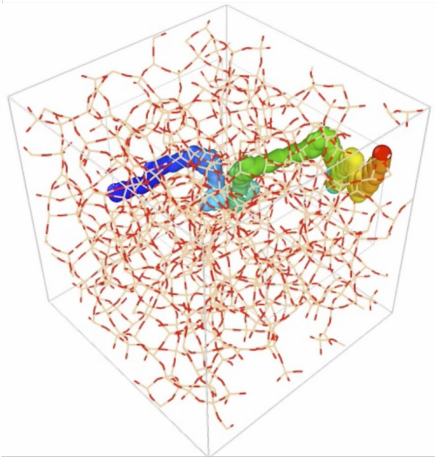


H_2O diffusion & reaction in SiO_2

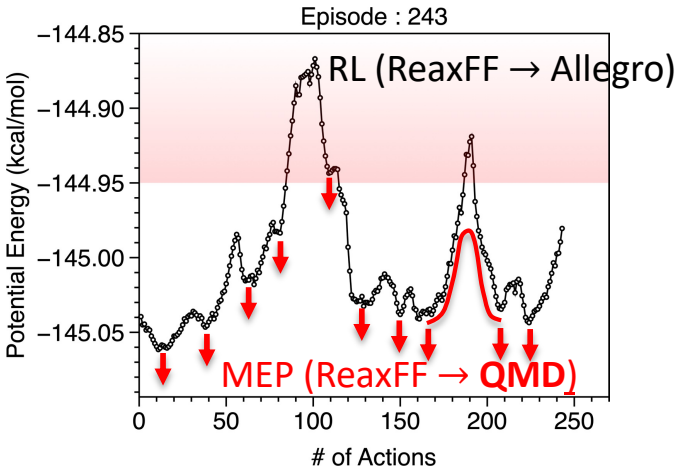


2-seconds trajectory

1. Explore

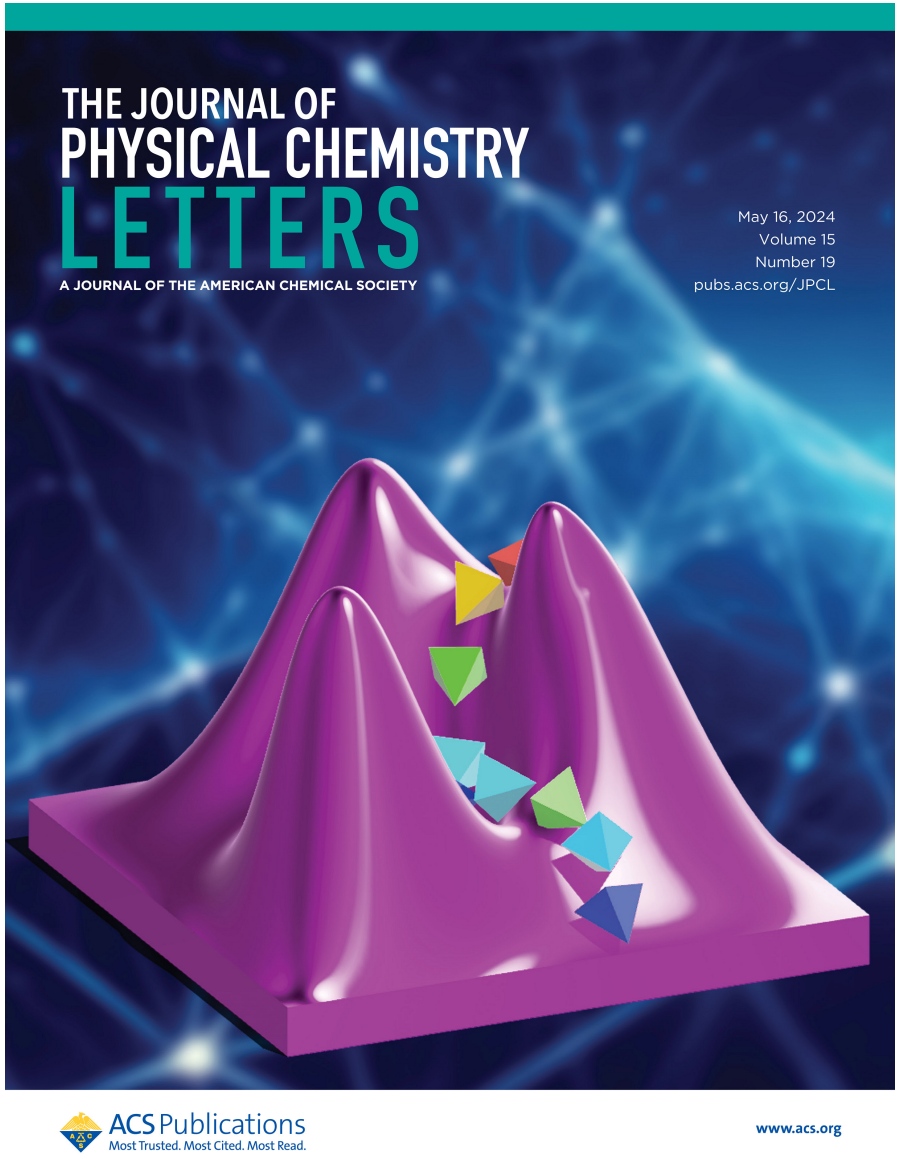


2. Refine



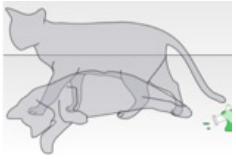
AI for long time!

RL to Enable Long-Time Dynamics

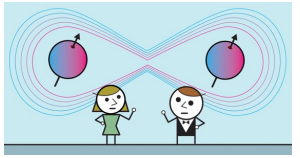


Nomura *et al*., J. *Phys. Chem. Lett*. **15**, 5288 ('24)

Quantum Computing (QC) for Science



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



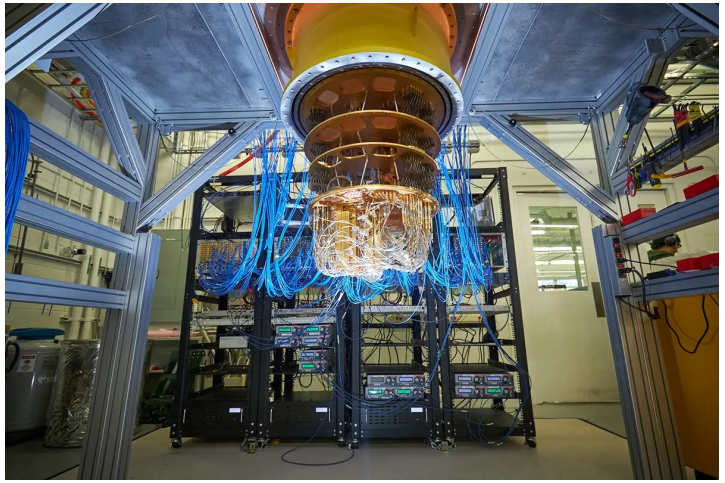
- **U.S. Congress (Dec. 21, '18) signed National Quantum Initiative Act to ensure leadership in quantum computing & its applications**
- **Quantum supremacy demonstrated by Google**

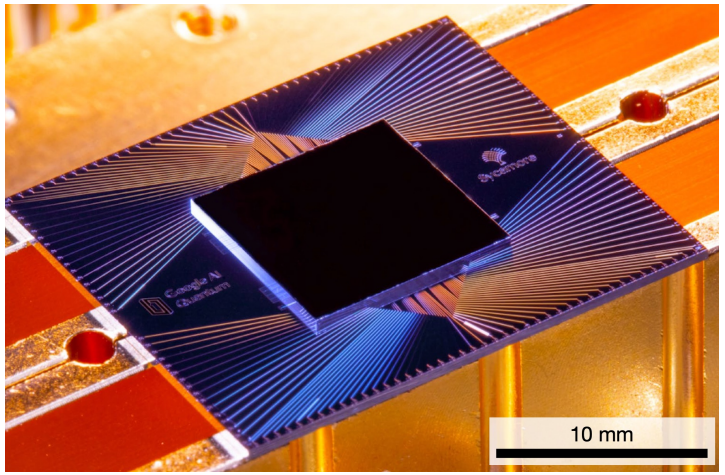
  F. Arute, *Nature* **574**, 505 ('19)
- **Quantum computing for science: *Universal simulator of quantum many-body systems***

  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)

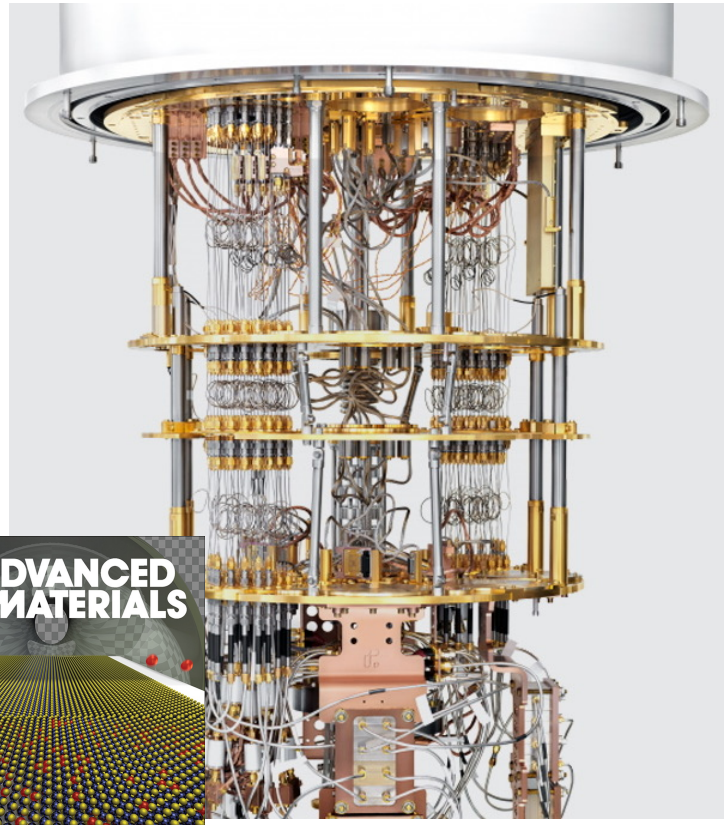


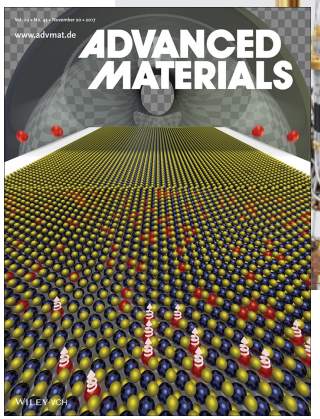


54-qubit Google Sycamore

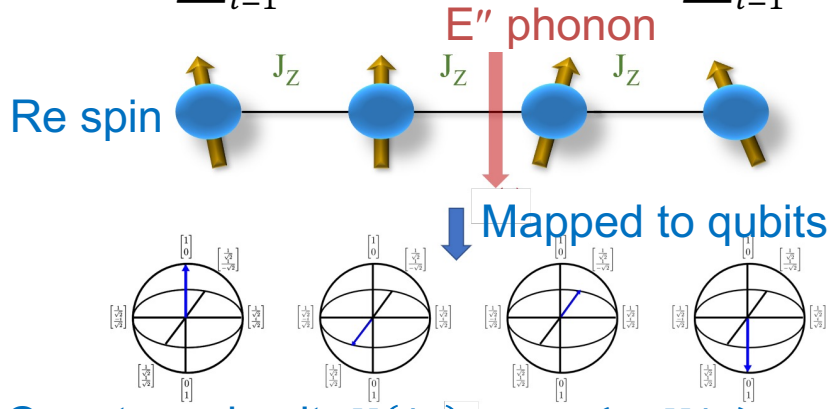
Quantum Computing of Magnetism

- **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"**

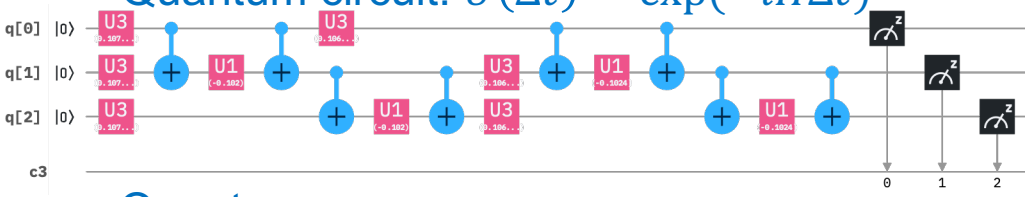




$$H(t) = -J_z \sum_{i=1}^{N-1} \boldsymbol{\sigma}_z^i \boldsymbol{\sigma}_z^{i+1} - \varepsilon_{ph} \sin(\omega_{ph} t) \sum_{i=1}^{N} \boldsymbol{\sigma}_x^i$$



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



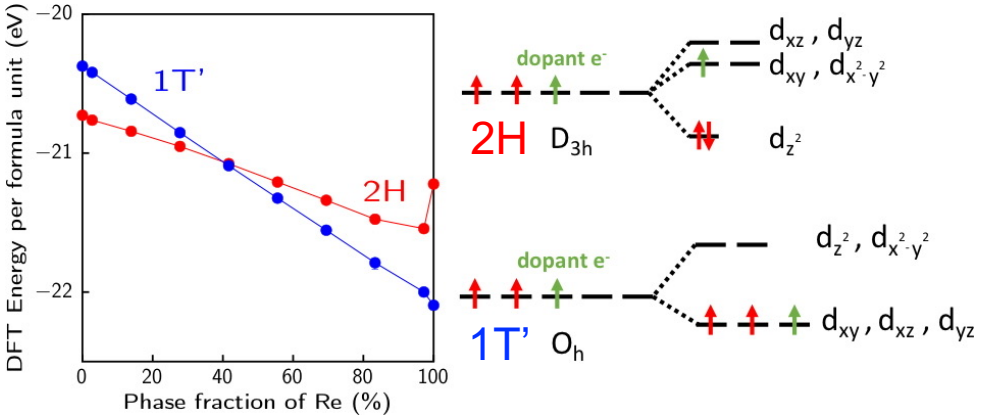
Quantum program

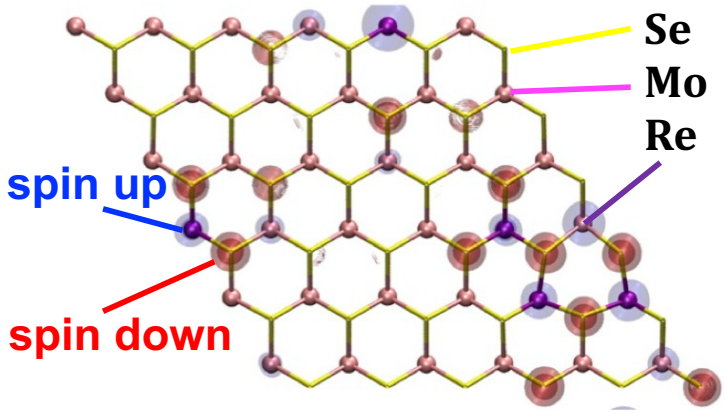
```
32  ····#define·the·two·non-commuting·terms·that·comprise·the·Hamiltonian¬
33  ····Hz·=·PauliTerm("Z",·0,·epsilon_0)¬
34  ····Hy·=·PauliTerm("Y",·0,·epsilon_ph*np.sin(w_ph*t))¬
35  ····#exponentiate·the·terms·of·the·Hamiltonian·for·use·in·Trotter·approx¬
36  ····exp_Hz·=·exponential_map(Hz)(delta_t/(2.0*hbar))¬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

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

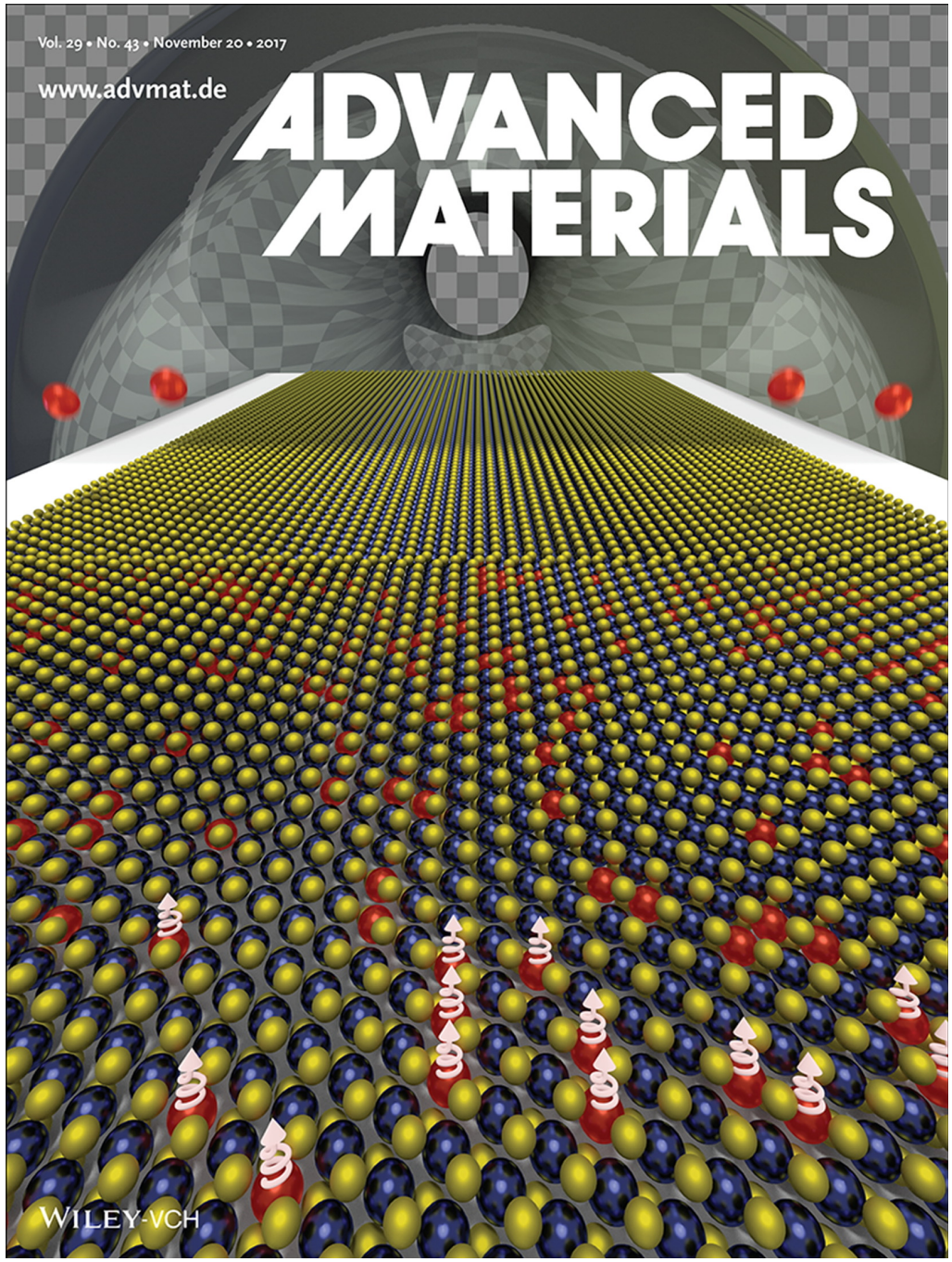
Semiconductor-to-Metal Transition *via* Doping

- **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**





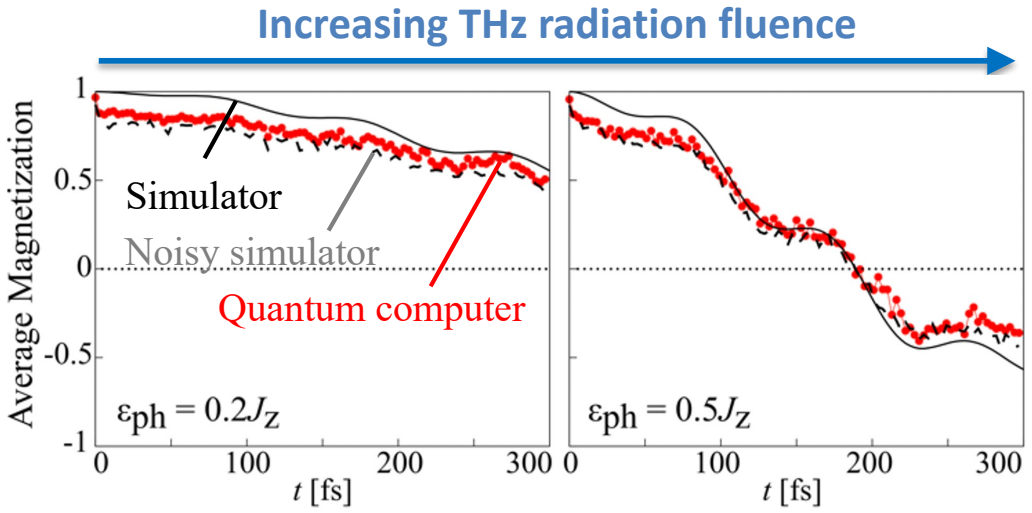
V. Kochat *et al.*, *Adv. Mater.* **29**, 1703754 ('17)



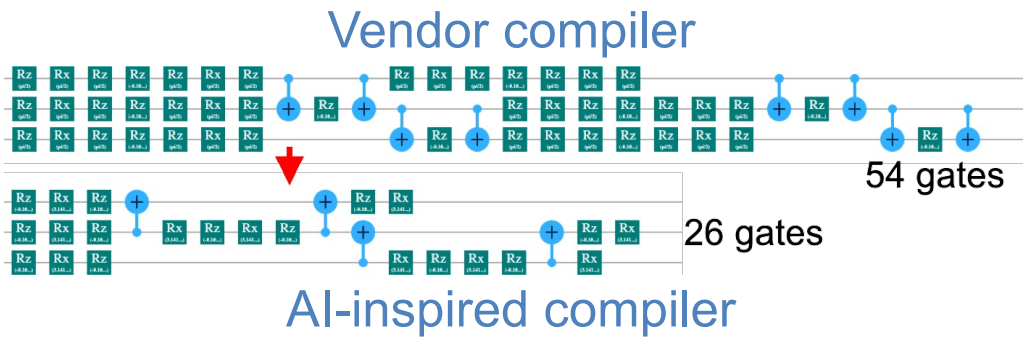
Quantum Dynamics on Quantum Computers

- **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**

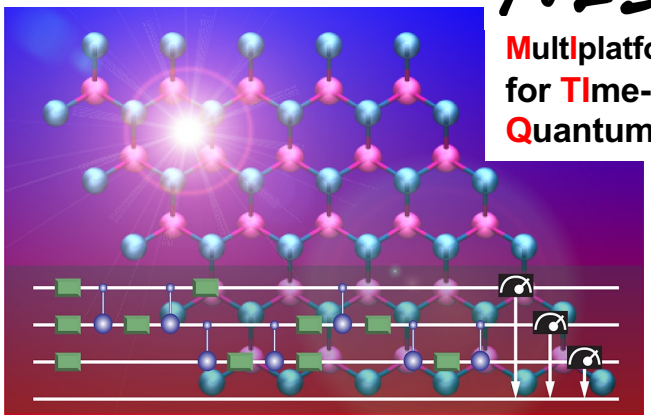


Bassman *et al.*,
Quantum Sci. Tech. **6**, 014007 ('21)

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

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

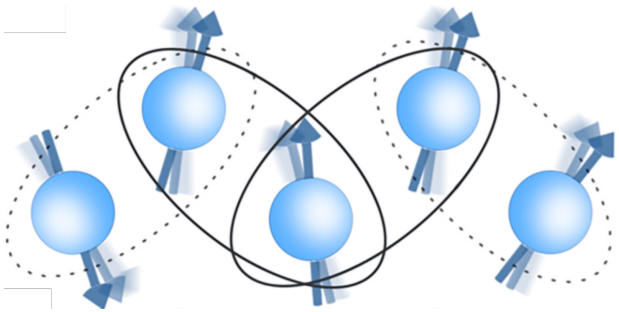
MISTIQS
Mult**I**platform **S**oftware for **TI**me-dependent **Q**uantum **S**imulation



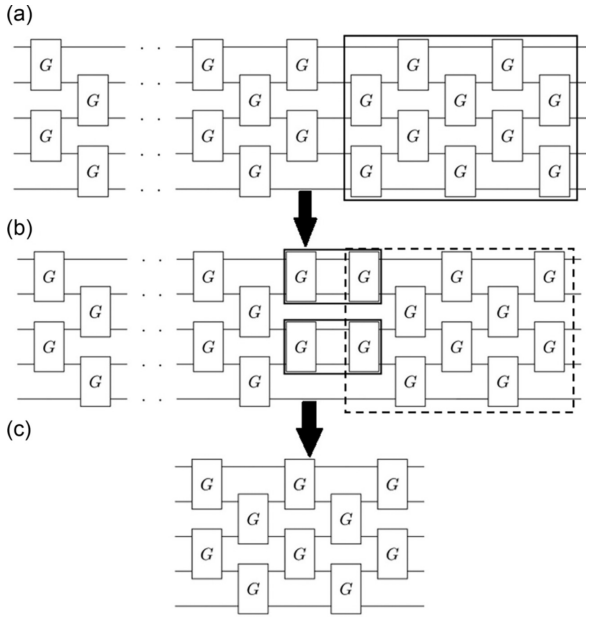
Powers *et al.*, *SoftwareX* **14**, 100696 ('21)
https://github.com/USCCACS/MISTIQS

Topological Quantum Dynamics

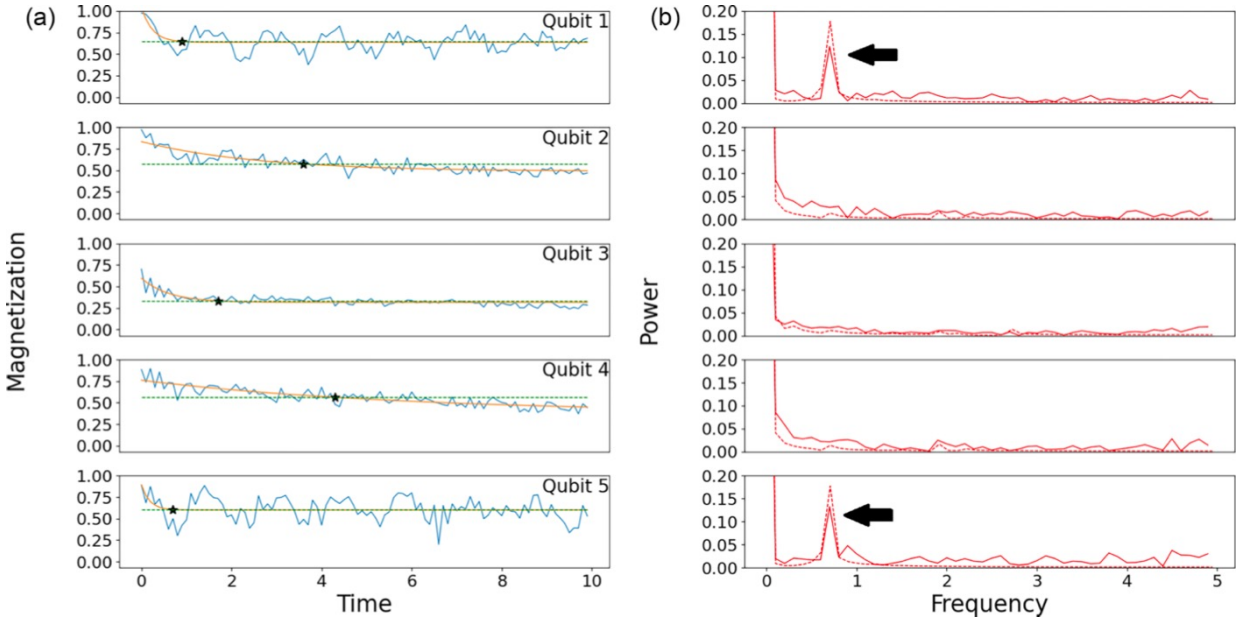
Quantum spin chain



Constant-depth quantum circuit

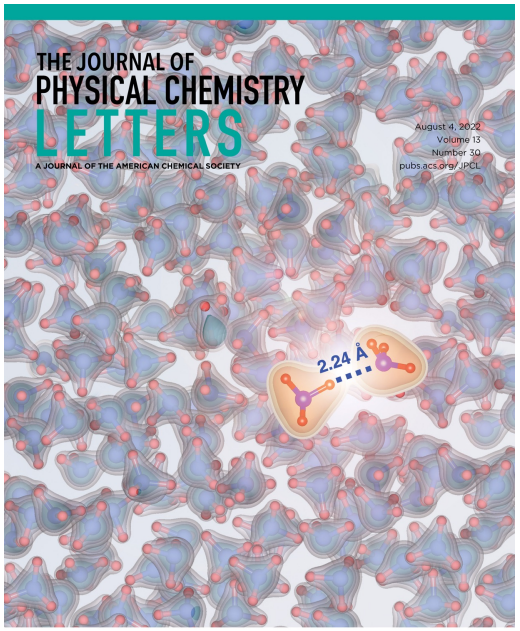


Topological surface mode

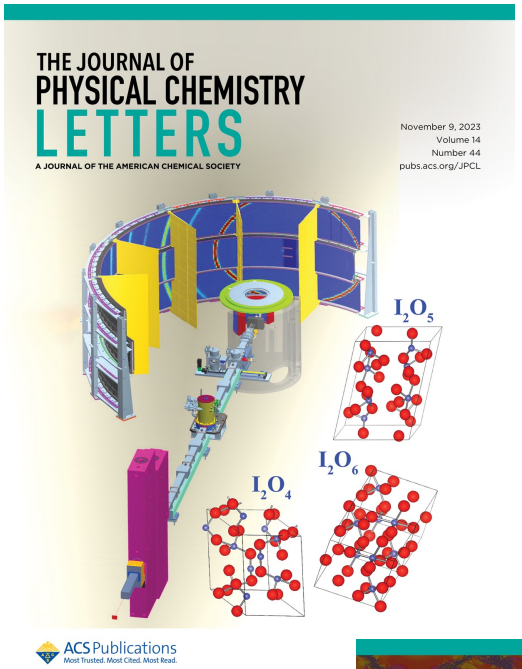


M. Mercado et al., *Phys. Rev. B* **110**, 075116 ('24)

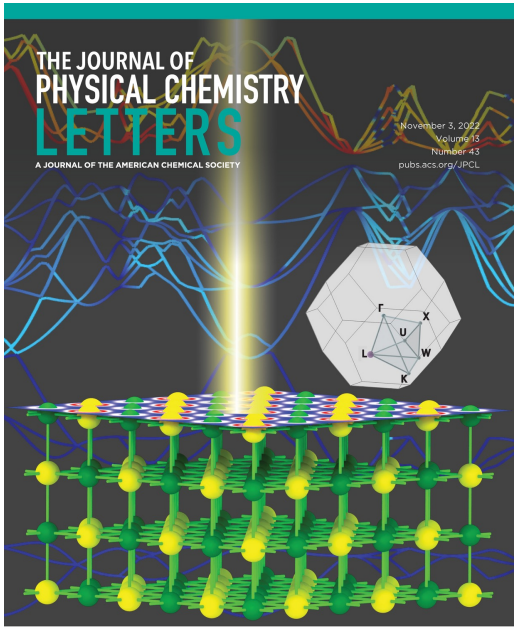
Other Applications



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

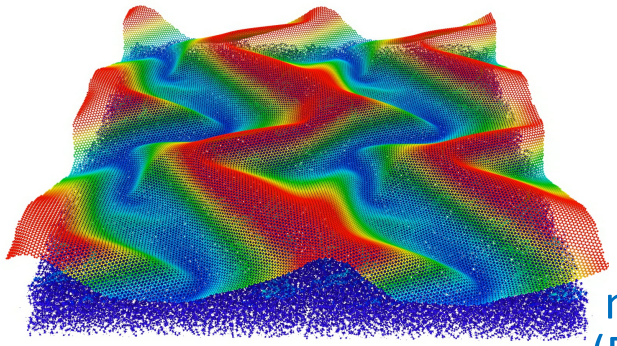


Neutron scattering (Nov. ’23)

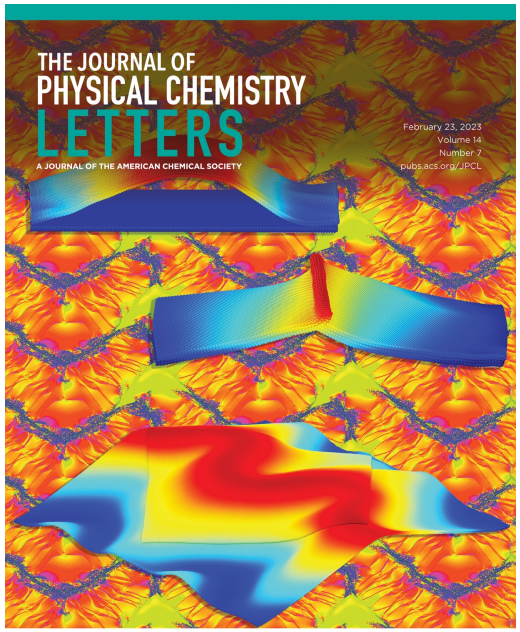


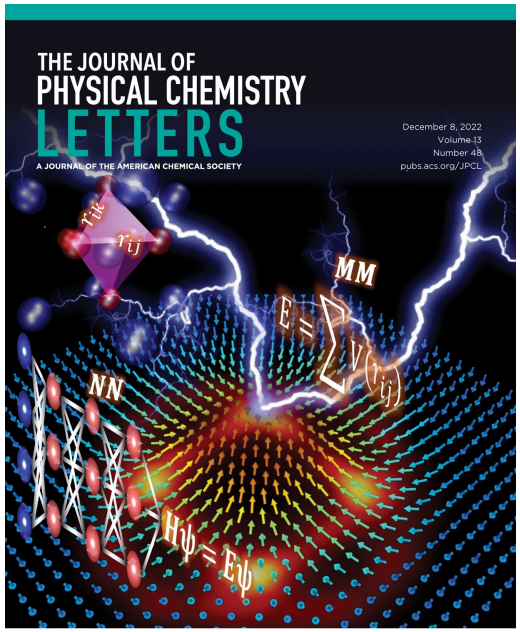
Phase-change memory (Nov. 3, ’22)

Topotronics (Dec. 8, ’22)



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

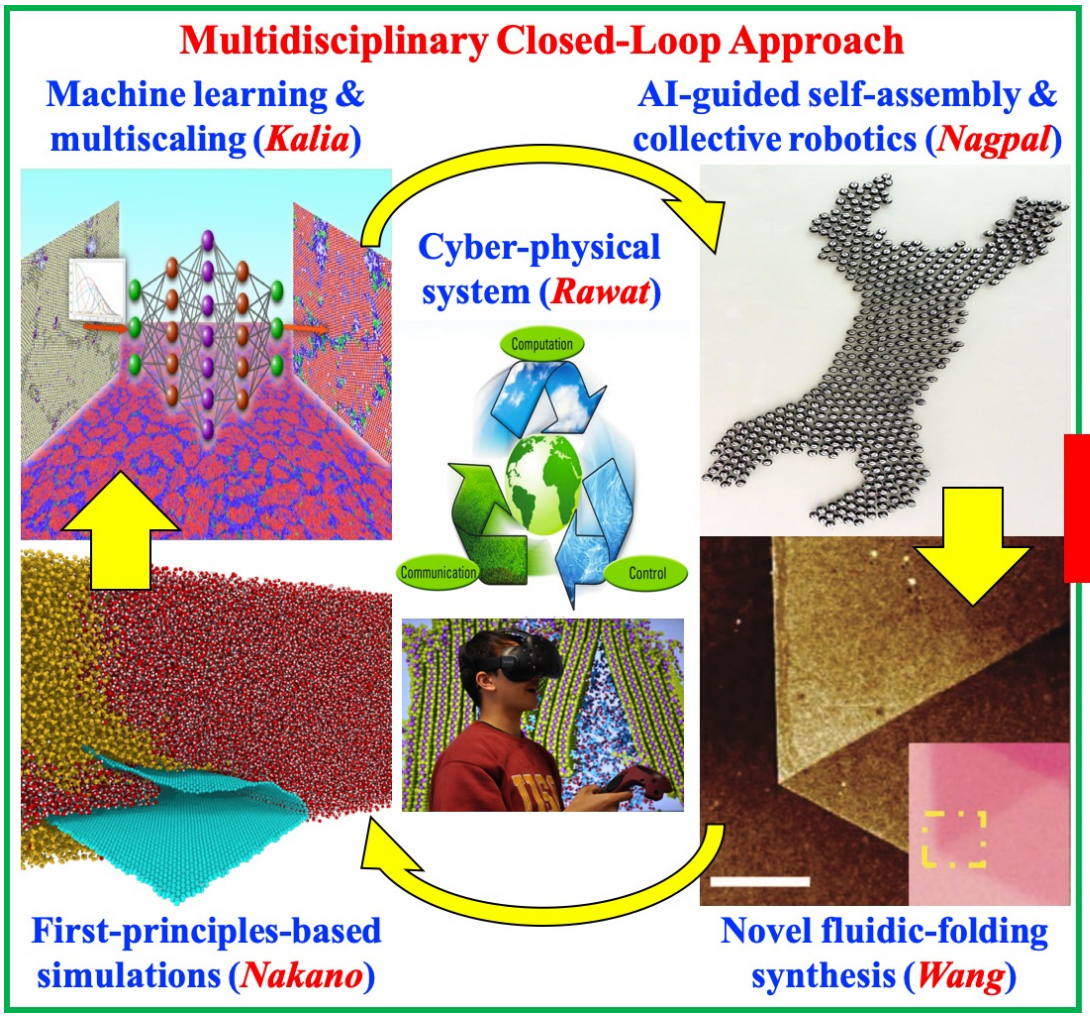


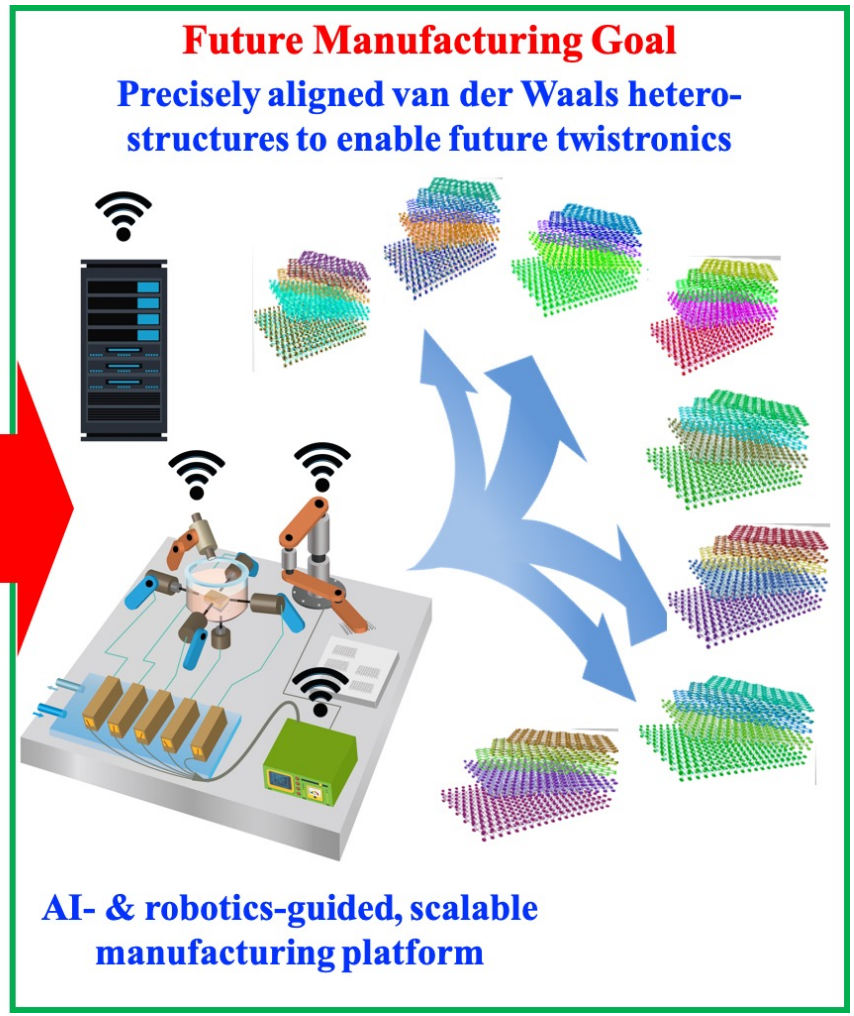


USC-Howard Future Manufacturing

FMRG: Artificial Intelligence Driven Cybermanufacturing of Quantum Material Architectures

$3.75M NSF project (2020-2025)





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

NSF Future Semiconductors Project

- **Identify atomistic & electronic mechanisms of emerging attoJoule (aJ) in-sensor neuromorphic computing without external power:**

  1. Protonic synapse that is deterministic & high speed with aJ energy consumption
  2. Retinal neurons for in-sensor image computing without external power

Conductivity switch in H-doped WO_3

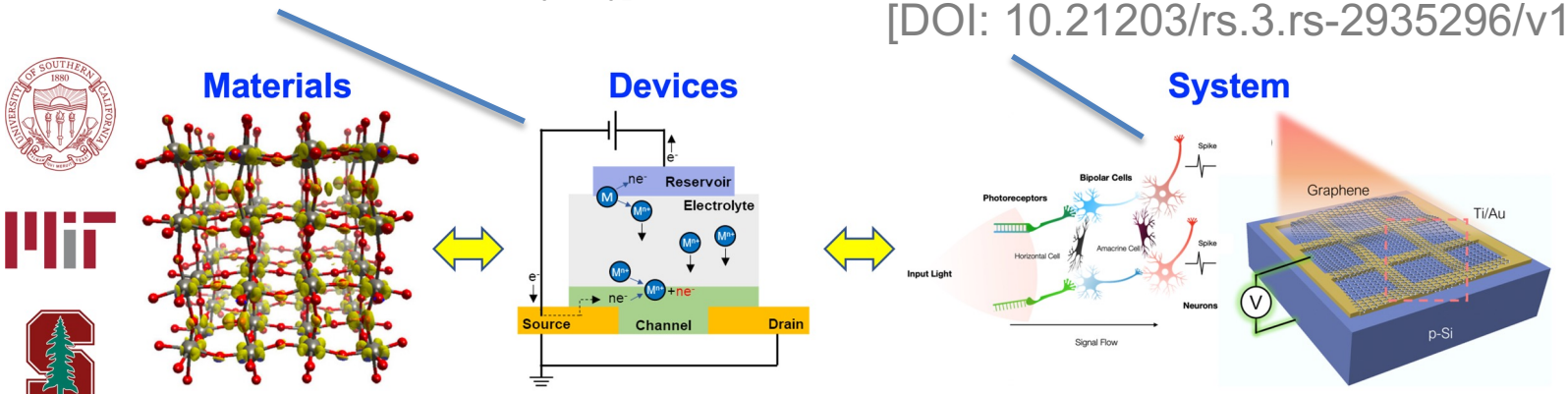
Protonic synapse (MIT)

[Onen *et al.*, *Science* **377**, 539 ('22)]

Photo-induced negative differential resistance (NDR) in graphene/Si

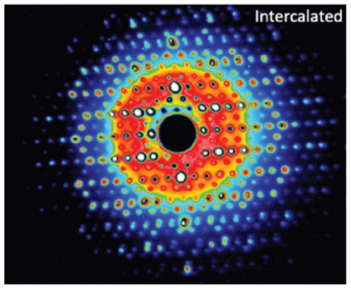
Oscillatory retinal neural network (USC)

[DOI: 10.21203/rs.3.rs-2935296/v1]



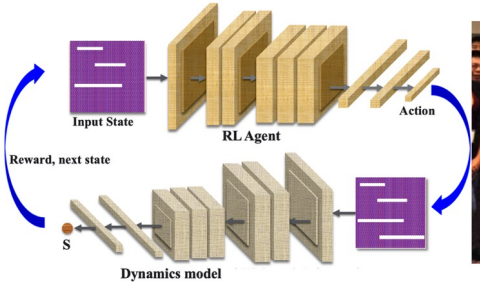
Dev, Krishnamoorthy, Kalia, Li — Wang, Yang, Zhang, Yildiz — del Alamo, Kapadia, Lin, Raina

UE/XS & AI/CS guided co-design and FeSe workforce support



UE/XS

Lindenberg, Wei



AI/CS

Vashishta, Levi, Nakano, Nomura

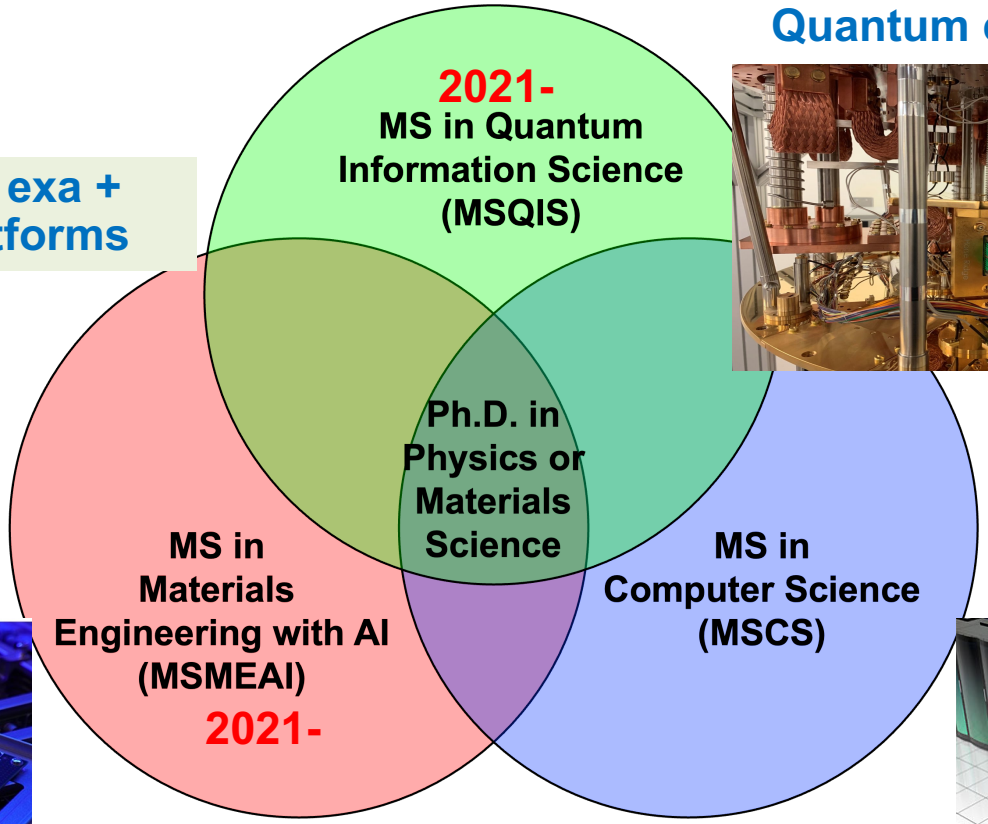
FuSe Workforce Development

Nikias, Katehi, Raghavendra, Rawat

Training Cyber-Science Workforce

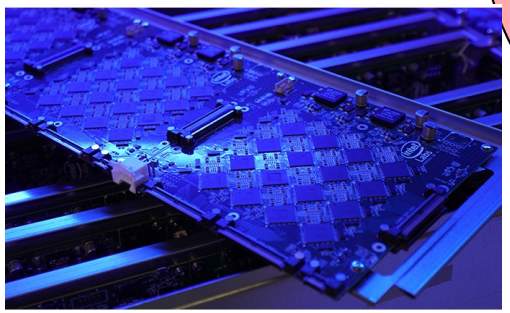
- New generation of computational scientists at the nexus of exascale computing, quantum computing & AI
- Unique dual-degree program: Ph.D. in materials science or physics, along with MS in computer science, MS in quantum information science or MS in materials engineering with AI

Cybertraining on exa + quantum + AI platforms



Horse Ridge II
Quantum computer

Neuromorphic
Pohoiki Springs

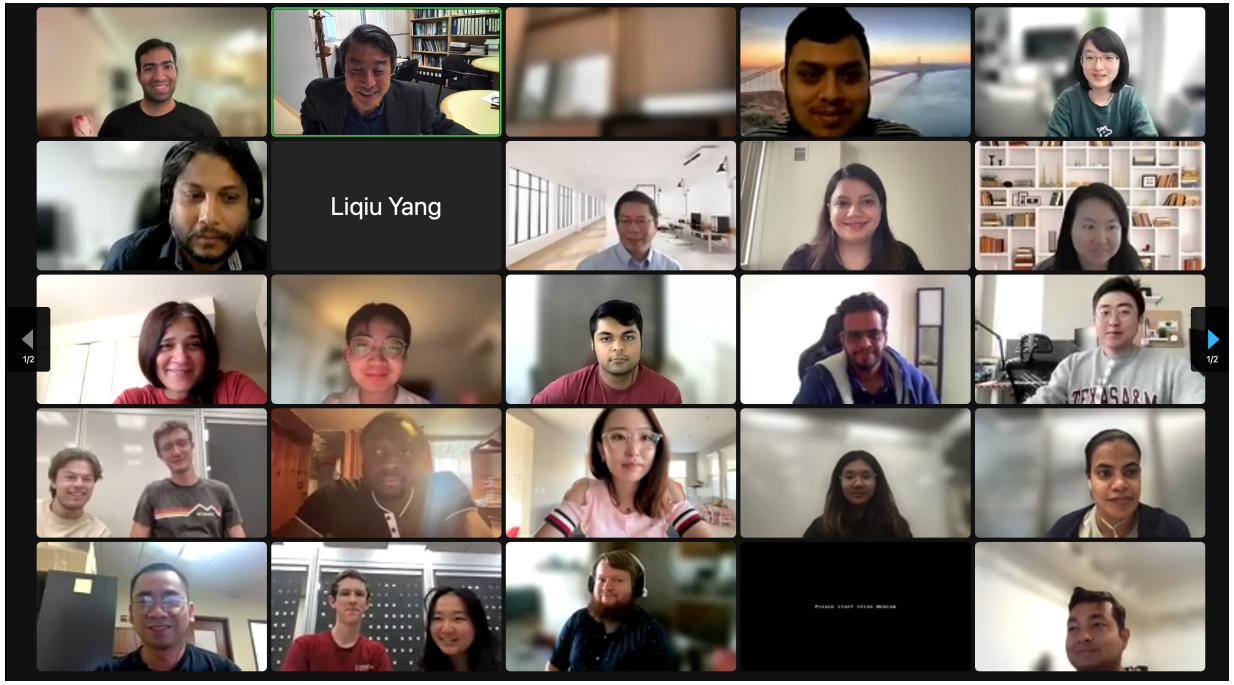


Exascale
Aurora

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**



Second CyberMAGICS workshop (June 29 – July 1, 2023)

$1M NSF CyberTraining (2021-25) project

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

Conclusion

1. **Large spatiotemporal-scale quantum molecular dynamics simulations enabled by divide-conquer-recombine**
2. **Broad materials & energy applications**