

# 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

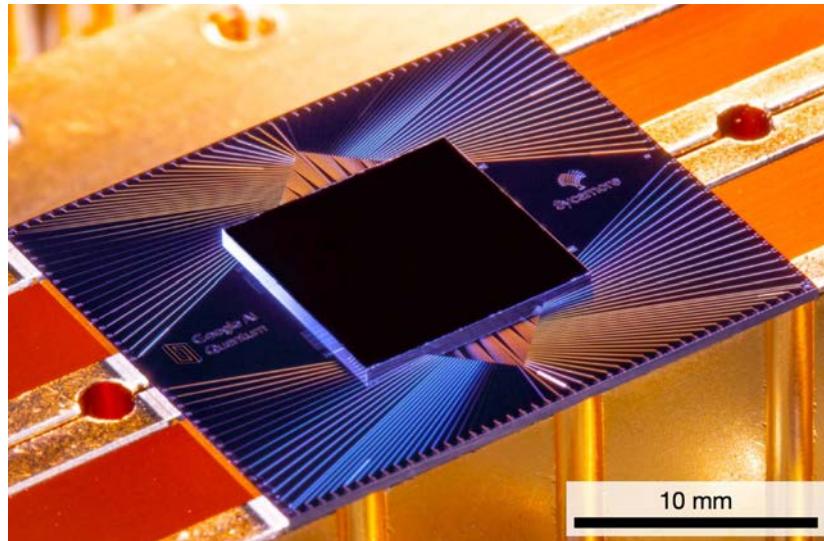
## Post-exascale Computing for Science



Compute Cambrian explosion



## Quantum Computing for Science



## 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)



Use all to advance science!

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

# USC Frontiers of Computing

USC launches computing into the next frontier

\$1B+ initiative including advanced computation,  
quantum computing, AI and ethics

<https://computing.usc.edu>

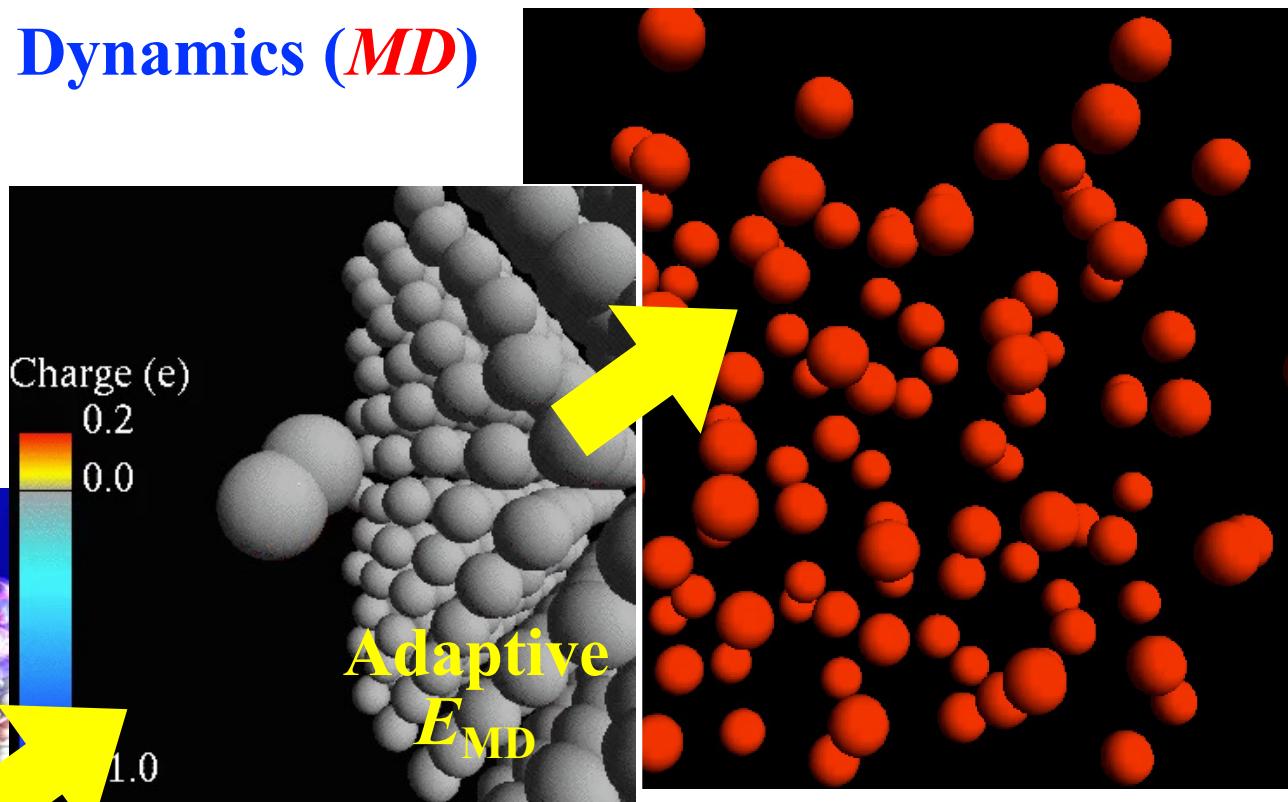
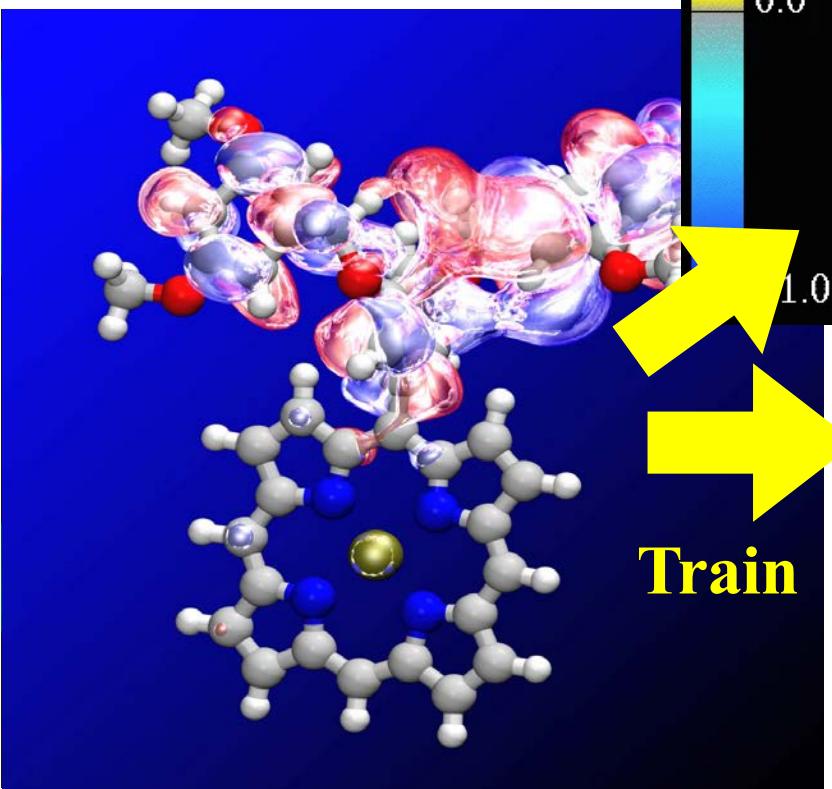
- New School of Advanced Computing: <https://sac.usc.edu>
- 30 senior & 60 junior & mid-level hires

# Molecular Dynamics & Machine Learning

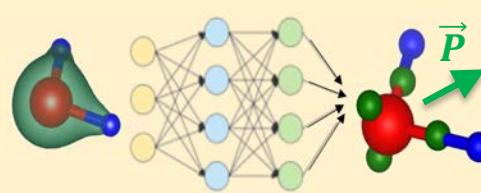
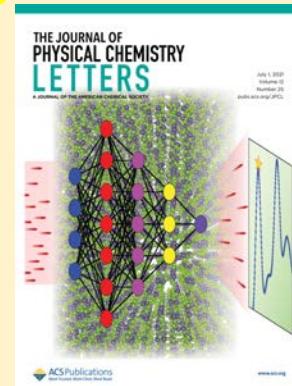
## Molecular Dynamics (*MD*)

### Reactive MD (*RMD*)

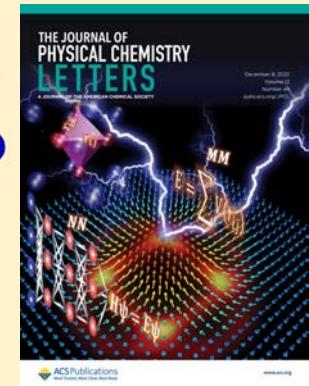
### Nonadiabatic quantum MD (*NAQMD*)



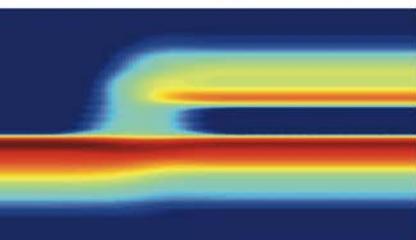
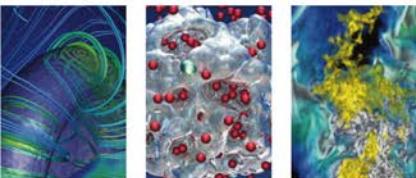
First principles-based neural-network quantum molecular dynamics (*NNQMD*)



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



# BES



NOVEMBER 3-5, 2015

ROCKVILLE, MARYLAND

# Exascale

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

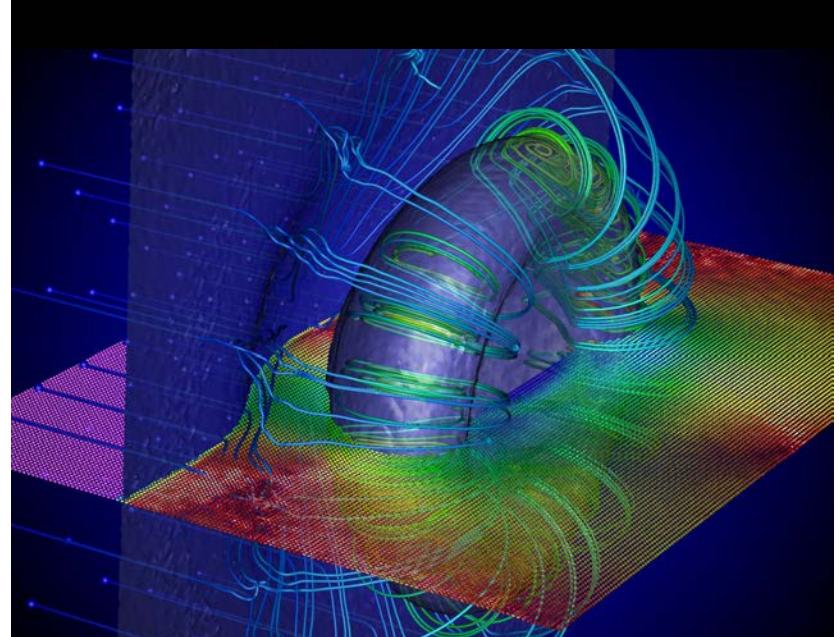
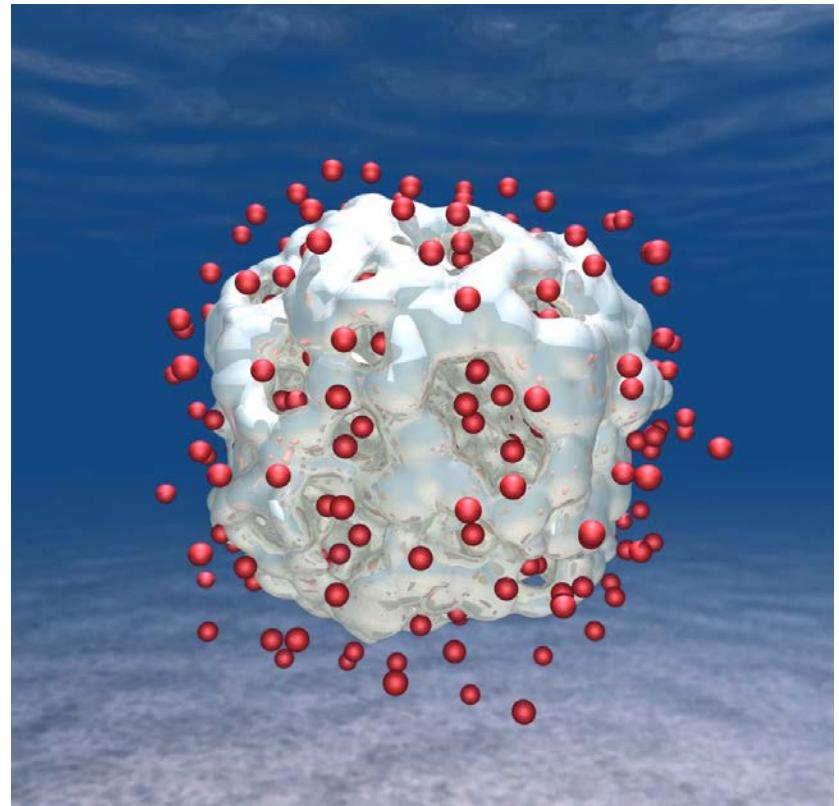
Shimamura *et al.*,  
*Nano Lett.*  
14, 4090 ('14)

*On-demand hydrogen  
production from water*

### 10<sup>9</sup>-atom RMD

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

*Fluid dynamics  
atom-by-atom*



# Quantum Molecular Dynamics (QMD)

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

**First molecular dynamics using an empirical interatomic interaction**

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



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

**Density functional theory (DFT)**

Hohenberg & Kohn, *Phys. Rev.* **136**, B864 ('64)

W. Kohn, *Nobel chemistry prize*, '98

$$\begin{array}{ccc} O(C^N) & \rightarrow & O(N^3) \\ 1 \text{ } N\text{-electron problem} & \text{intractable} & N \text{ 1-electron problems} \\ & & \text{tractable} \end{array}$$

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

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 &  $N_{\text{atom}}$  nuclei, with the Hamiltonian

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

nucleus momentum  
electron position      nucleus position

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

Nº 20

## ANNALEN DER PHYSIK VIERTE FOLGE. BAND 84

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

# Academic Genealogy

## LINDSAY BASSMAN PEDIGREE

### CARL GAUSS

1777 - 1855  
*University of Göttingen*  
Math/Physics 1799

### CHRISTOPHER GUDERMANN

1798 - 1852  
*University of Göttingen*  
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  $\rho(r)$

- 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

Kohn & Vashishta, General Density Functional Theory ('83)



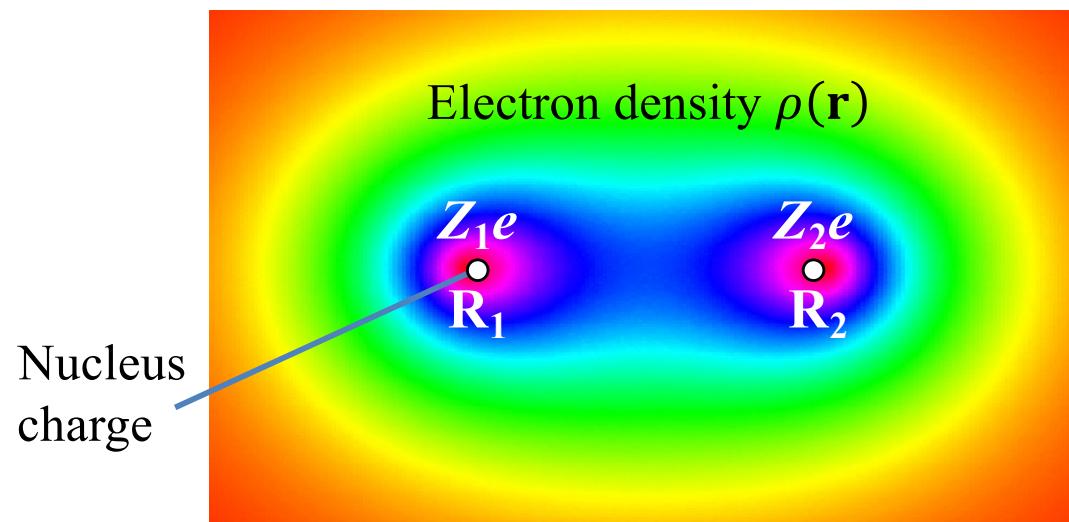
Sholl & Steckel, Density Functional Theory: A Practical Introduction ('23)

# 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

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

KS wave function    KS energy

- KS potential

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

exchange-correlation (xc) potential

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

step function    chemical potential

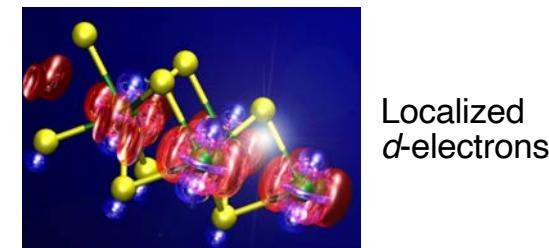
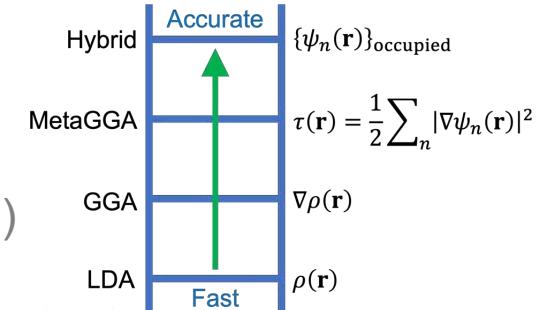
$$v_{\text{xc}}(\mathbf{r}) \equiv \frac{\delta E_{\text{xc}}}{\delta \rho(\mathbf{r})}$$

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

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

$$\delta E_{\text{DFT+U}} / \delta n_i = \epsilon_{\text{DFT}} + U \left( \frac{1}{2} - n_i \right)$$

Anisimov *et al.*, *Phys. Rev. B* **44**, 943 ('91)

## DFT-D: van der Waals (vdW) functional for molecular crystals & layered materials

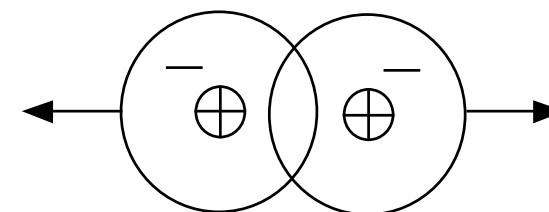
$$E_{\text{disp}} = -s_6 \sum_{i < j} \frac{c_{ij}}{R_{ij}^6} f_{\text{damp}}(R_{ij})$$

Grimme, *J. Chem. Phys.* **132**, 154104 ('10)

## vdW: Nonlocal correlation functional

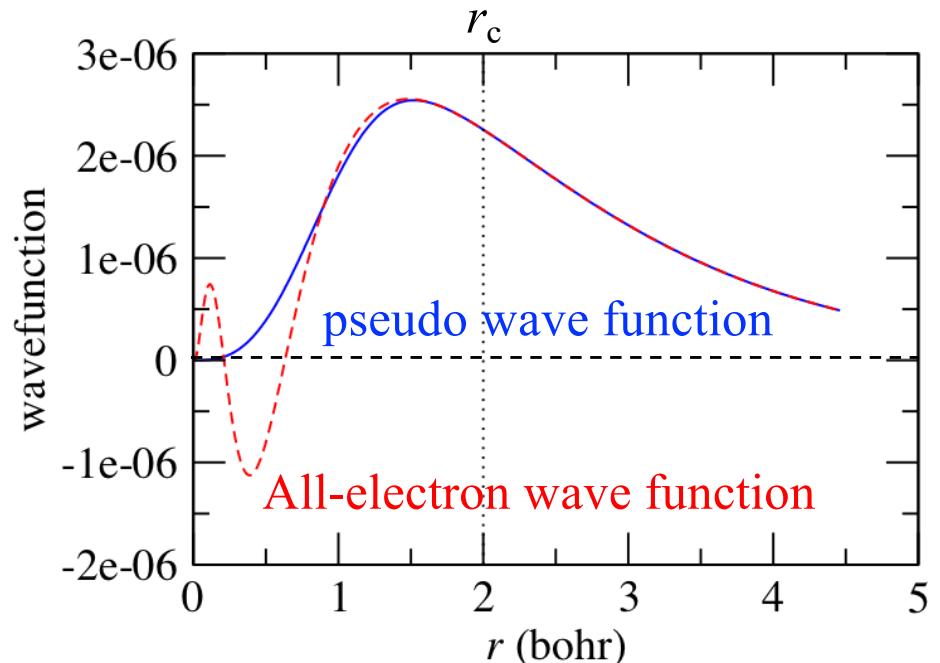
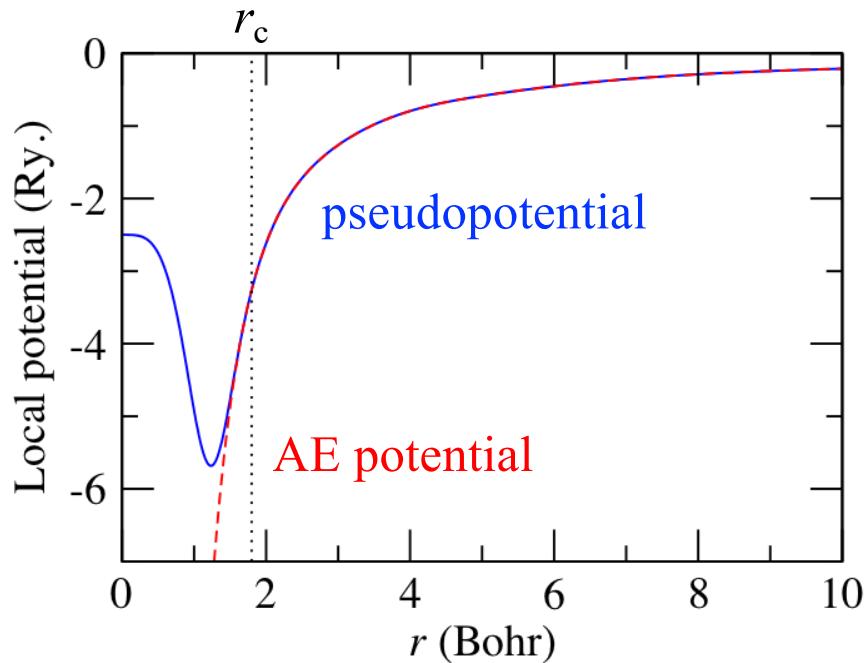
$$E_c^{\text{nl}} = \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \rho(\mathbf{r}) \phi(\mathbf{r}, \mathbf{r}') \rho(\mathbf{r}')$$

Dion *et al.*, *Phys. Rev. Lett.* **92**, 246401 ('04)



# Abstraction: Pseudopotential

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



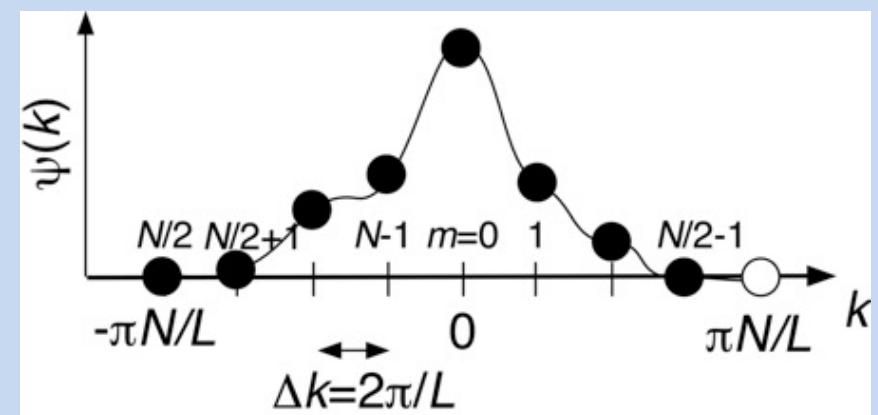
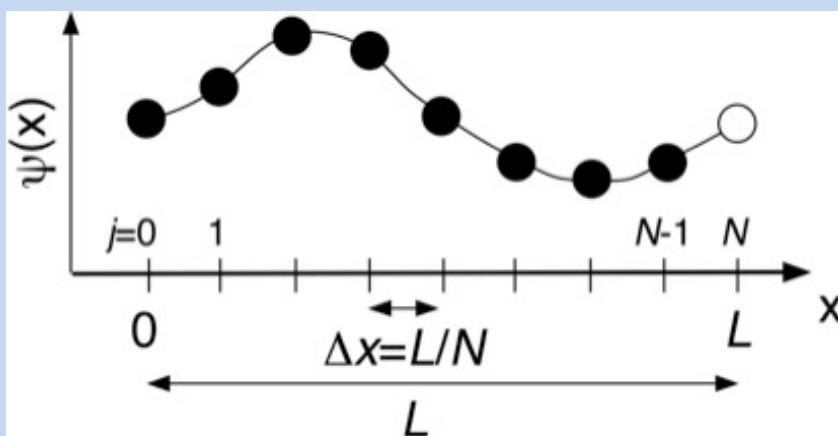
- Commonly used pseudopotentials
  - > Norm-conserving: Troullier & Martins, *Phys. Rev. B* **41**, 1993 ('91)
  - > Ultrasoft: Vanderbilt, *Phys. Rev. B* **41**, 7892 ('90)
  - > Projector augmented wave (PAW): Blochl, *Phys. Rev. B* **50**, 17953 ('94)

# 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

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

1D example

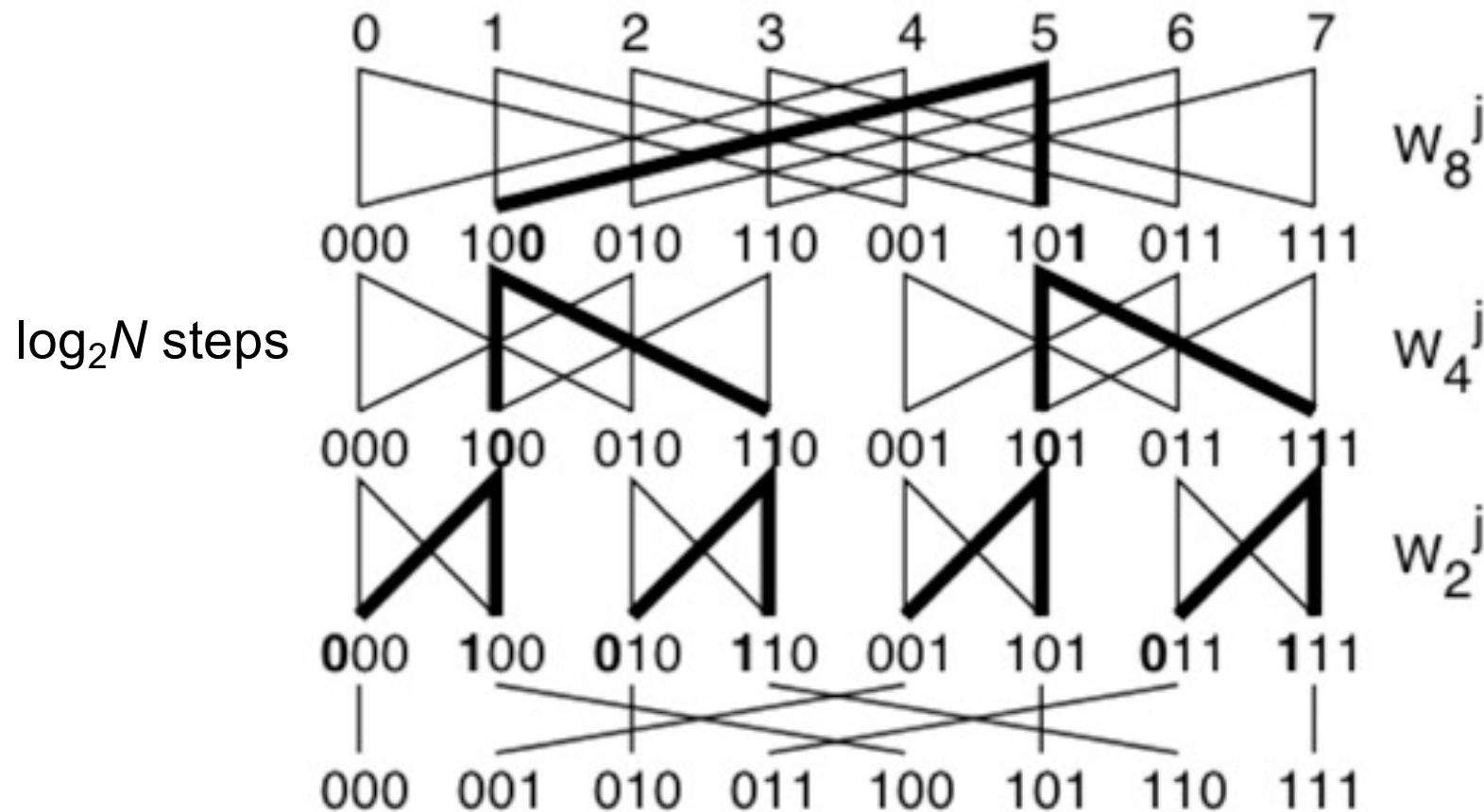


$$x_j = \frac{jL}{N}; \quad k_n = \frac{2\pi n}{L}$$

# Numerics: Fast Fourier Transform

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

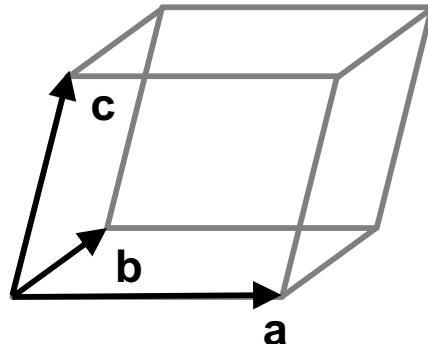
$$\psi(x_j) = \sum_{k_n} \psi_{k_n} \exp(i k_n x_j)$$



Butterfly (hypercube) data-exchange network

# Periodic Solid

- Consider a periodic solid with the unit cell spanned by vectors  $\mathbf{a}$ ,  $\mathbf{b}$  &  $\mathbf{c}$



- Fourier transform of a periodic function

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

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

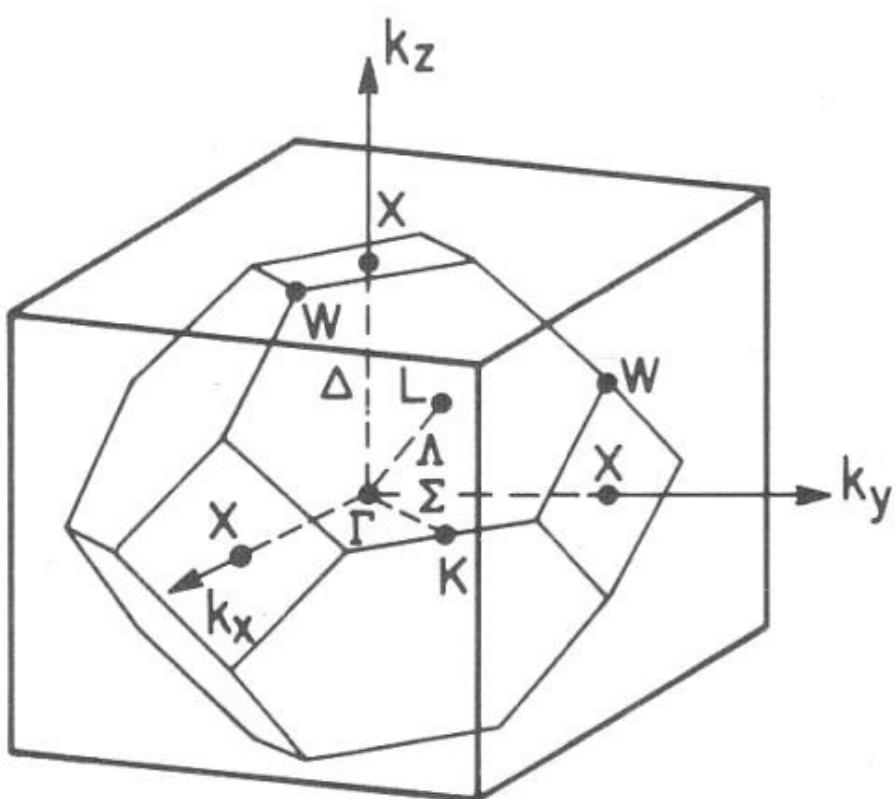
- Bloch's theorem

$$\begin{aligned} \psi_{n\mathbf{k}}(\mathbf{r}) &= \exp(i\mathbf{k} \cdot \mathbf{r}) u_{n,\mathbf{k}}(\mathbf{r}) \\ &= \sum_{\mathbf{G}} u_{n,\mathbf{k}}(\mathbf{G}) \exp(i(\mathbf{k} + \mathbf{G}) \cdot \mathbf{r}) \end{aligned}$$

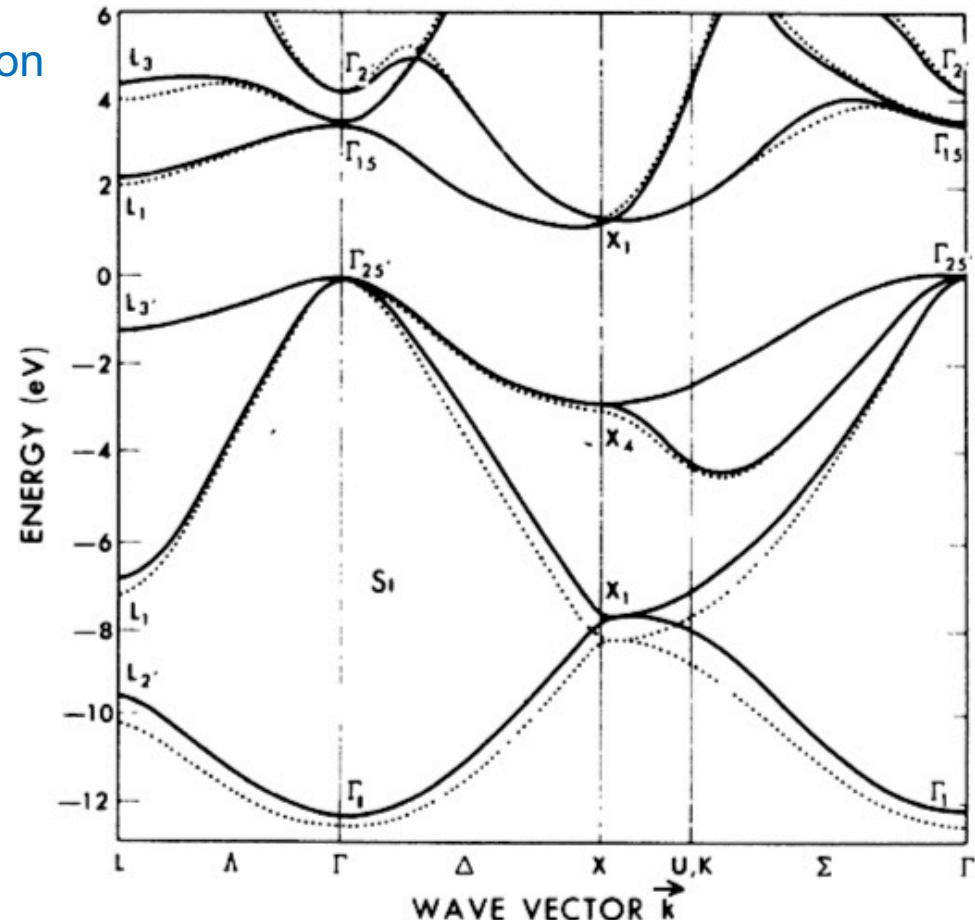
$\mathbf{k} \in$  first Brillouin zone in the reciprocal space

# Electronic Bands: Infinite Lattice

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



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

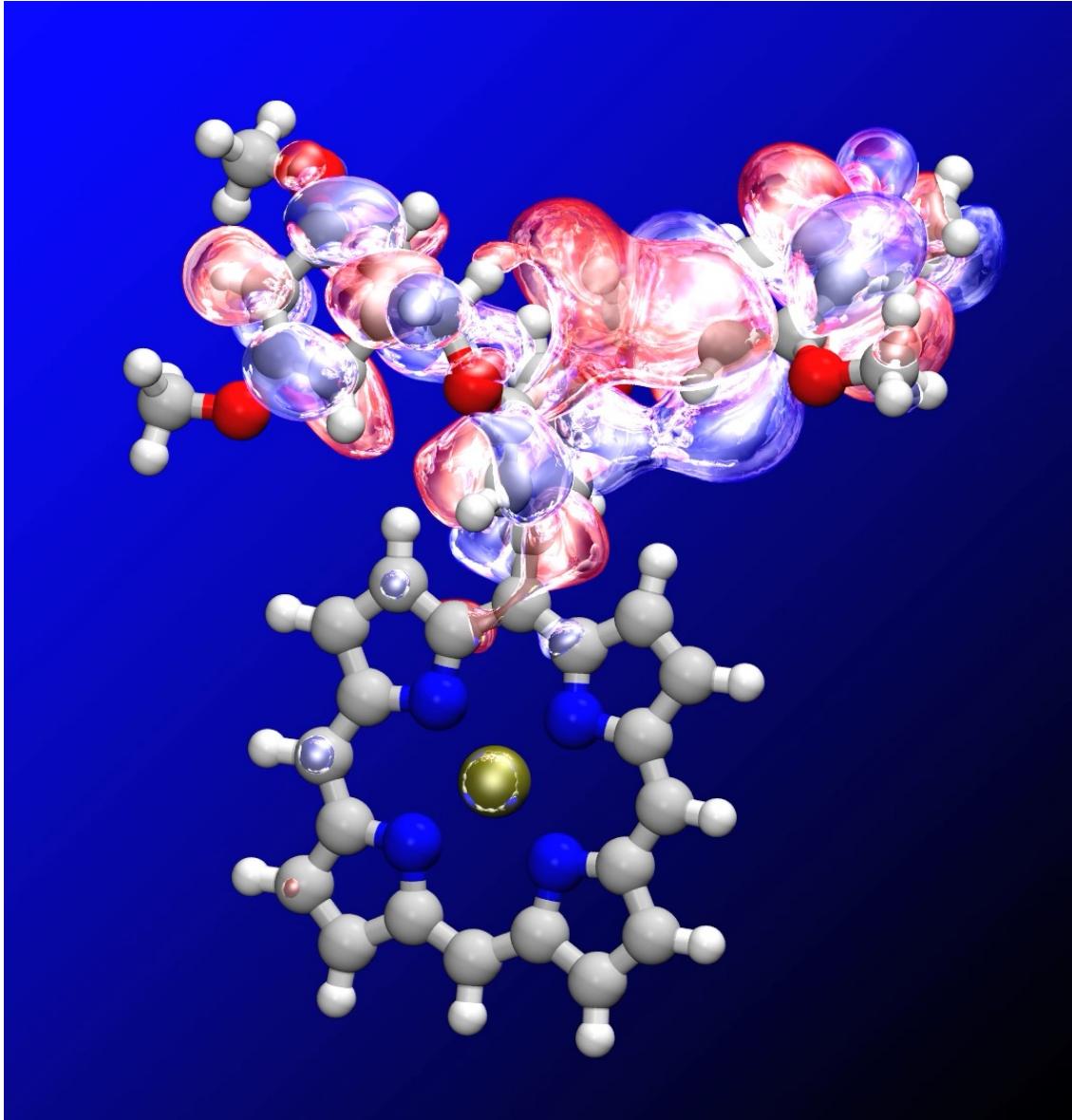
Given  $\{\psi_n, \epsilon_n\}$ ,  
determine  $\mu$  and  
compute  $\rho(\mathbf{r})$

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

Chemical potential

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

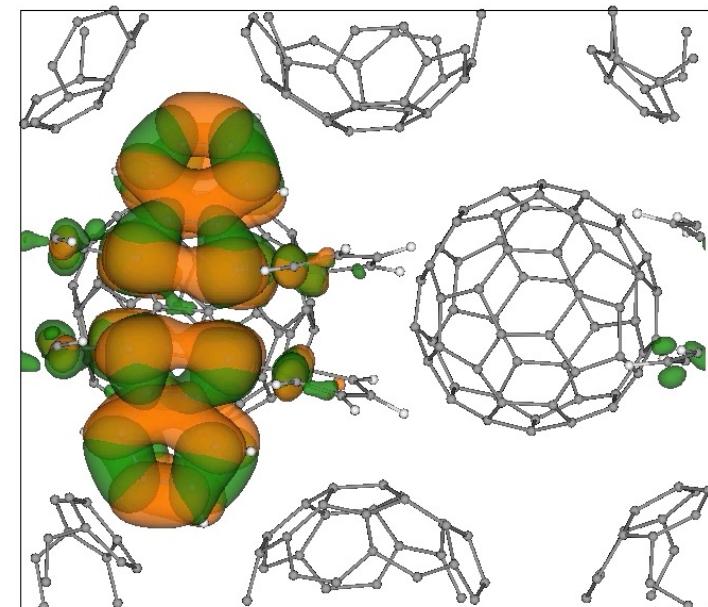
# Nonadiabatic Quantum Molecular Dynamics



*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<sub>60</sub>



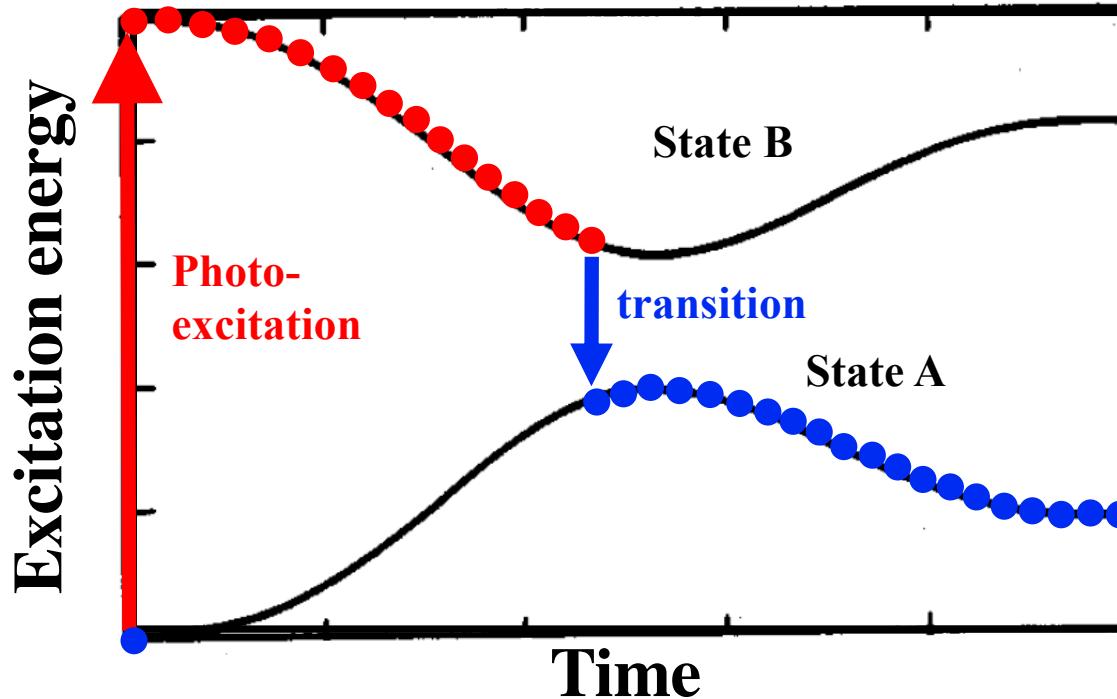
quasi-electron; quasi-hole

- **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

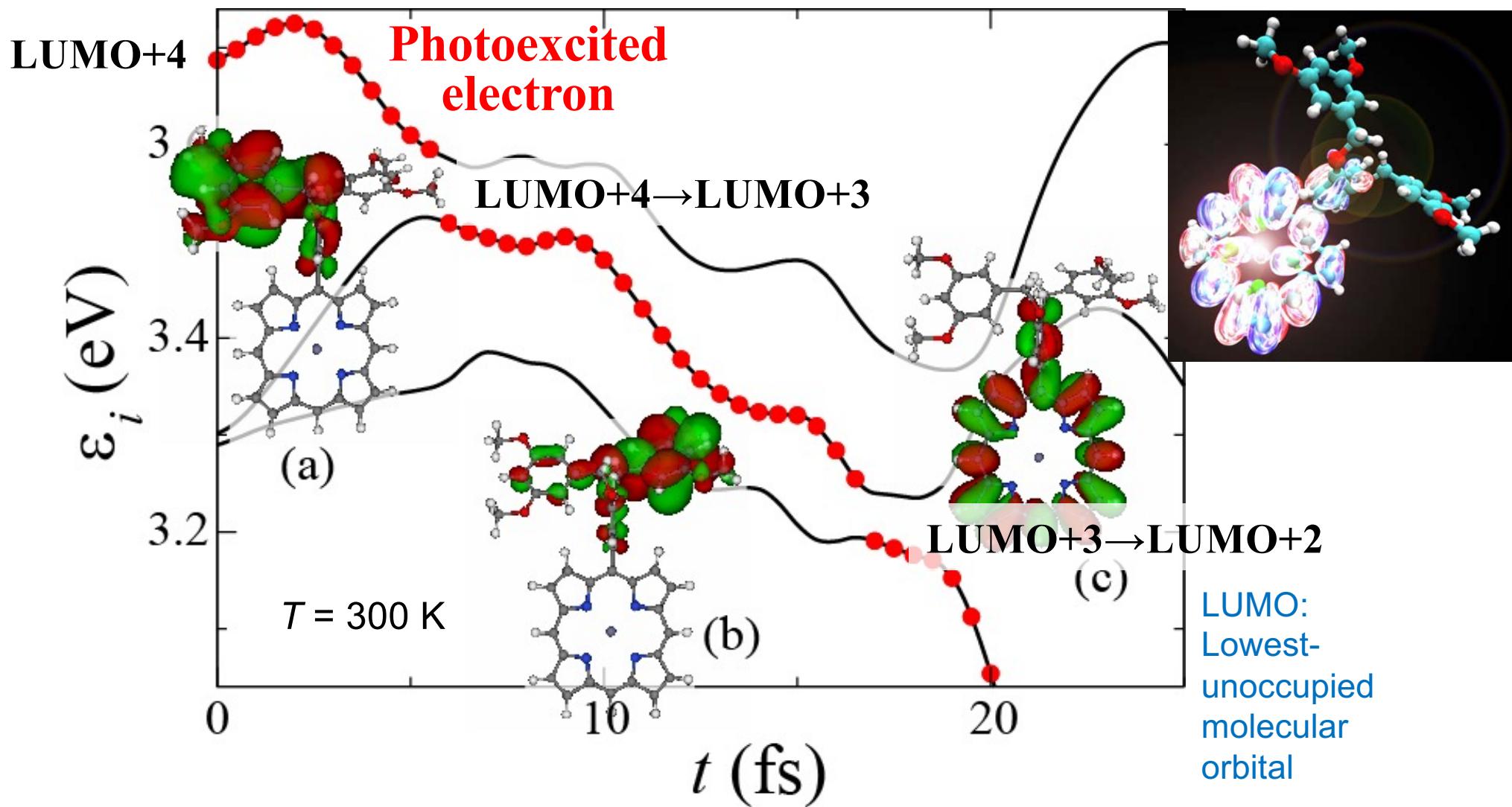
$K$ -th excitation frequency

$$\Psi(\mathbf{r}, t) = \sum_J C_J^{(I)}(t) \Phi_J(\mathbf{r}; \mathbf{R}(t)) \quad C_I^{(I)}(0) = \delta_{I,J}$$
$$\frac{d}{dt} C_J^{(I)}(t) = - \sum_k C_k^{(I)}(t) \left( i\omega_K \delta_{JK} + \langle \Phi_J | \frac{\partial}{\partial t} | \Phi_K \rangle \right)$$

$J$ -th adiabatic excited state

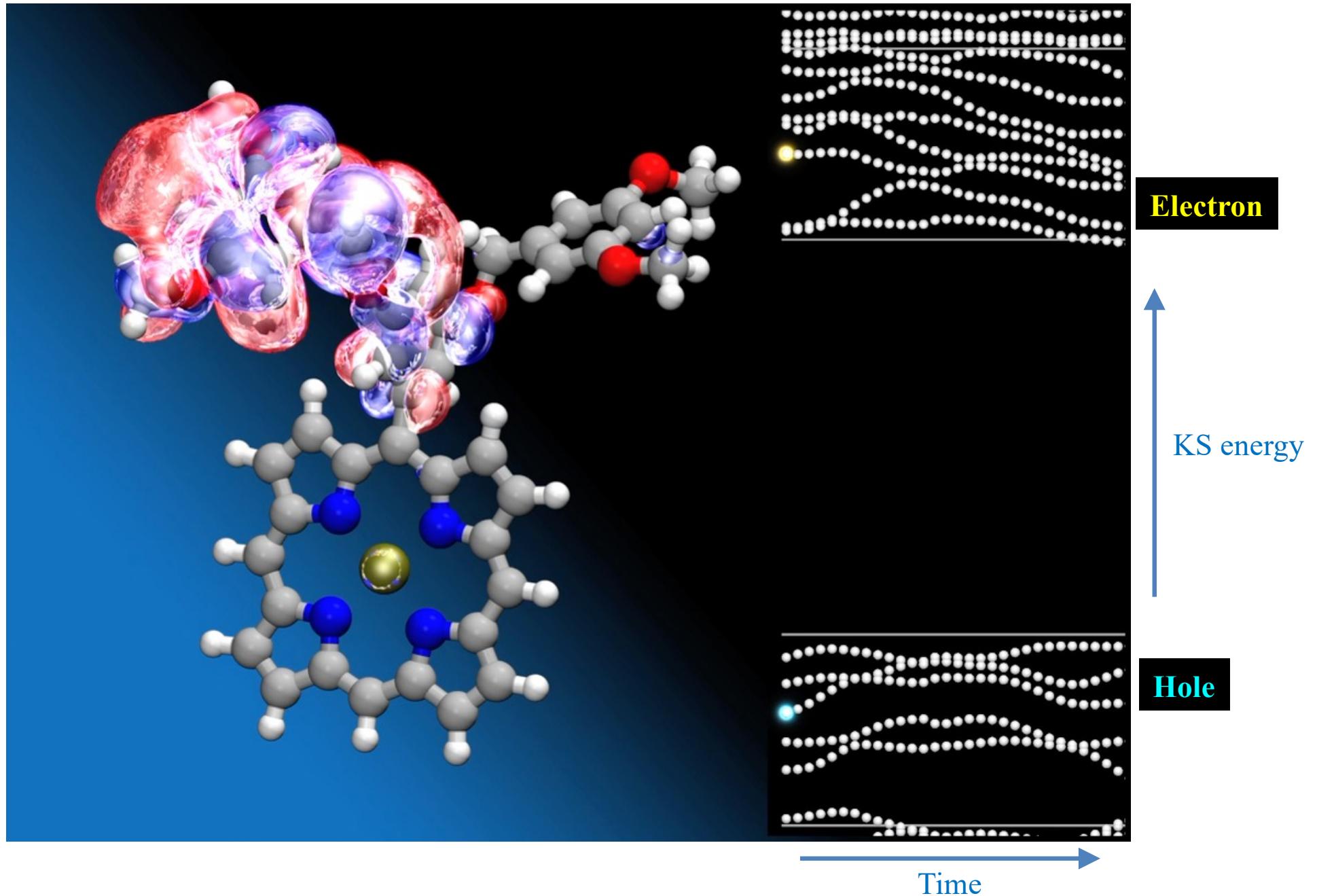
Electronic transition assisted by nuclei motion

# Example: Electron Transfer in a Dendrimer



- 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]

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

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

electron    hole

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

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

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

*I-th excitation energy*

Kohn-Sham energy

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

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

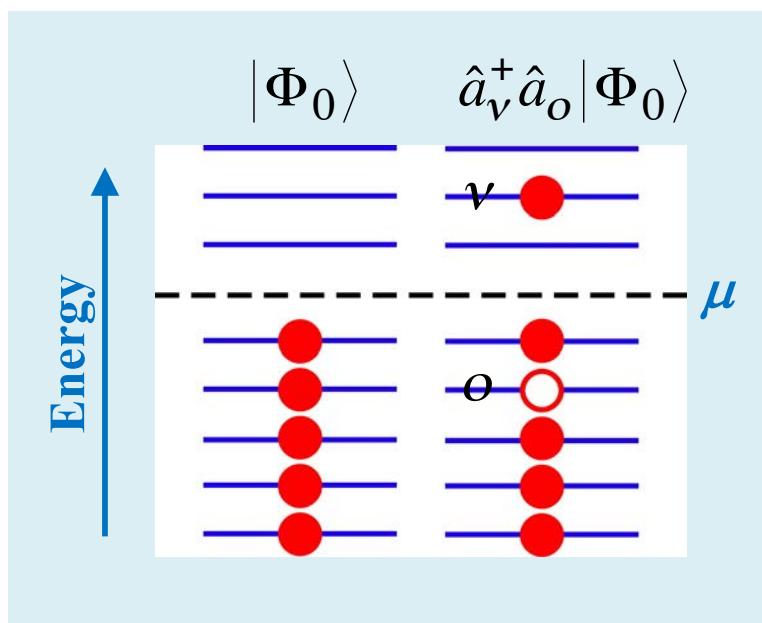
Coulomb & exchange-correlation interaction matrix elements

# Electronic Excited States

- $I$ -th excited state

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

electron-hole pair      ground state



# QXMD Code

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- 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](https://github.com/USCCACS/QXMD_DEV)

**Software download site:**

<https://cybermagiccs.netlify.app/software>

# Current & Future Supercomputing

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



## Innovative & Novel Computational Impact on Theory & Experiment

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

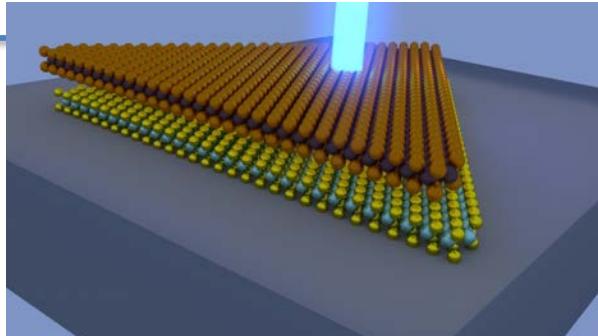
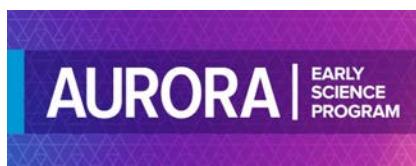
- Atomistic simulations on million cores (pre-exascale)



786,432-core IBM Blue Gene/Q

281,088-core Intel Xeon Phi

560-node (2,240-GPU) AMD/NVIDIA Polaris



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...



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

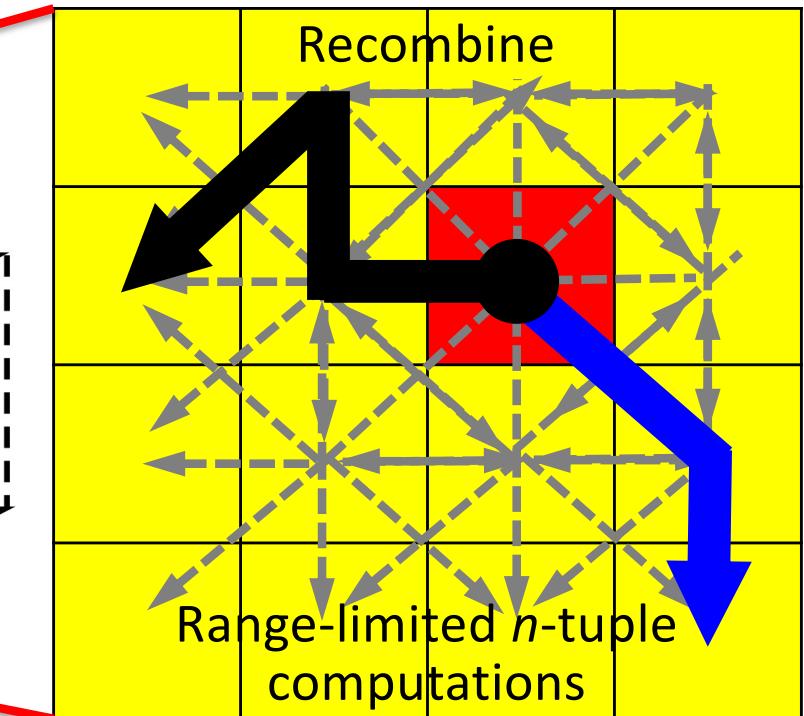
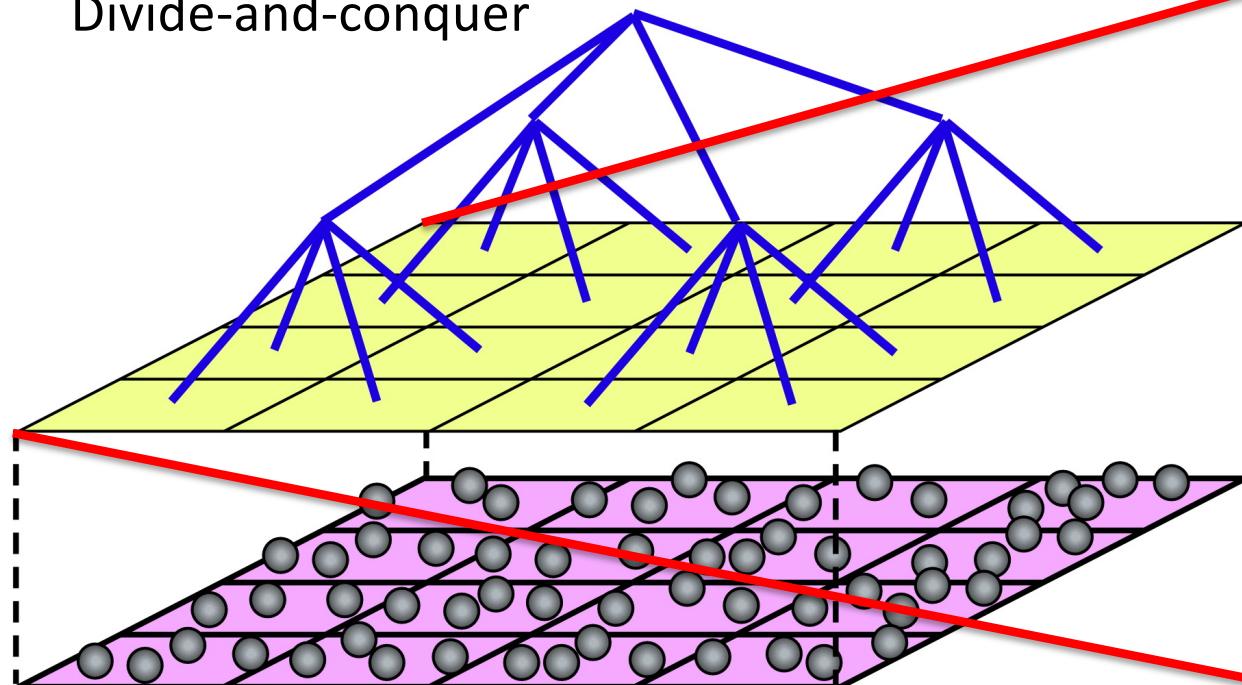
Michael Feldman | November 15, 2017 04:34 CET

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

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

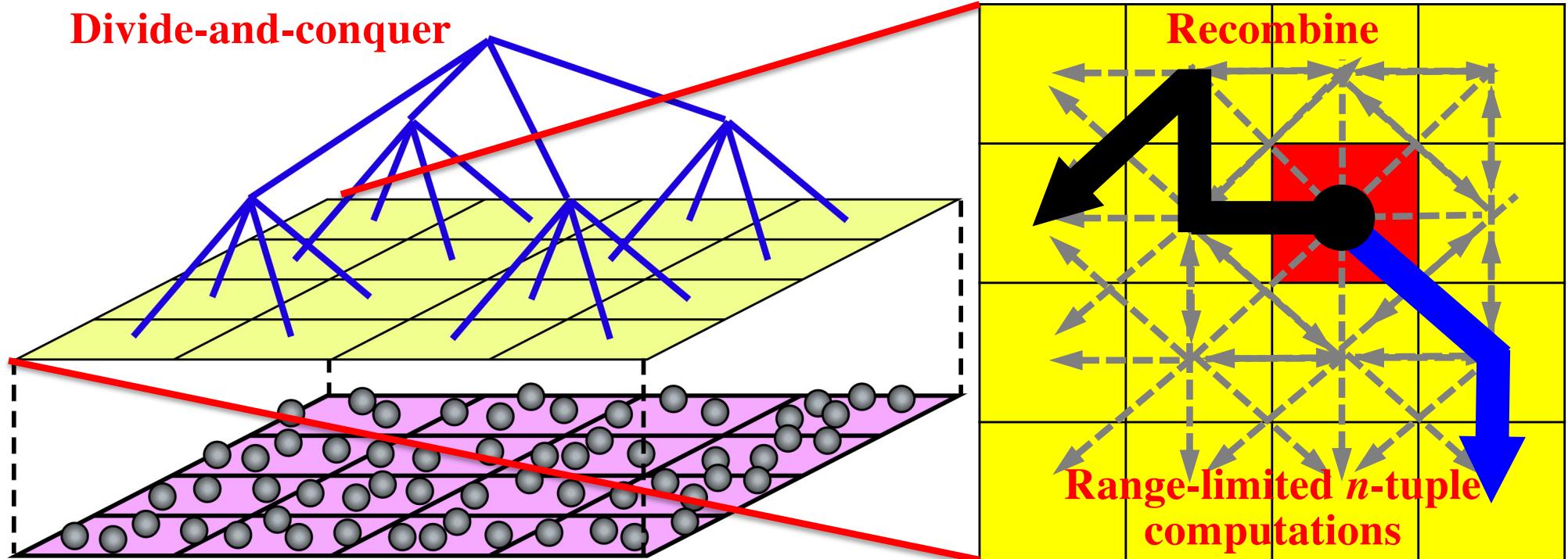
F. Shimojo et al., *J. Chem. Phys.* **140**, 18A529 ('14);  
K. Nomura et al., *ACM/IEEE SC14* ('14)

Divide-and-conquer



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

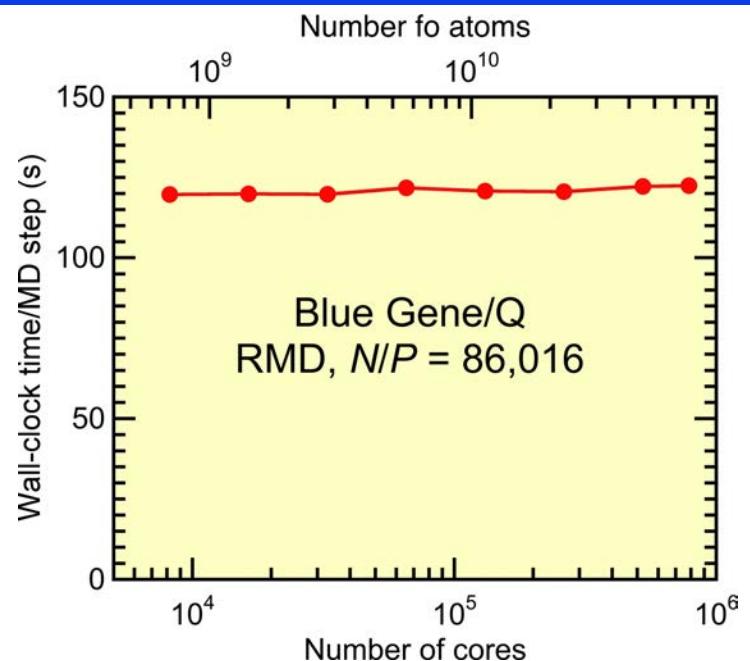
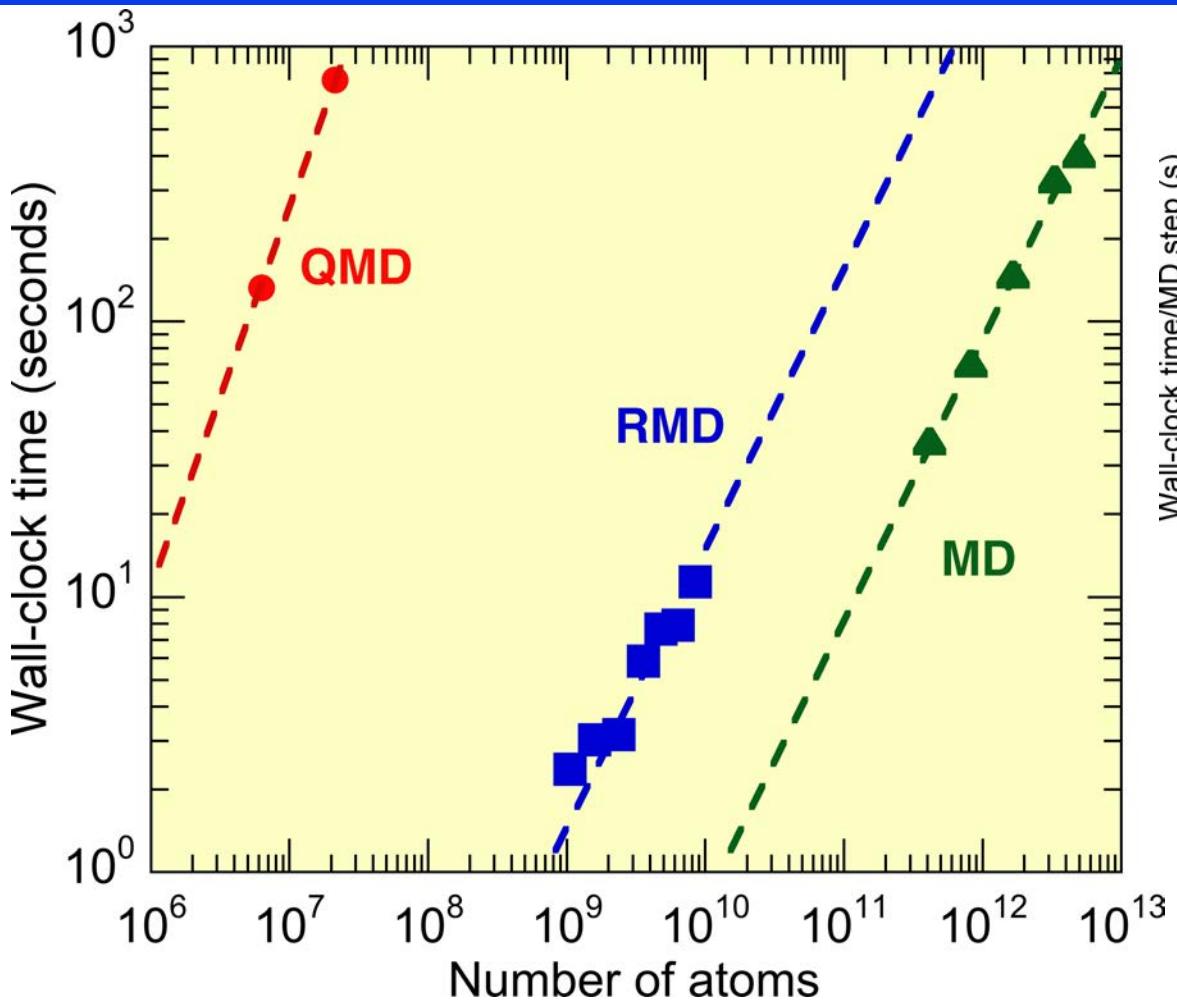
# Divide-Conquer-Recombine (DCR) Engines



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)

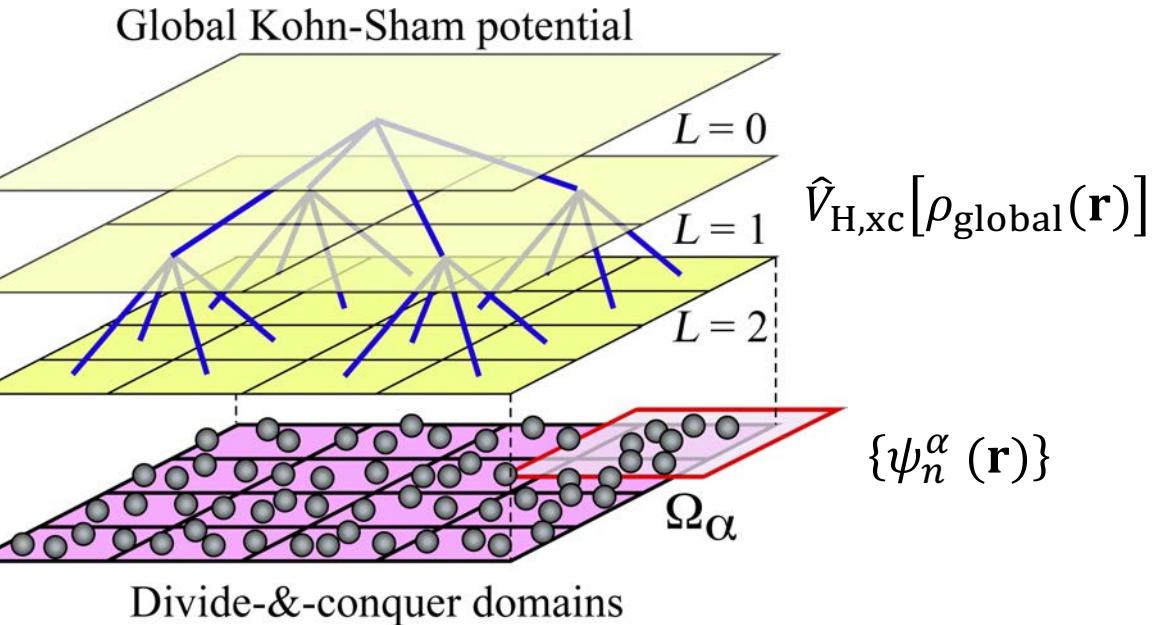
# Scalable Simulation Algorithm Suite



**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  $\text{SiO}_2$
  - 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:  $\Omega = \bigcup_\alpha \Omega_\alpha$
- Domain Kohn-Sham equations

Global-local  
self-consistent  
field (SCF)  
iteration

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

- Global & domain electron densities

$$\rho_{\text{global}}(\mathbf{r}) = \sum_\alpha p_\alpha(\mathbf{r}) \rho_\alpha(\mathbf{r}) \quad \rho_\alpha(\mathbf{r}) = \sum_n [\psi_n^\alpha]^2 \Theta(\mu - \epsilon_n^\alpha)$$

Domain support function

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

Global chemical potential

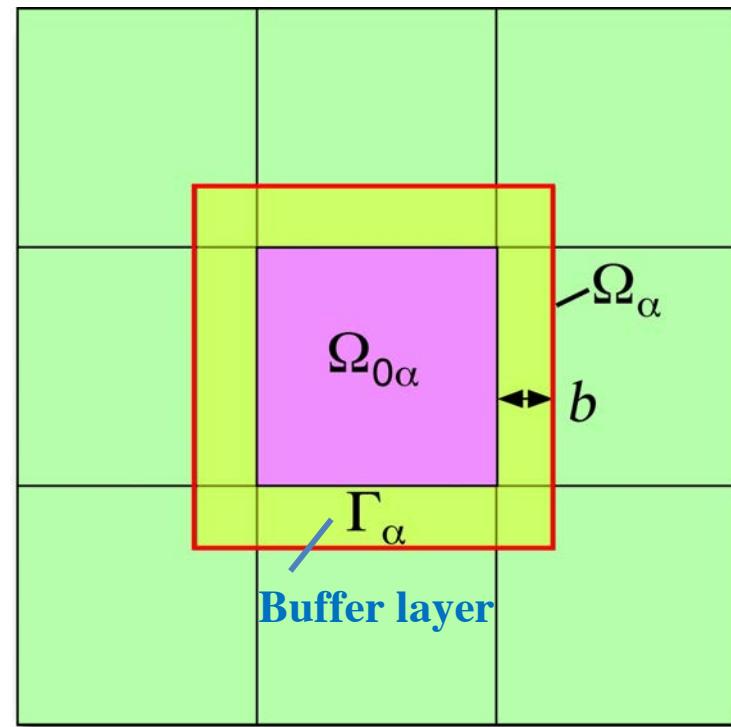
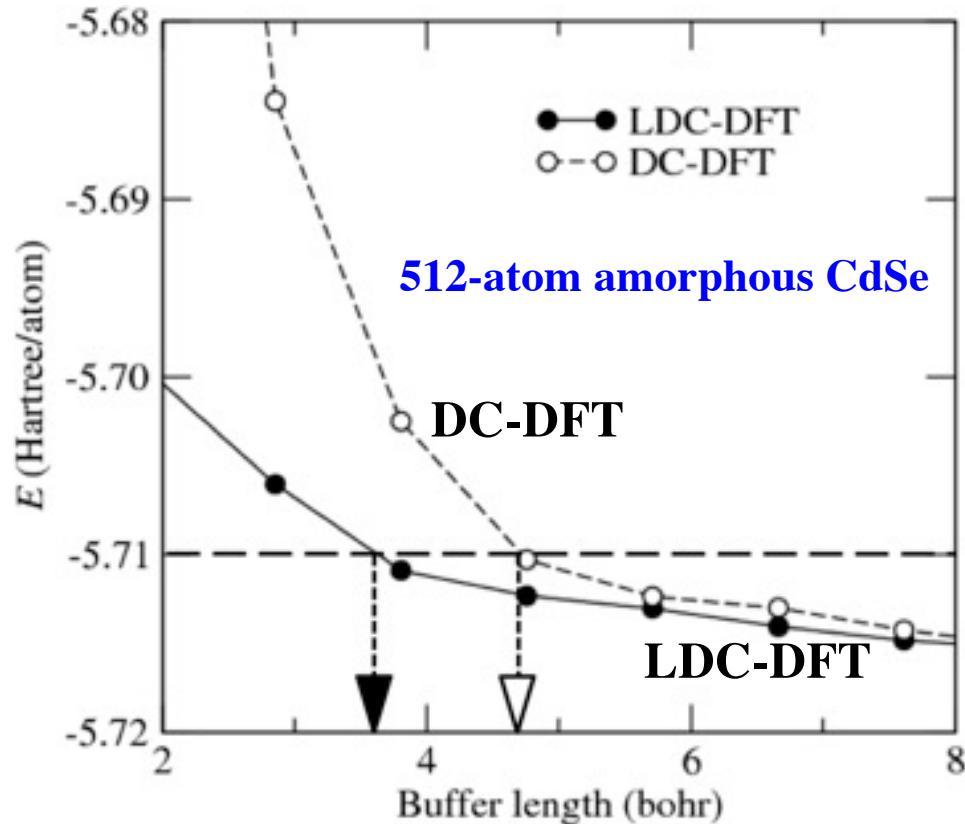
$$N = \int d\mathbf{r} \rho_{\text{global}}(\mathbf{r})$$

# Lean Divide-&-Conquer (LDC) DFT

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

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

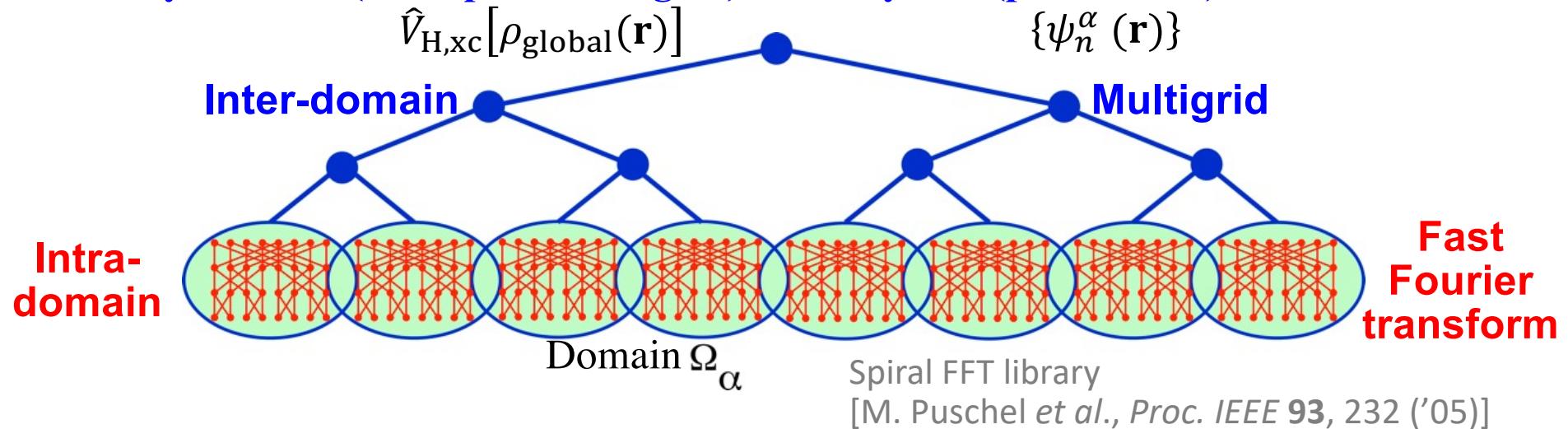
- More rapid energy convergence of LDC-DFT compared with nonadaptive DC-DFT



- Factor 2.03 (for  $\nu = 2$ )  $\sim 2.89$  (for  $\nu = 3$ ) reduction of the computational cost with an error tolerance of  $5 \times 10^{-3}$  a.u. (per-domain complexity:  $n^{\nu}$ )

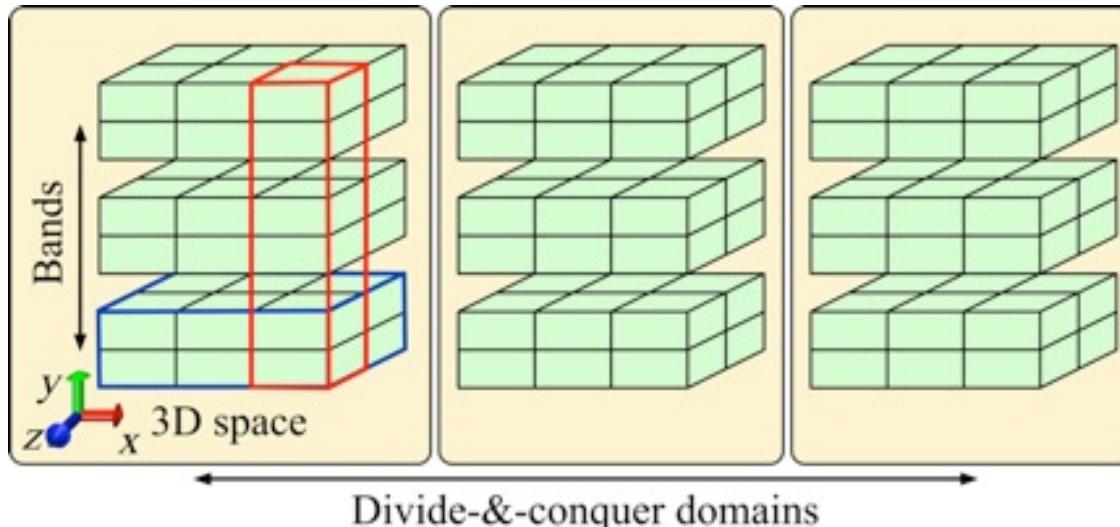
# Hierarchical Computing

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



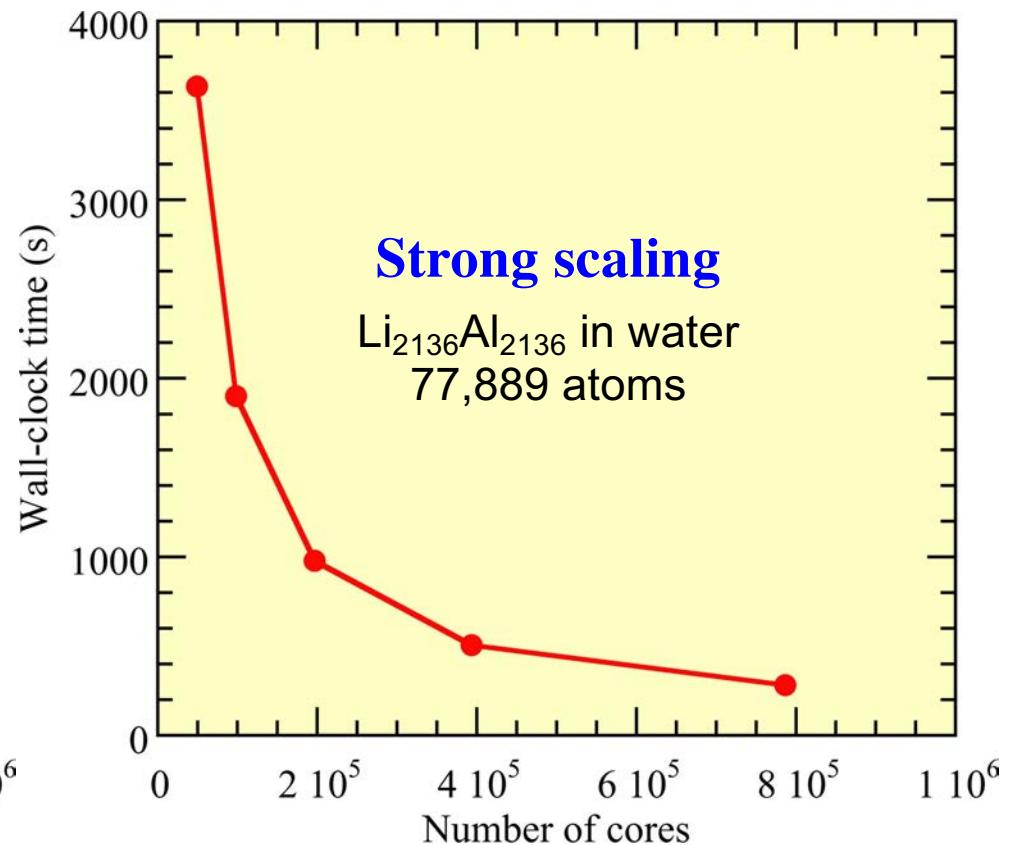
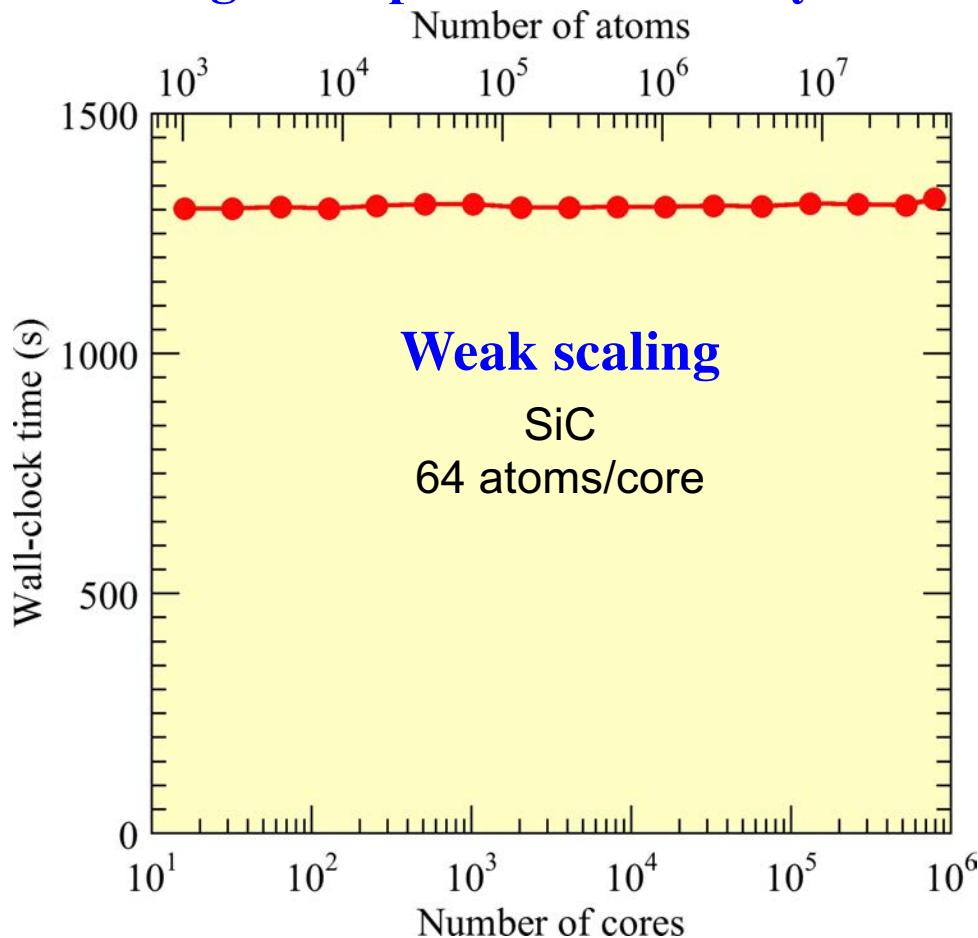
cf. globally- sparse-yet-locally-dense eigensolver [J. H. Lam et al., *Nature Commun.* **15**, 3479 ('24)]

- 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]

# 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)

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



$$\Psi = [|\psi_1^\alpha\rangle, \dots, |\psi_{N_{\text{band}}}^\alpha\rangle] \quad \tilde{\mathbf{B}}(i) = [|\beta_{i,1}\rangle, \dots, |\beta_{i,N_{\text{atom}}}\rangle] \quad [\tilde{\mathbf{D}}(i,j)]_{I,J} = D_{ij,I} \delta_{IJ}$$

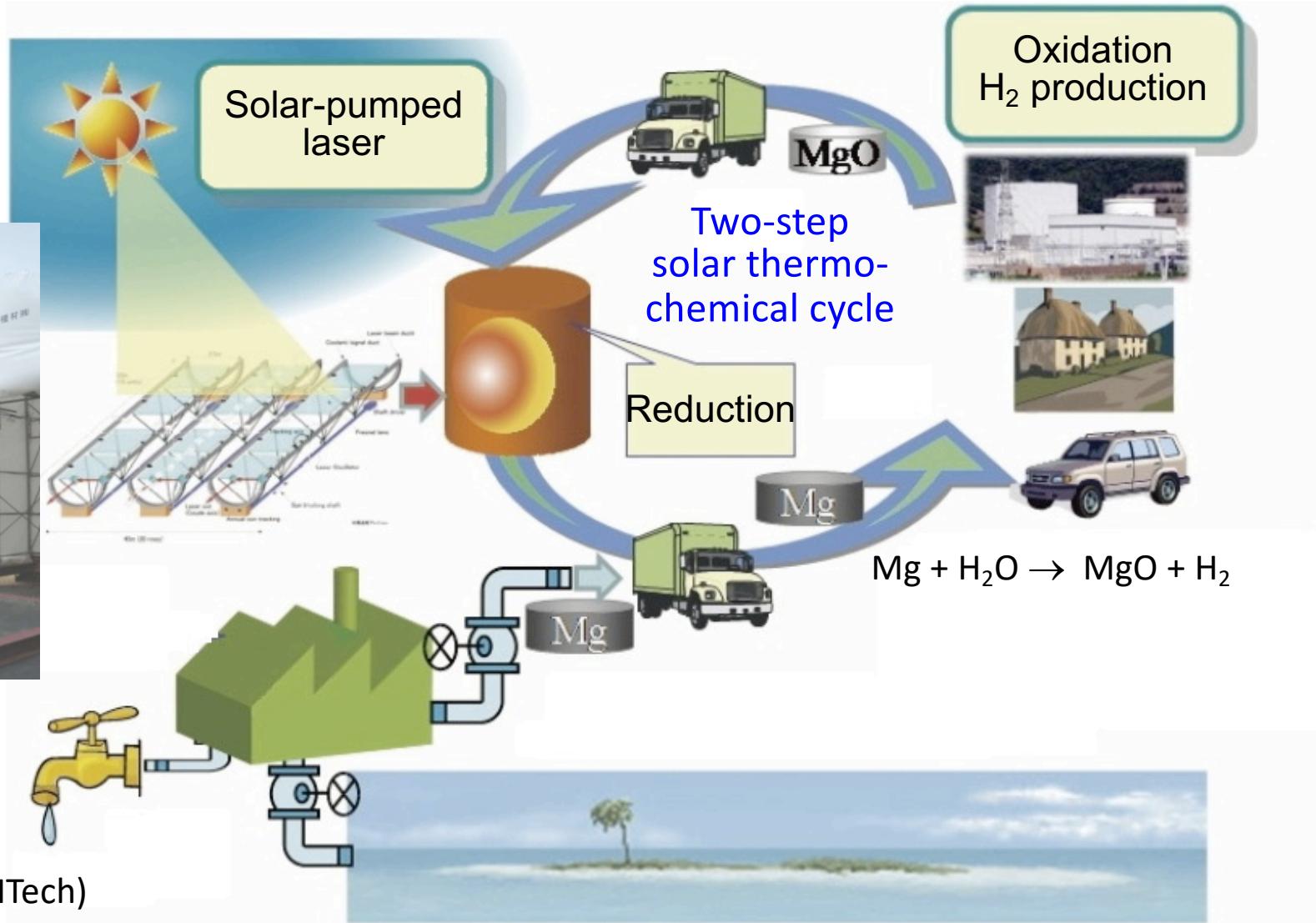
$$\hat{v}_{\text{nl}}\Psi = \sum_{i,j}^L \tilde{\mathbf{B}}(i) \tilde{\mathbf{D}}(i,j) \tilde{\mathbf{B}}(j)^T$$

- **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)

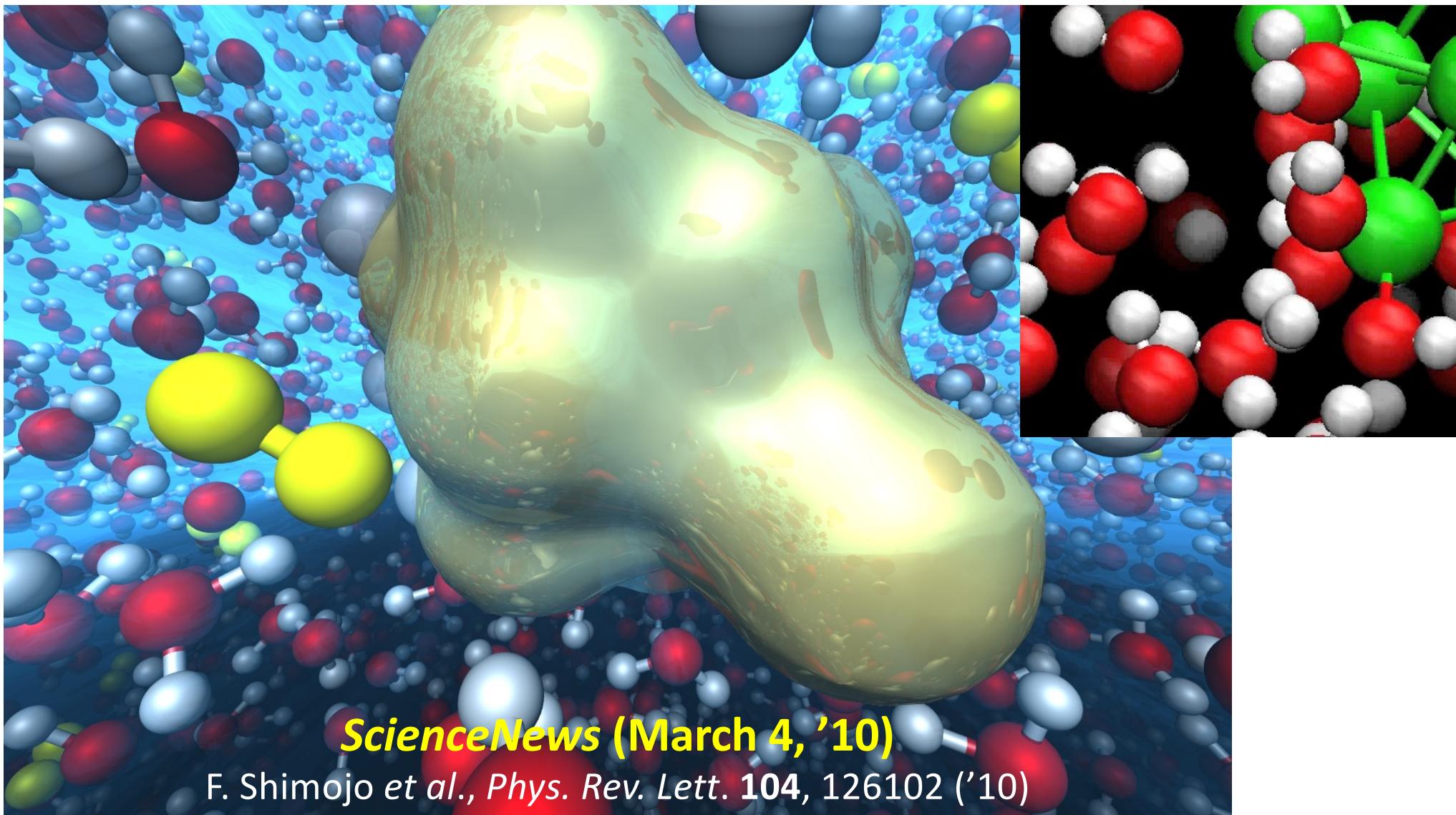
S. C. Tiwari *et al.*, *HPC Asia* ('20) best paper

# Renewal Energy Cycle by Metal Carriers



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

# Nanotechnology Solution



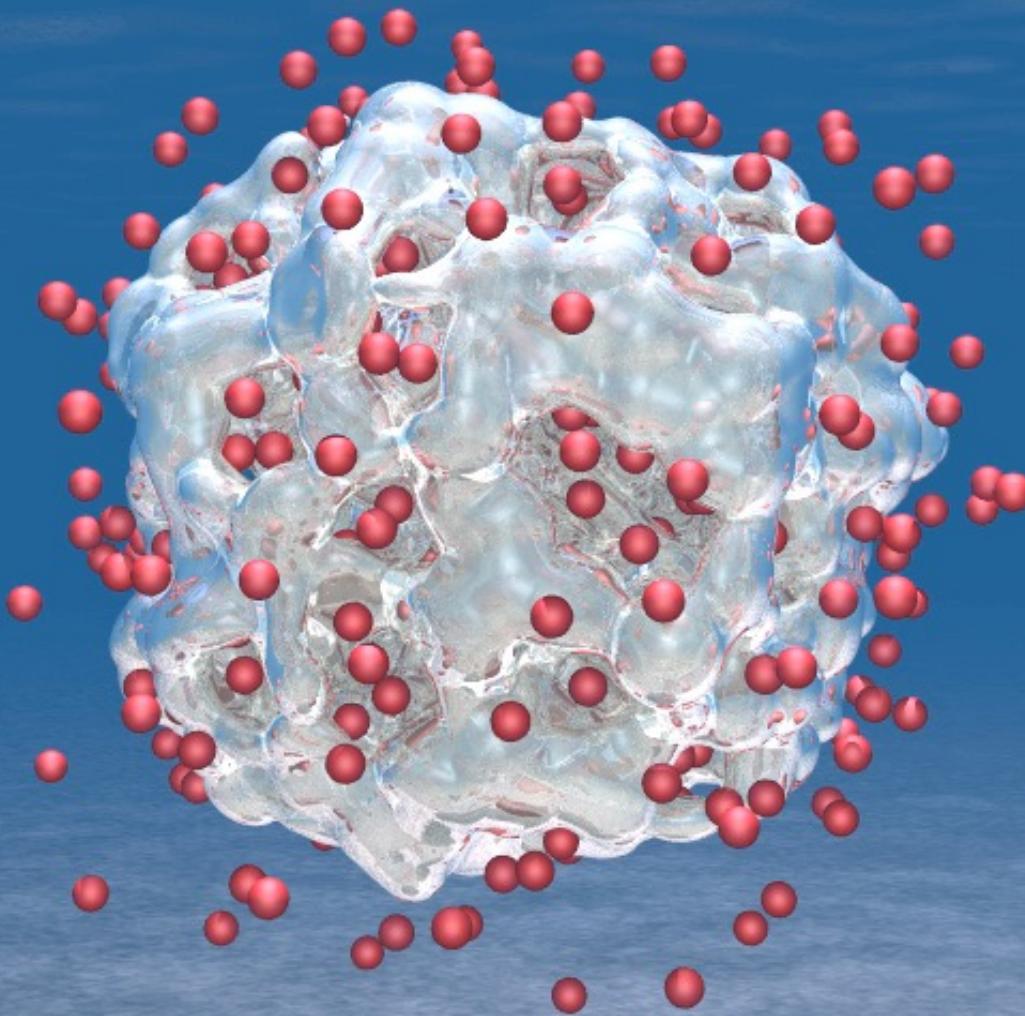
- QMD simulation shows rapid H<sub>2</sub> production from water by a superatom\* (Al<sub>17</sub>), but the technology is not scalable to larger particle sizes

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

# H<sub>2</sub> Production from Water Using LiAl Particles

16,661-atom QMD simulation of Li<sub>441</sub>Al<sub>441</sub> in water  
on 786,432 IBM Blue Gene/Q cores

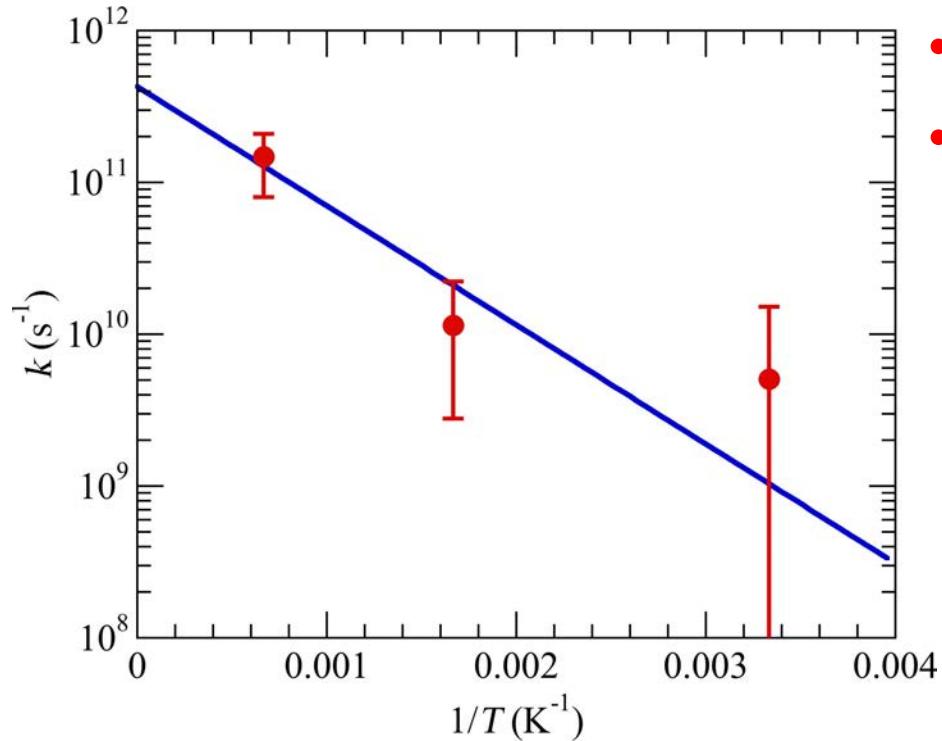
K. Shimamura *et al.*,  
*Nano Lett.* **14**, 4090 ('14)



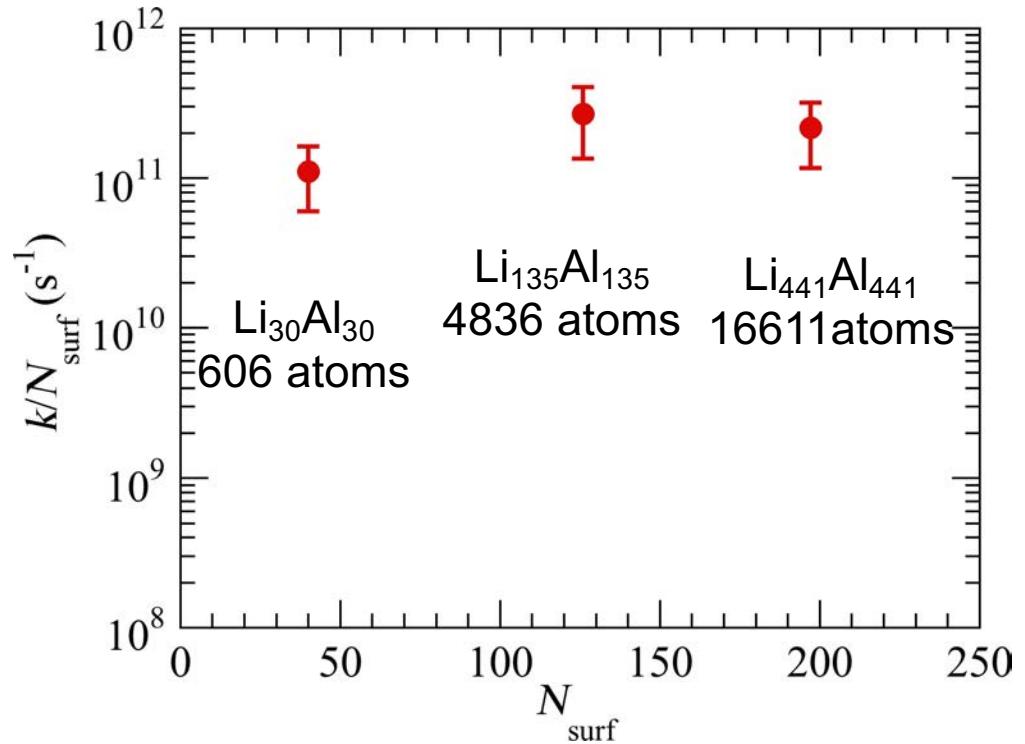
21,140 time steps (129,208 self-consistent-field iterations)

# Rapid & Scalable H<sub>2</sub> Production

- Orders-of-magnitude faster H<sub>2</sub> production from water than with pure Al



- Activation barrier = 0.068 eV
- Reaction rate =  $1.04 \times 10^9$  (s<sup>-1</sup>) per LiAl pair at 300 K

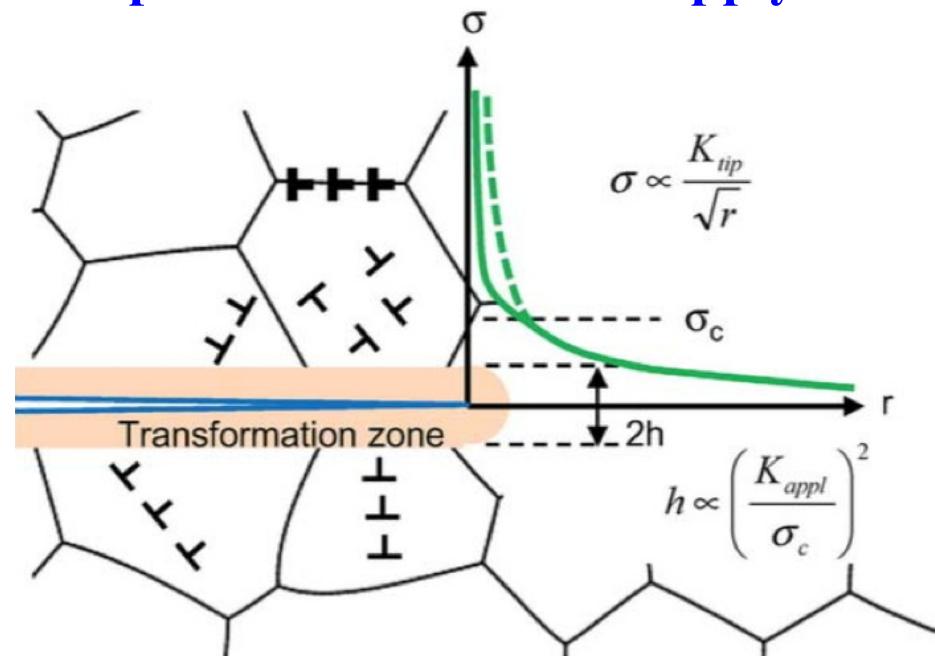
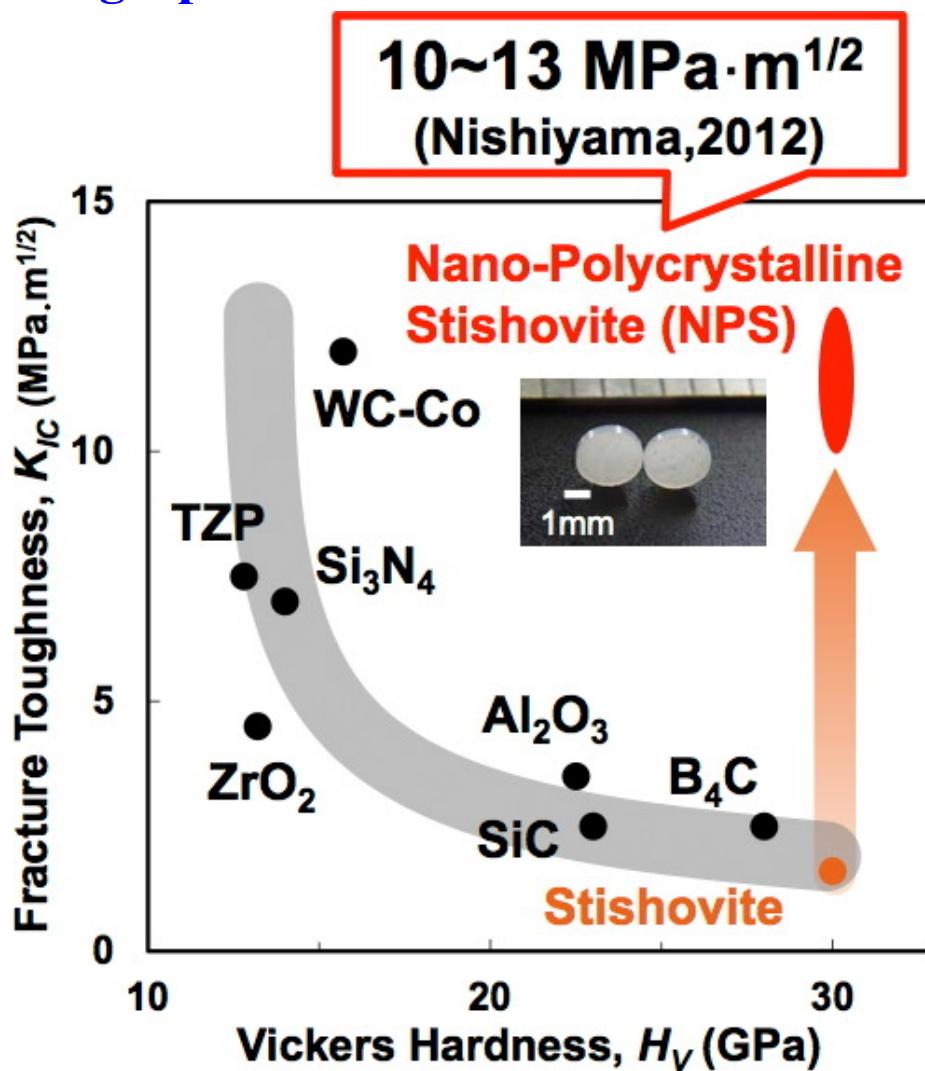


- Reaction rate does not decrease for larger particles → industrial scalability

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

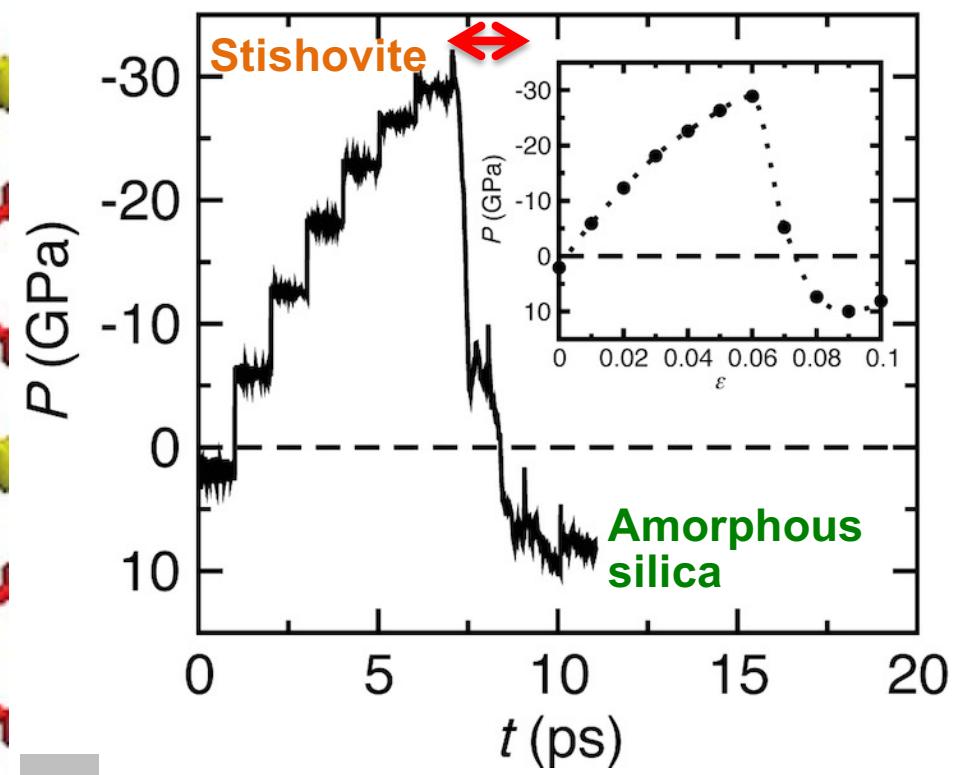
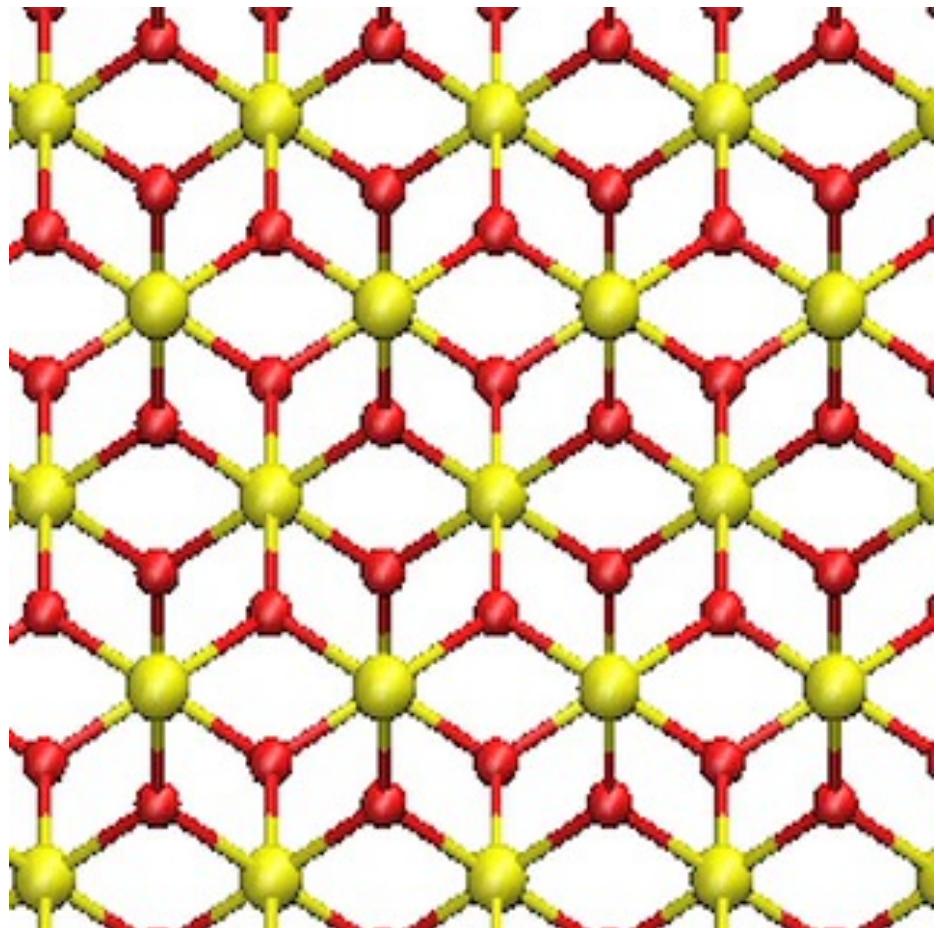


K. Yoshida *et al.*, *Sci. Rep.* **5**, 10993 ('15);  
*Acta Mater.* **124**, 316 ('17)

- 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  $\sim 30$  GPa

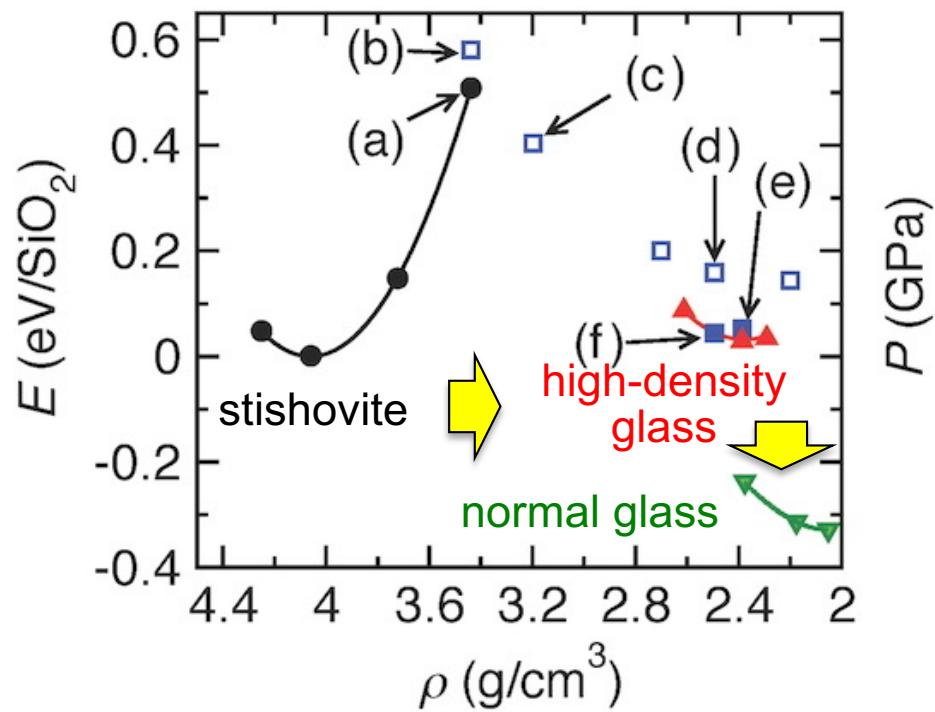
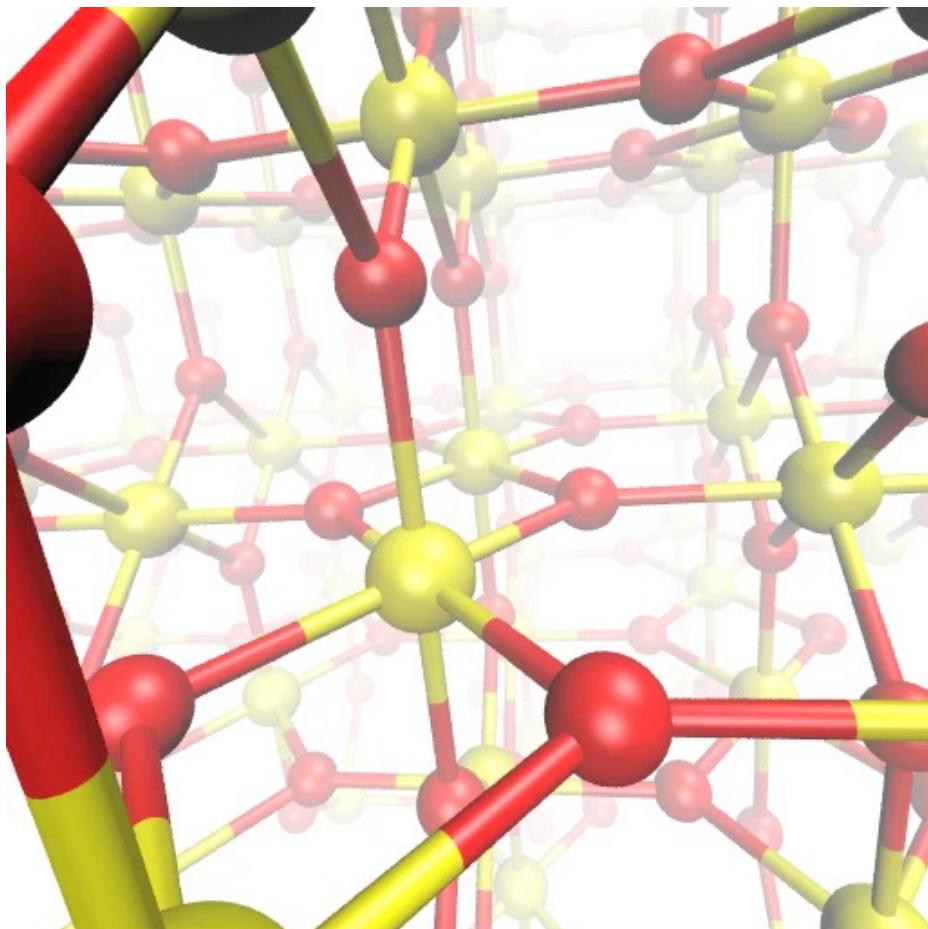


*Volume<sub>amorphous silica</sub>*  $\sim 2 \times$  *Volume<sub>stishovite</sub>*

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

# Rapid Amorphization Mechanism

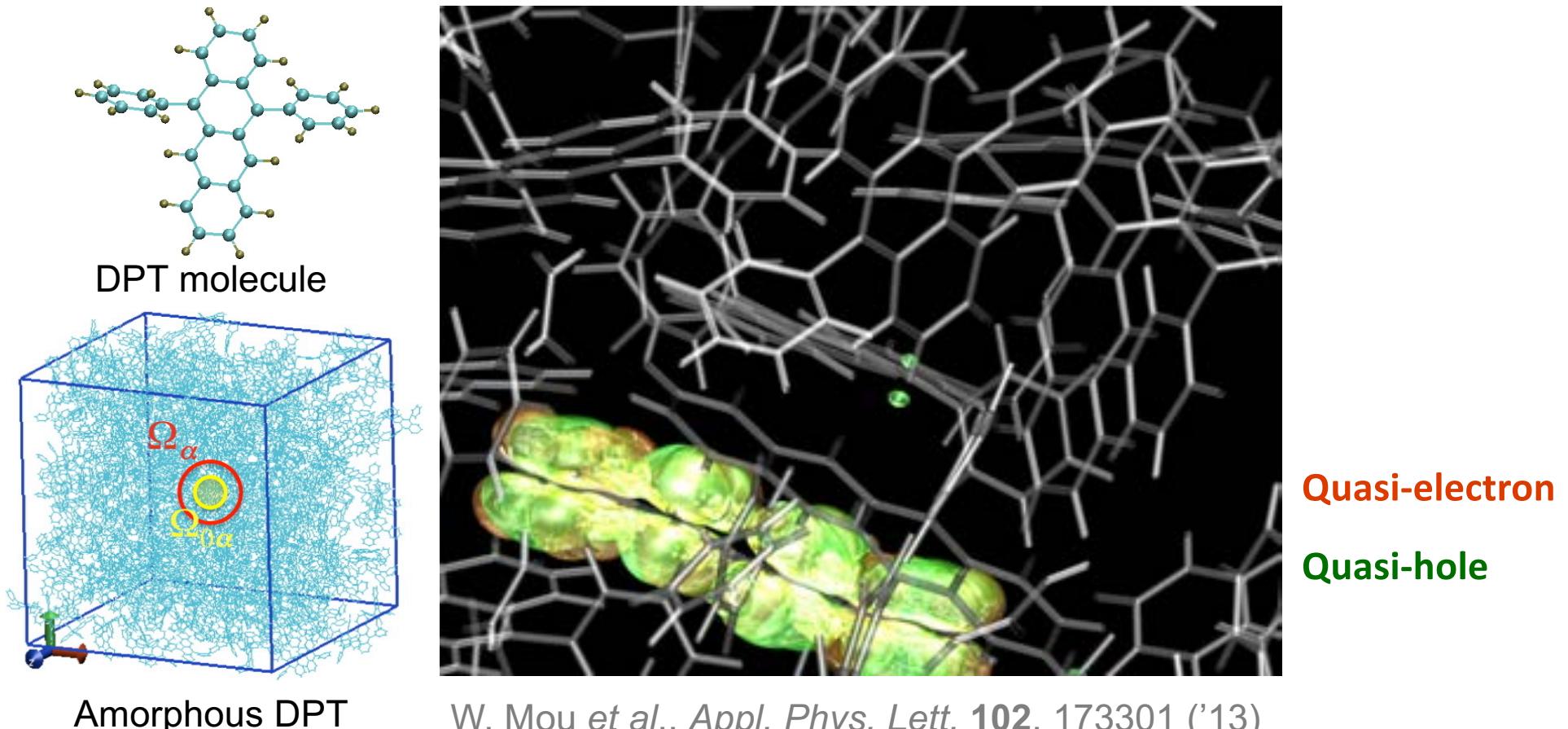
- Found a displacive amorphization mechanism that only involves short-distance collective motions of atoms, thereby facilitating the rapid transformation



- 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

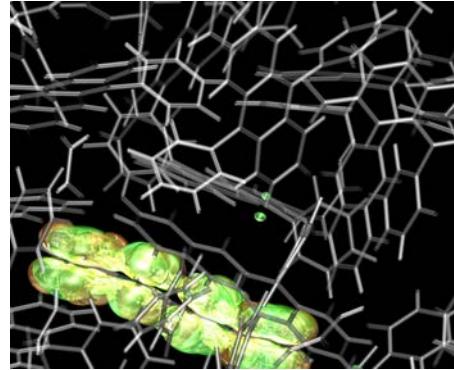
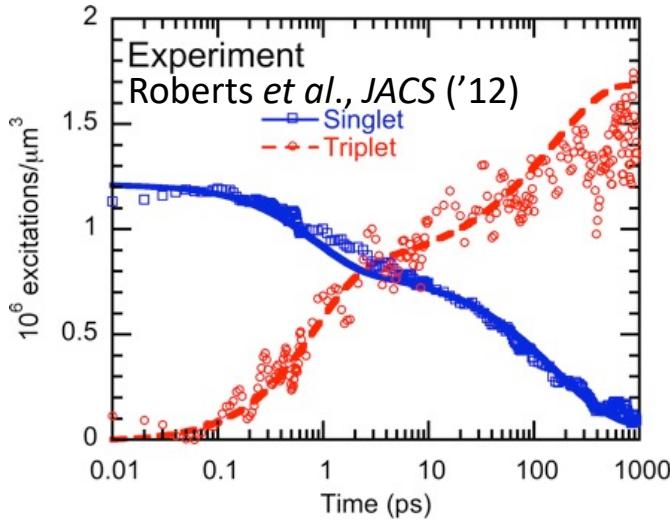
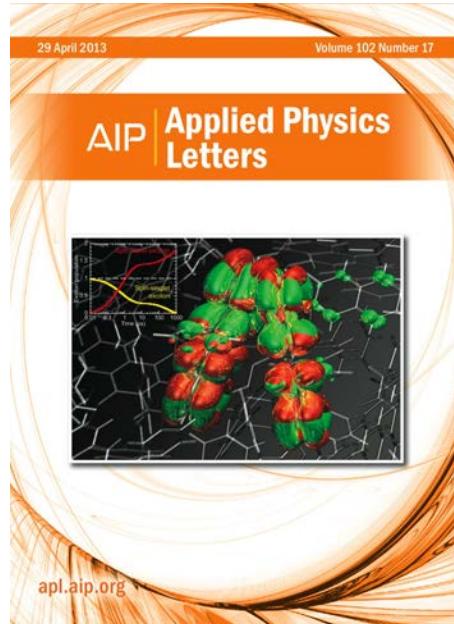
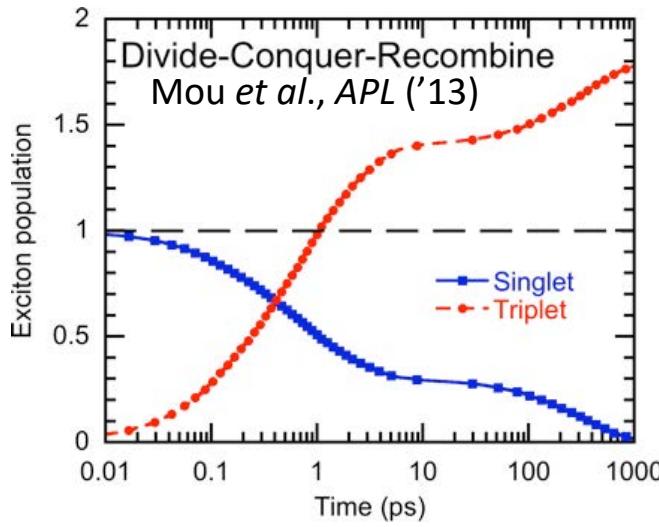
- Photo-current doubling by splitting a singlet exciton into 2 triplet excitons
- Singlet fission in mass-produced disordered organic solid → efficient low-cost solar cells
- Experimental breakthrough: SF found in amorphous diphenyl tetracene (DPT)



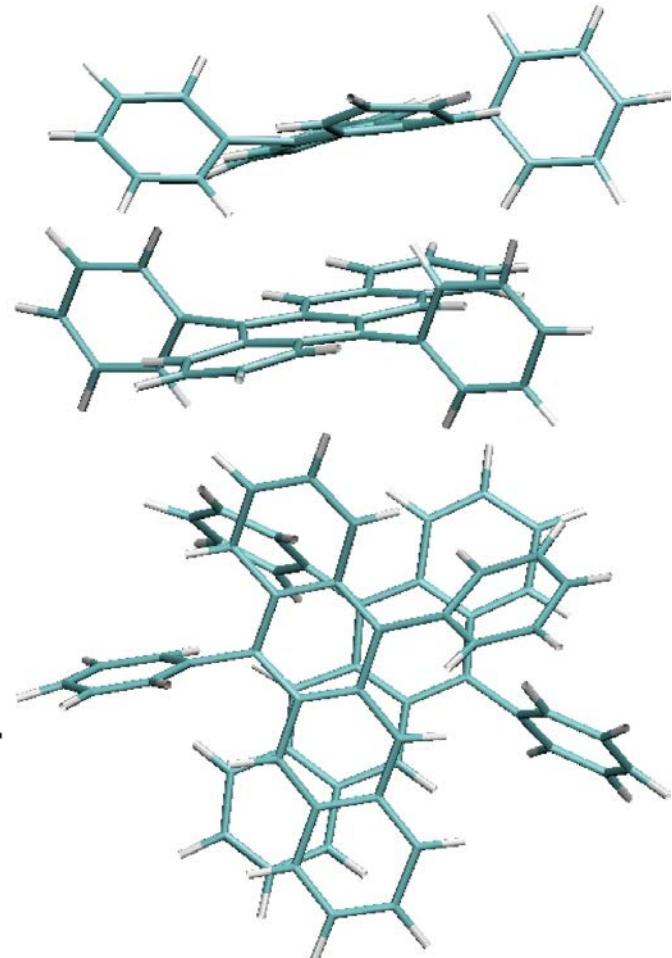
- Divide-conquer-recombine nonadiabatic QMD (phonon-assisted exciton dynamics) + time-dependent perturbation theory (singlet-fission rate) + kinetic Monte Carlo calculations of exciton population dynamics in 6,400-atom amorphous DPT

# 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



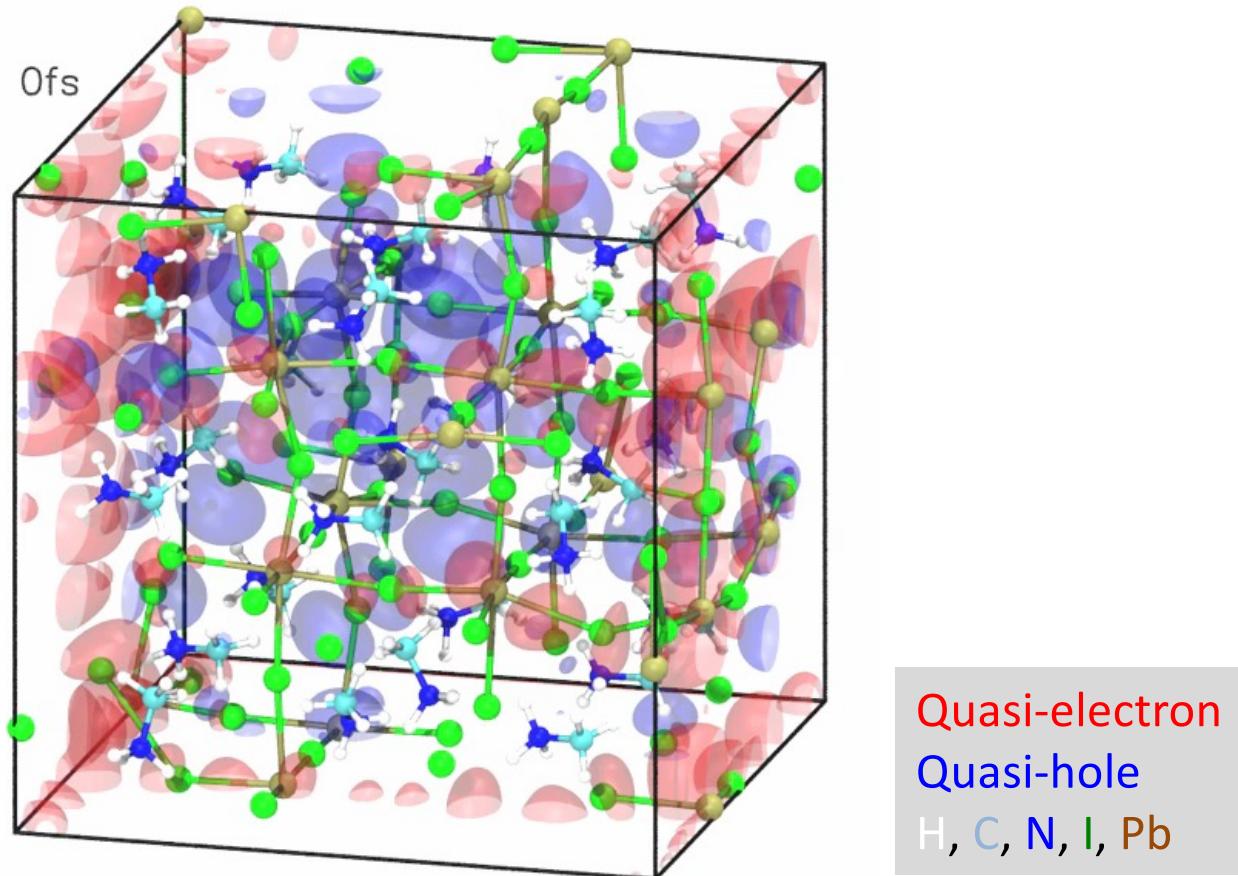
Side view  
Top view



# Photoexcited Carriers in $\text{MAPbI}_3$

- Organometal halide perovskites (e.g. methylammonium lead iodide,  $\text{CH}_3\text{NH}_3\text{PbI}_3$  or  $\text{MAPbI}_3$ ) for solar cells with high power conversion efficiency > 20%

[Stranks & Snaith, *Nat. Nanotechnol.* **10**, 391 ('15)]

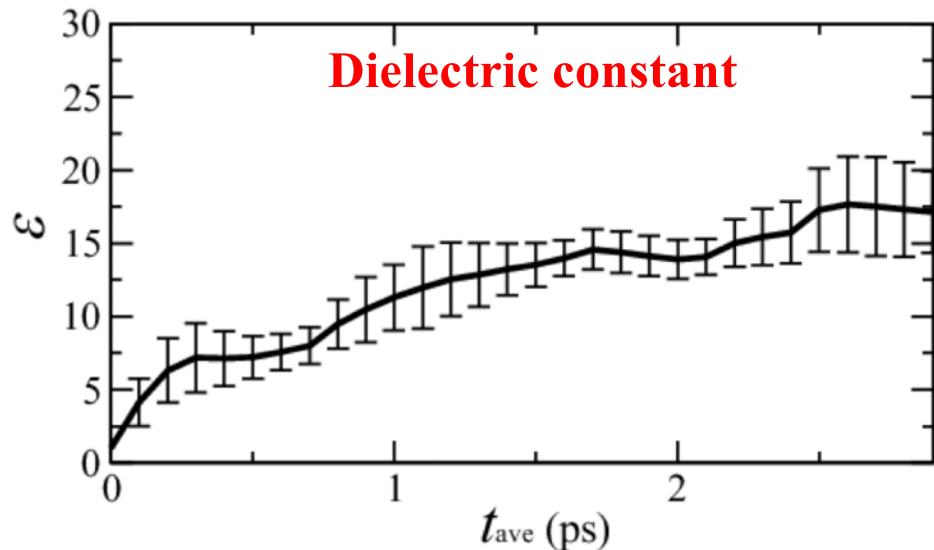


- Nonadiabatic QMD simulation

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  $D_e = (1.16 \pm 0.31) \times 10^{-2} \text{ cm}^2/\text{s}$  &  $D_h = (1.01 \pm 0.42) \times 10^{-2} \text{ cm}^2/\text{s}$

Expt:  $D_e = (1.7 \pm 1.1) \times 10^{-2} \text{ cm}^2/\text{s}$  &  $D_h = (1.1 \pm 0.7) \times 10^{-2} \text{ cm}^2/\text{s}$  [Stranks *et al.*, *Science* **342**, 341 ('13)]

# Screening Role of Methylammonium Sublattice

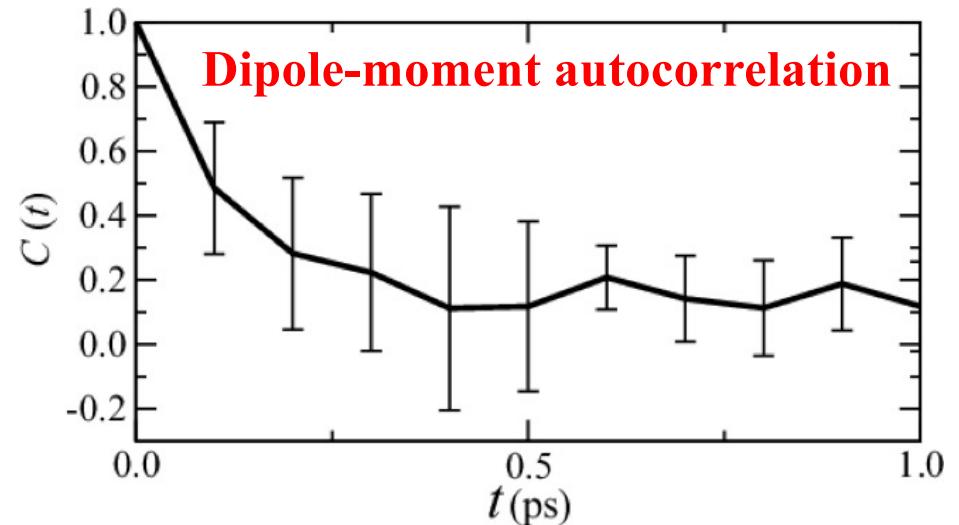


$$\epsilon = 1 + \frac{4\pi}{3k_B TV} (\langle \mathbf{M}^2 \rangle - \langle \mathbf{M} \rangle^2)$$

time average  
dipole moment

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

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  $\sim 1$  ps**

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

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

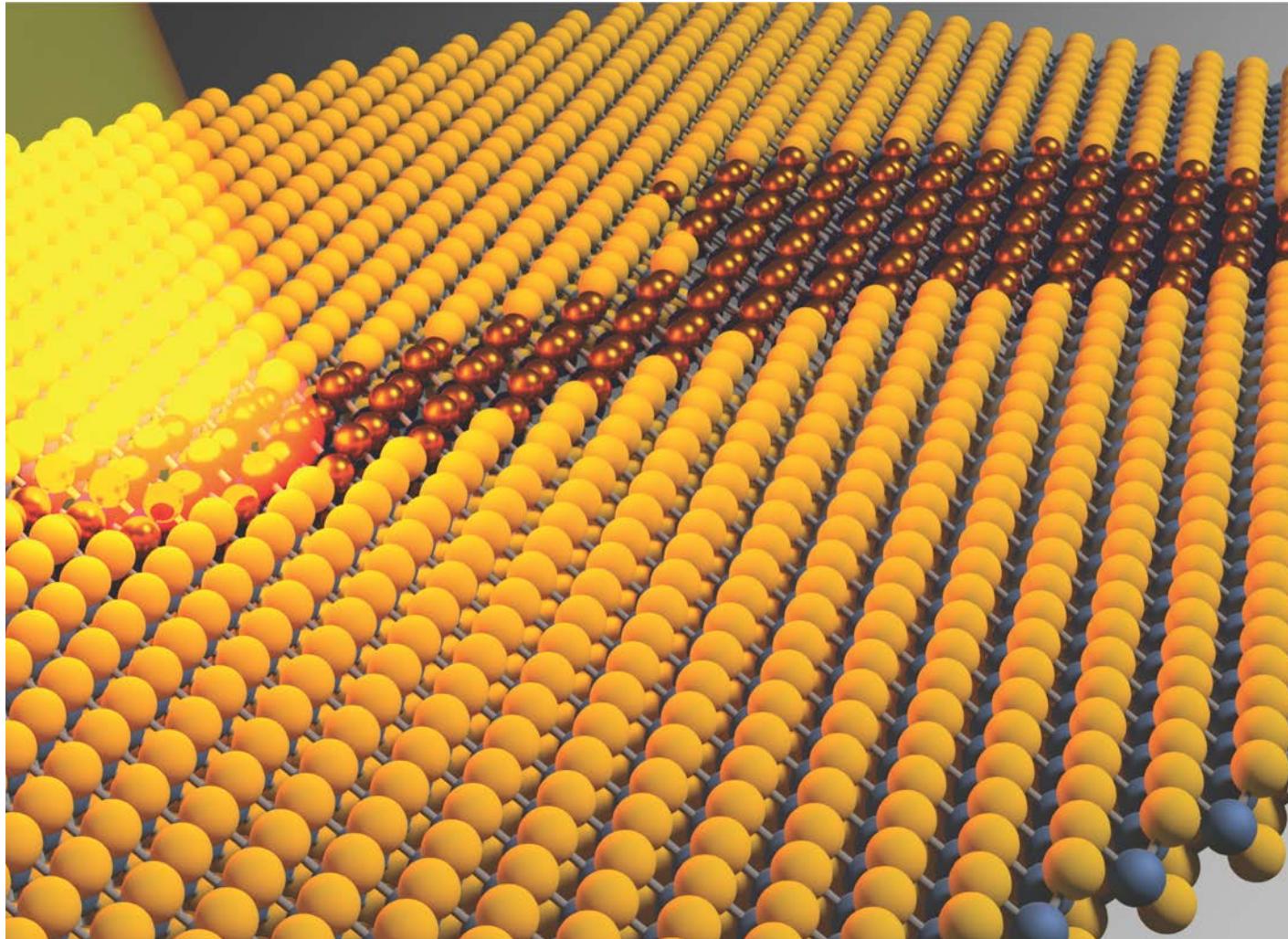
- Large dielectric constant of MA sublattice causes small exciton binding energy, **0.012 ± 0.009 eV** (experimental upper bound = **0.05 eV** [D'Innocenzo *et al.*, *Nat. Commun.* **5**, 3586 ('14)])
- MA sublattice quickly screens out electrostatic electron-hole attraction to unbind an exciton & generate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

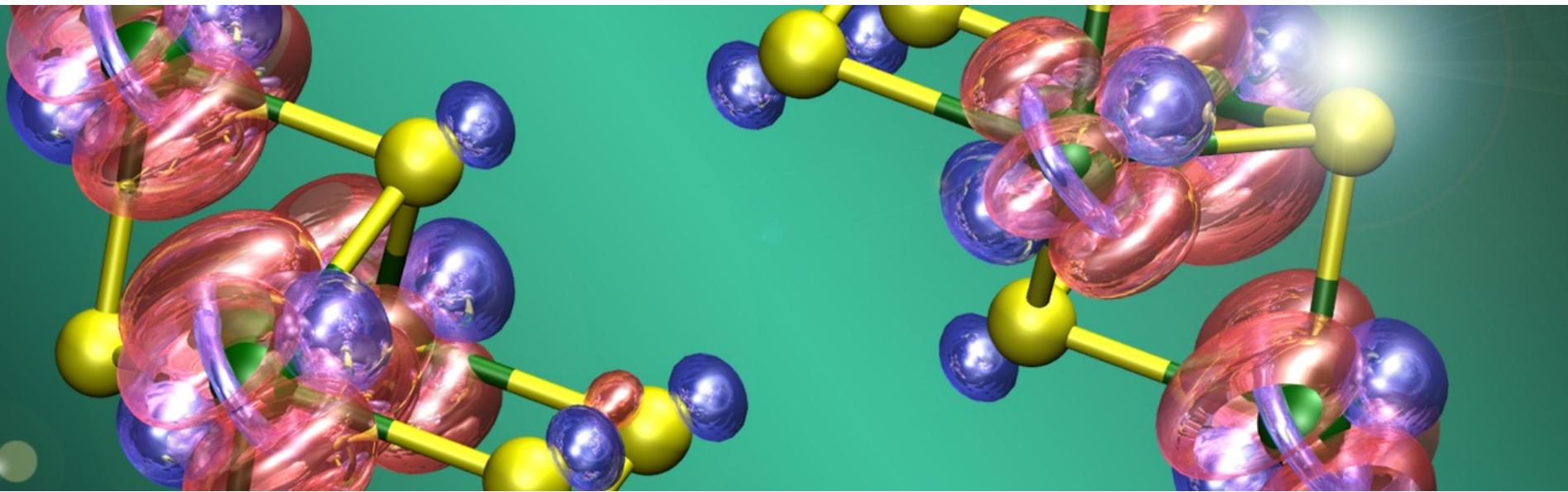
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**Goal:** Use ultrafast laser pulses to transform material structures & properties  
(e.g., semiconductor-to-metal) on demand

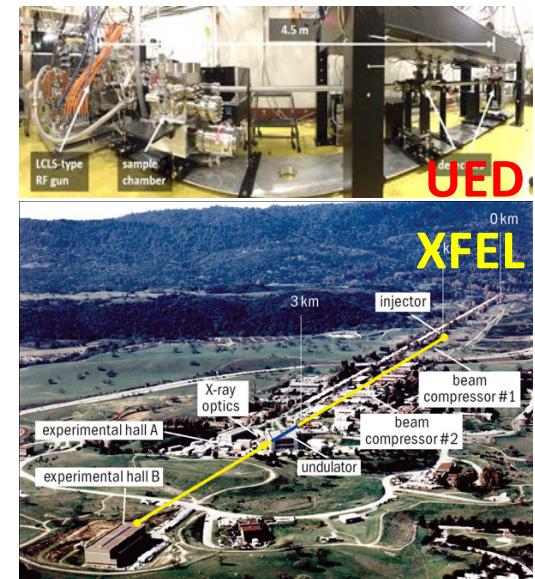


A. Krishnamoorthy *et al.*, *Nanoscale* **10**, 2742 ('18); journal cover

# Simulation-Experiment Synergy



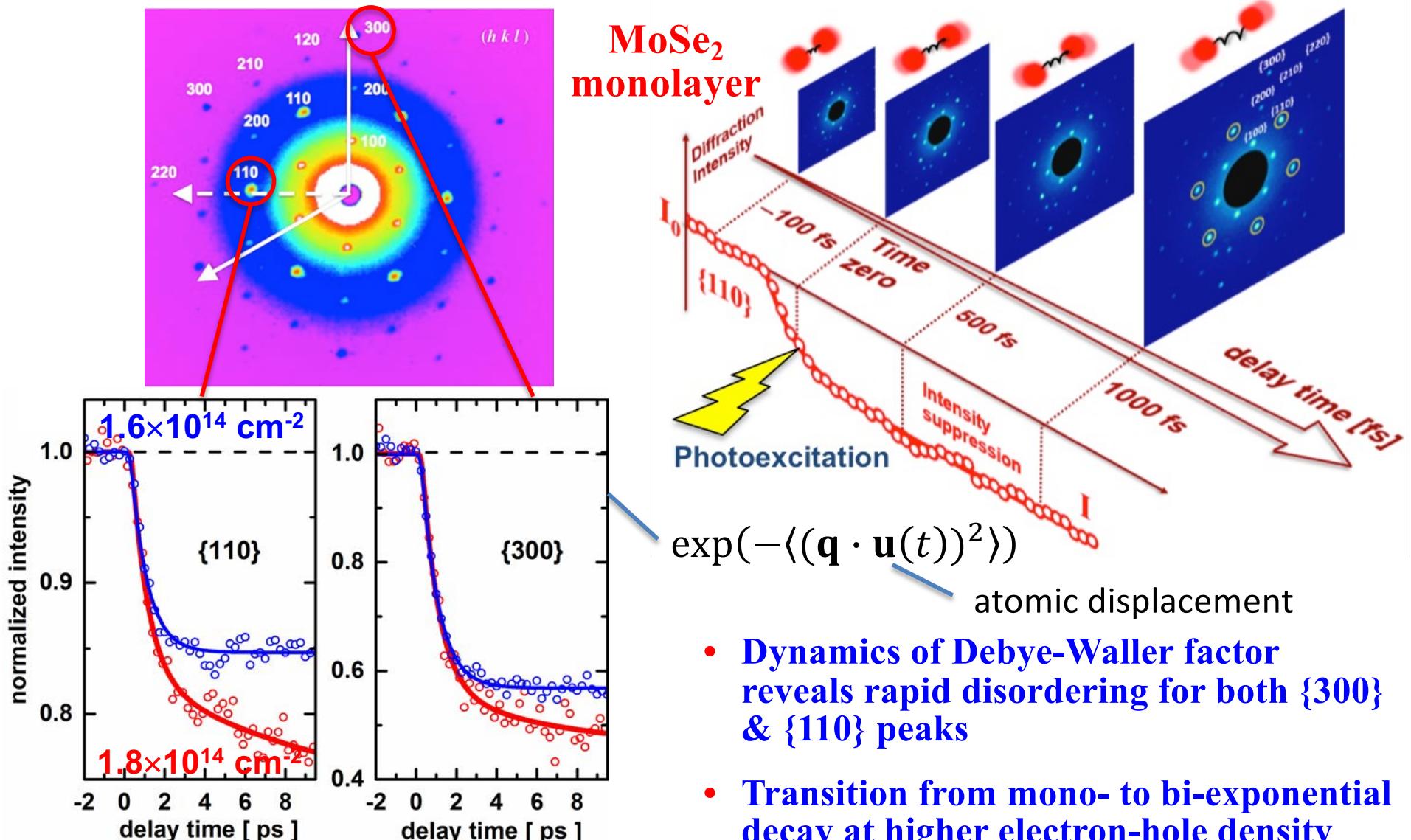
- In ultrafast ‘electron & X-ray cameras,’ laser light hitting a material is almost completely converted into nuclear motions — key to switching material properties on & off at will for future electronics applications.
- High-end nonadiabatic quantum molecular dynamics simulations reproduce the ultrafast 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

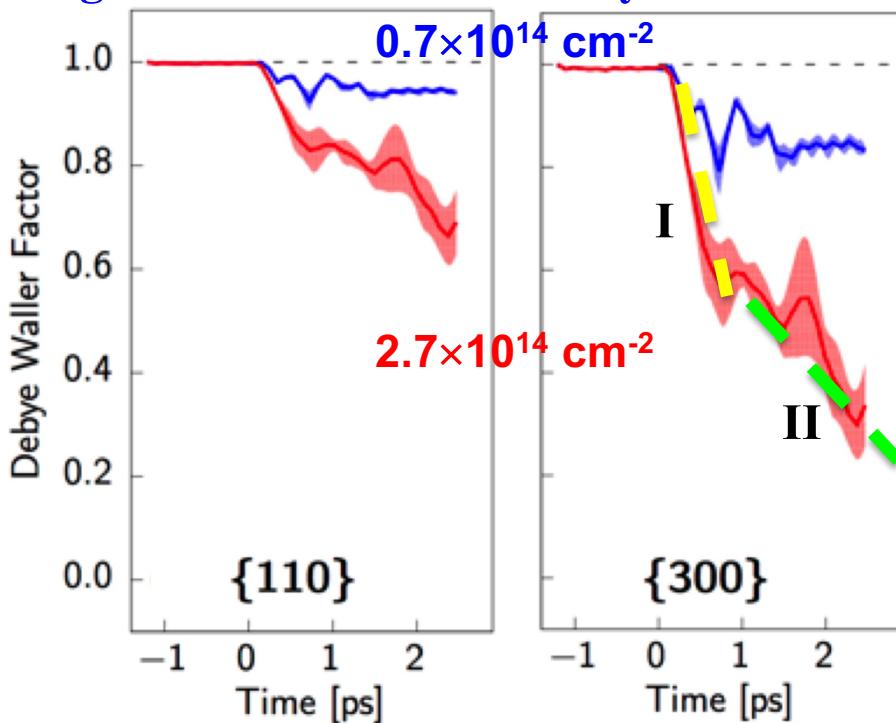
- Ultrafast electron diffraction experiment shows nearly perfect energy conversion from electronic excitation to lattice motions within ps



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

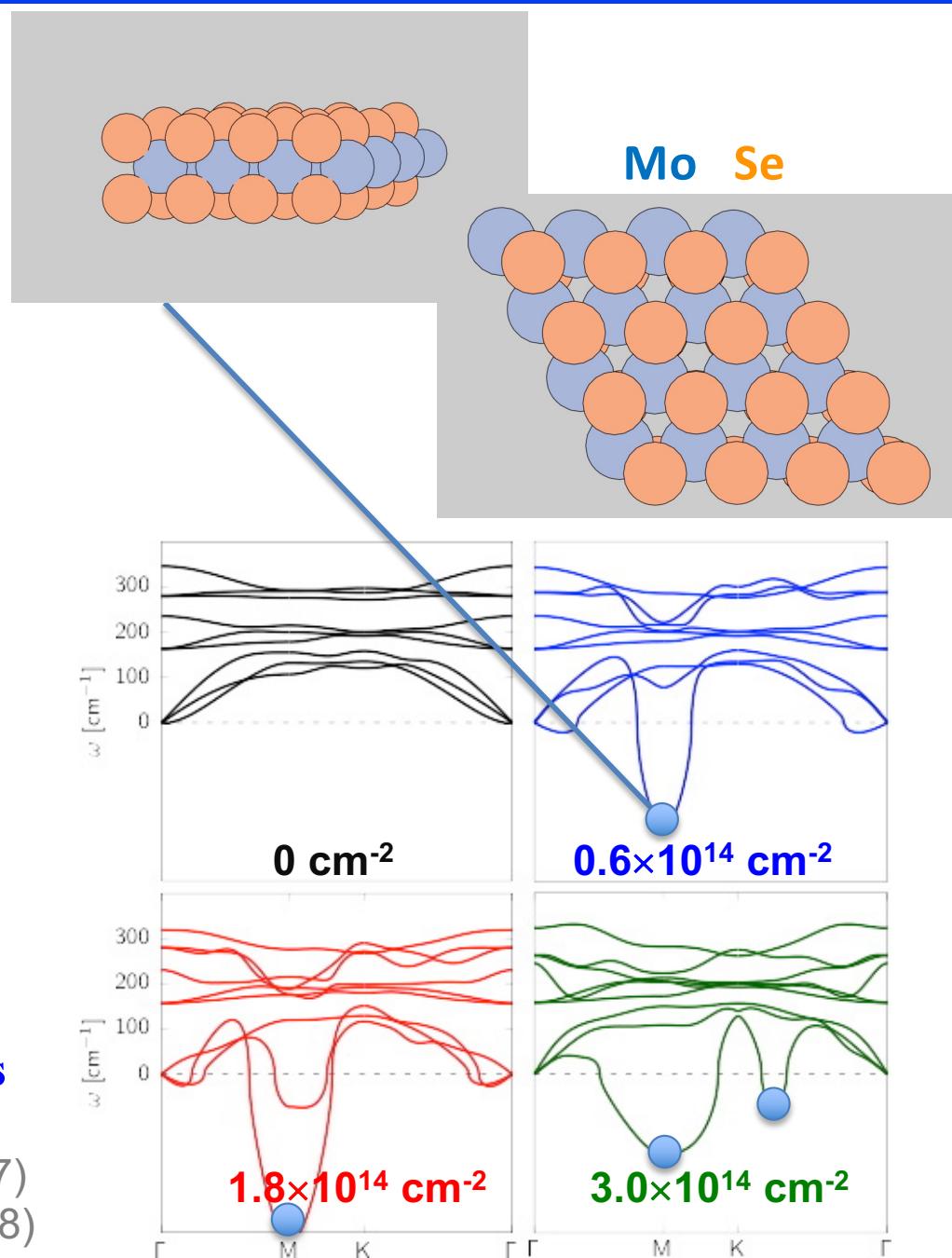
# Strong Electron-Lattice Coupling in MoSe<sub>2</sub>

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



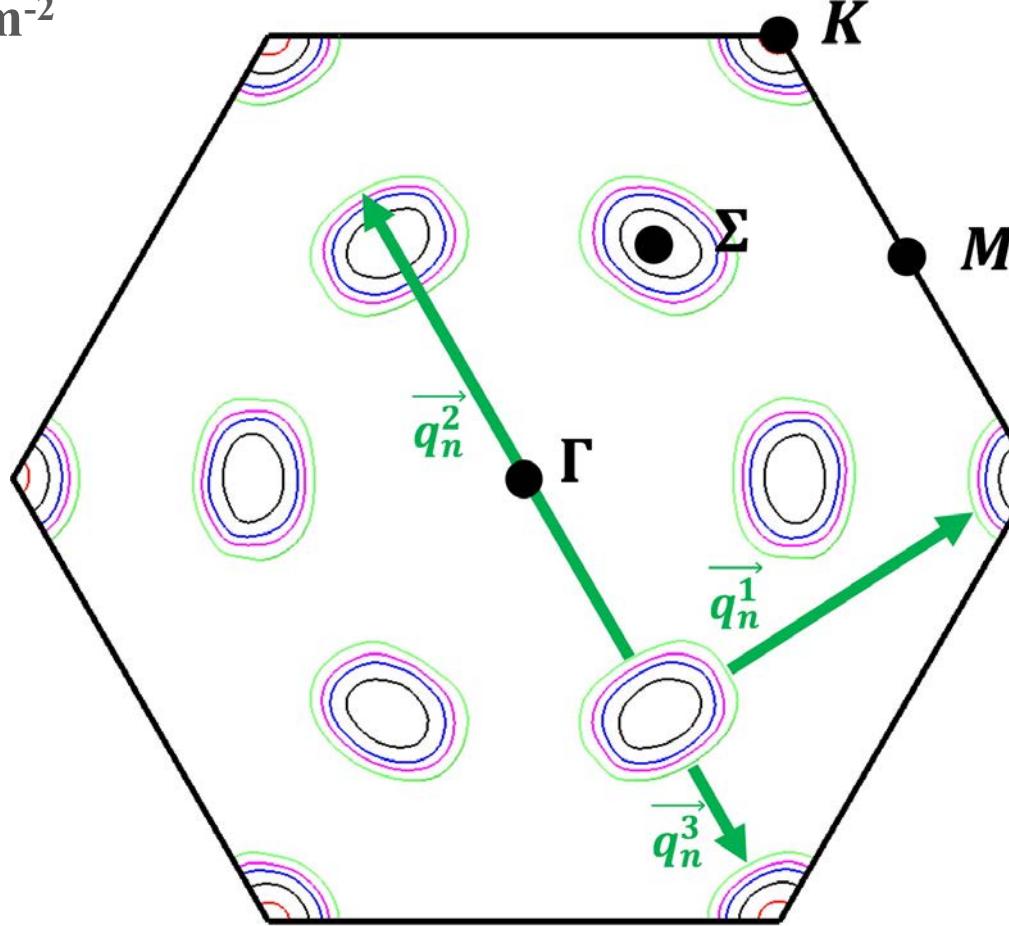
- Rapid lattice dynamics is explained by the softening of M-point ( $\frac{1}{2} 0 0$ ) phonon
- Bi-exponential transition is explained by the softening of additional phonon modes at higher electron-hole densities

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  $n(\text{e-h})$  ranging from 0.2 to  $2 \times 10^{14} \text{ cm}^{-2}$

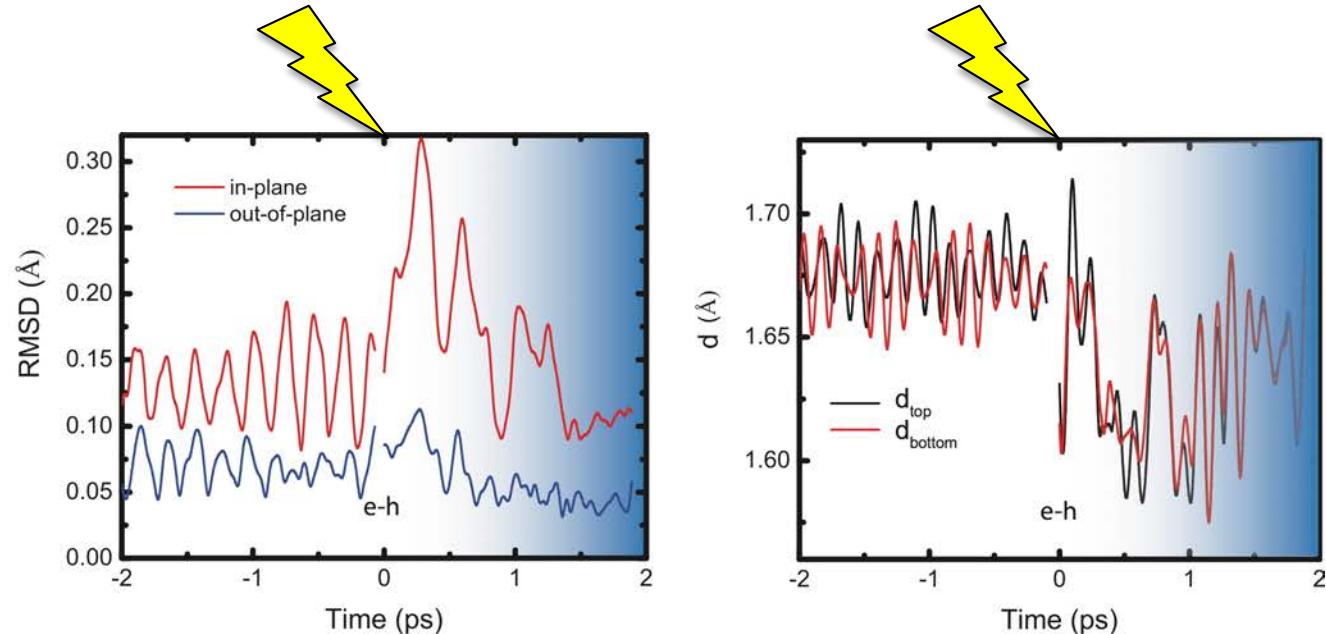
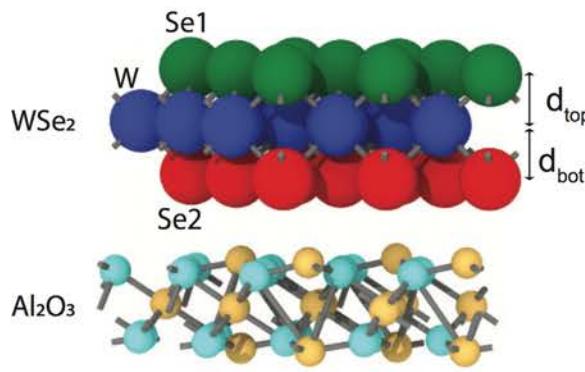


$$n(\text{e-h}) = 0.22, 1, 2, 3, 4 \times 10^{14} \text{ cm}^{-2}$$

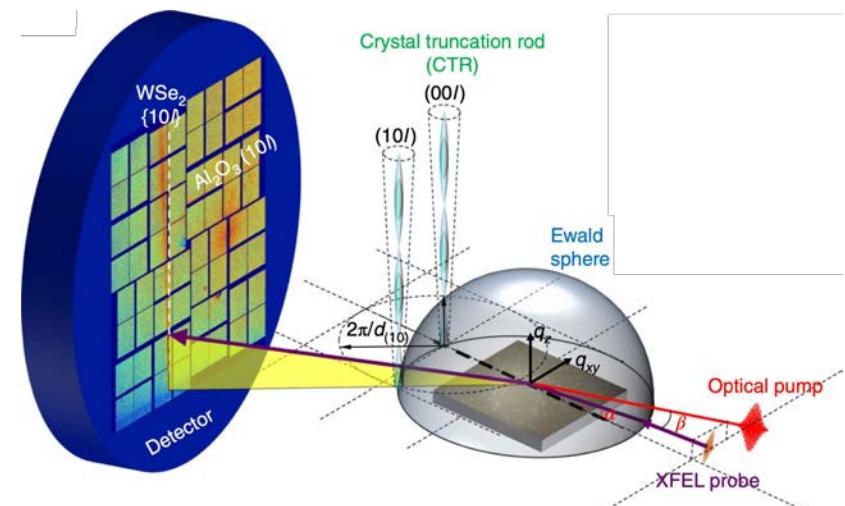
- While the Fermi surface is localized at K-points at minimal excitation (red), it occupies  $\Sigma$ -pockets at larger  $n(\text{e-h})$  (black & blue), enabling electron scattering by emitting  $\vec{q}_n^1$  ( $M$ ),  $\vec{q}_n^2$  ( $\Sigma$ ) and  $\vec{q}_n^3$  ( $K$ ) phonons

# WSe<sub>2</sub> Monolayer on Al<sub>2</sub>O<sub>3</sub> Substrate

- NAQMD simulation to study photoexcitation dynamics of WSe<sub>2</sub> monolayer on Al<sub>2</sub>O<sub>3</sub> substrate



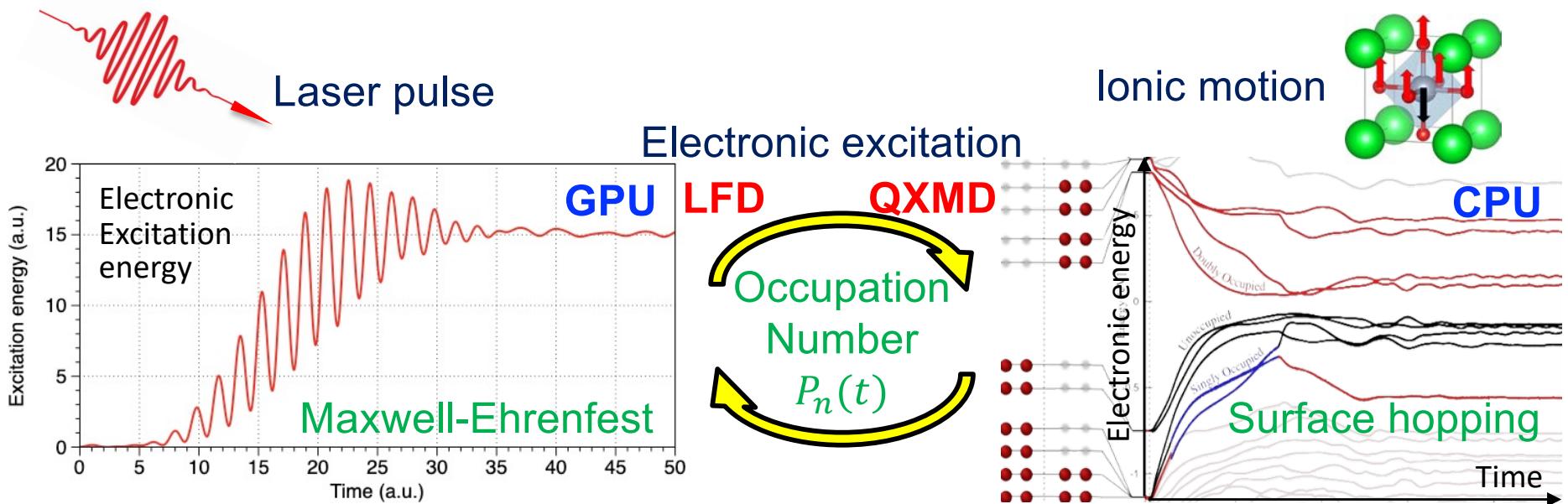
- Enhanced in-plane atomic displacements upon photoexcitation
- Photo-induced intralayer contraction of W-Se distances
- Good agreement with femtosecond surface X-ray scattering experiments at Stanford SLAC/LCLS



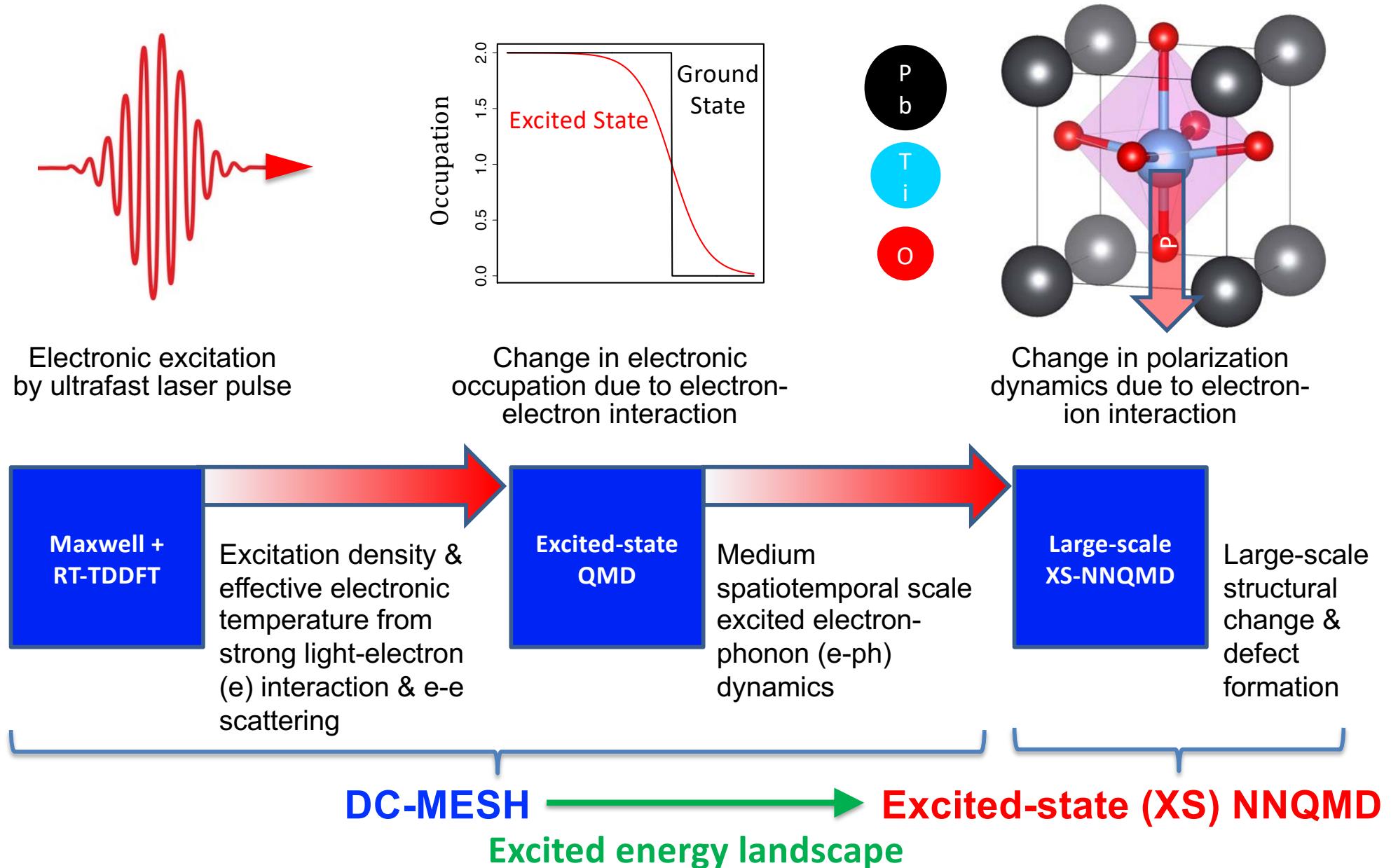
Tung, et al., *Nature Photonics* 13, 425 ('19)

# 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



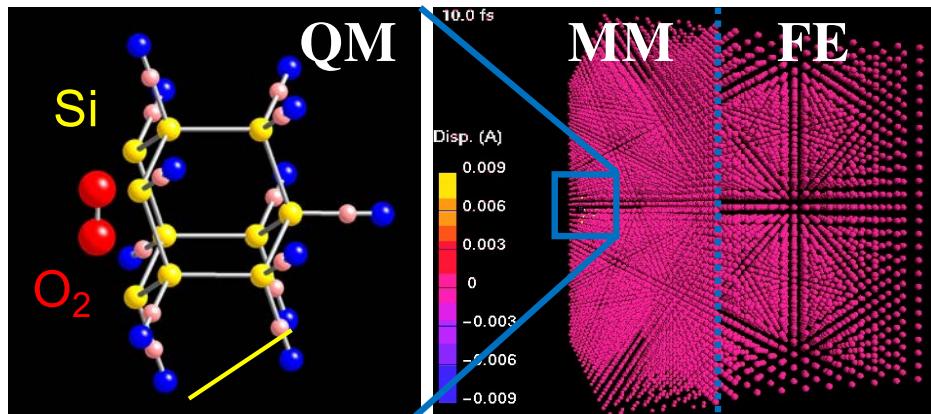
# Multiscaling from DC-MESH to XS-NNQMD



# 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)

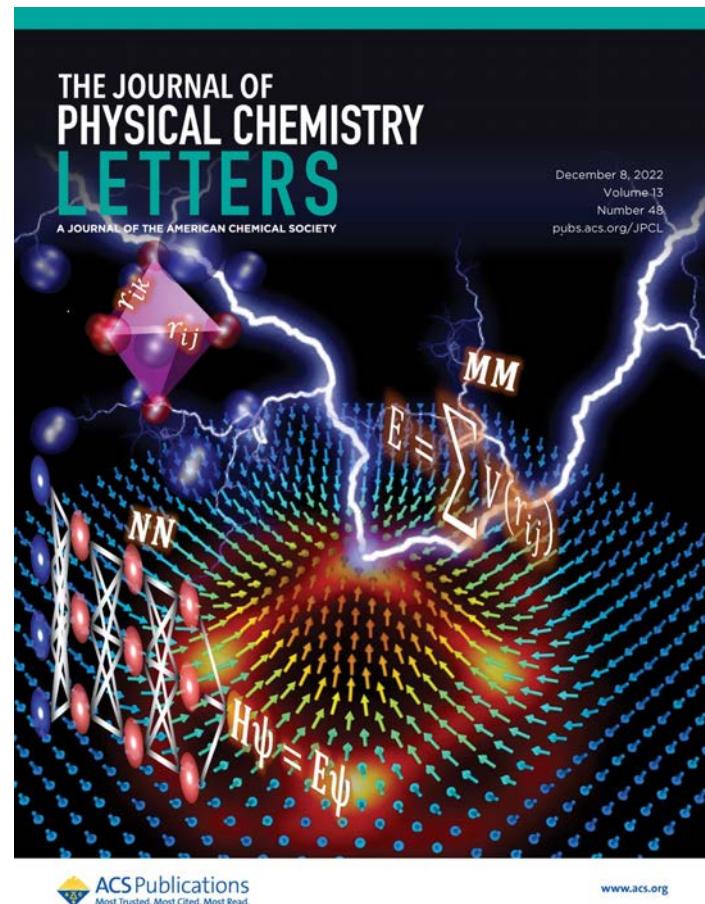
Warshel, *Angew. Chem.* **53**, 10020 ('14)



**QM/MM/FE (finite-element method)**

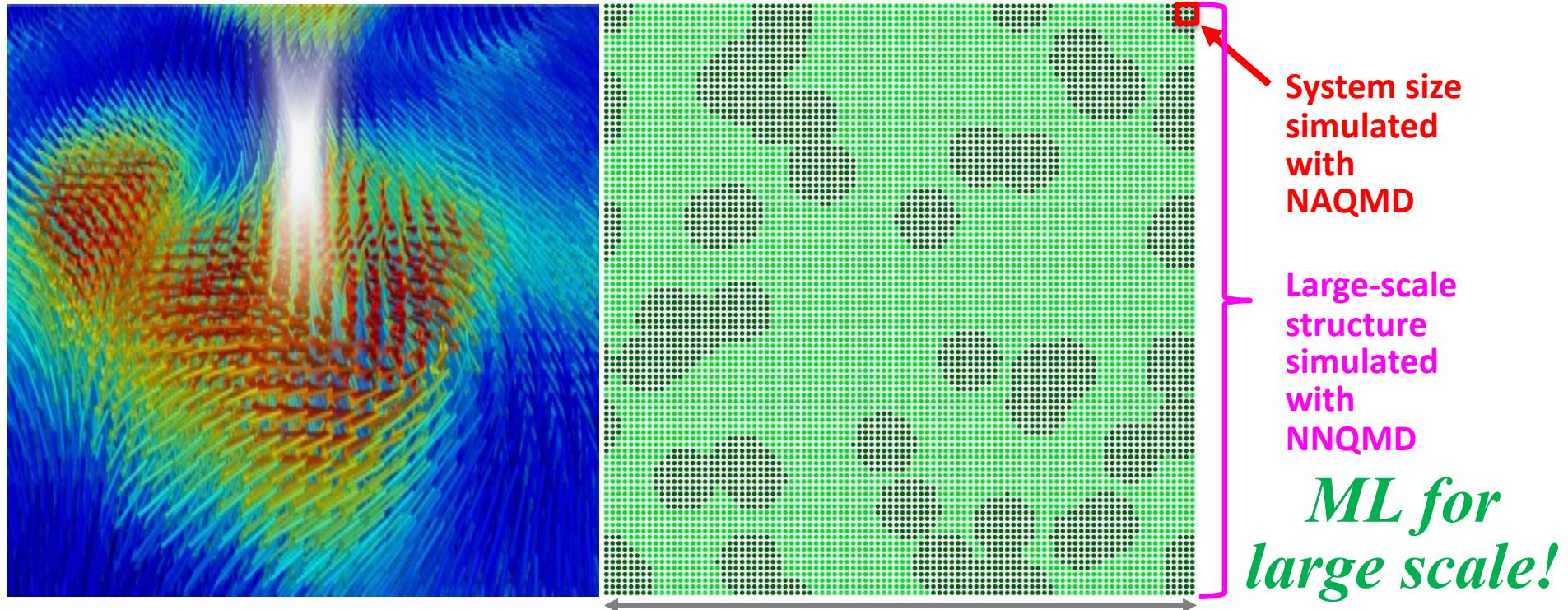
Ogata et al, *Comput. Phys. Commun.* **138**, 143 ('01)

- **NN/MM:** NNQMD for ferroelectric (PbTiO<sub>3</sub>: PTO) embedded in MM for paraelectric (SrTiO<sub>3</sub>: STO) to apply appropriate strain boundary condition



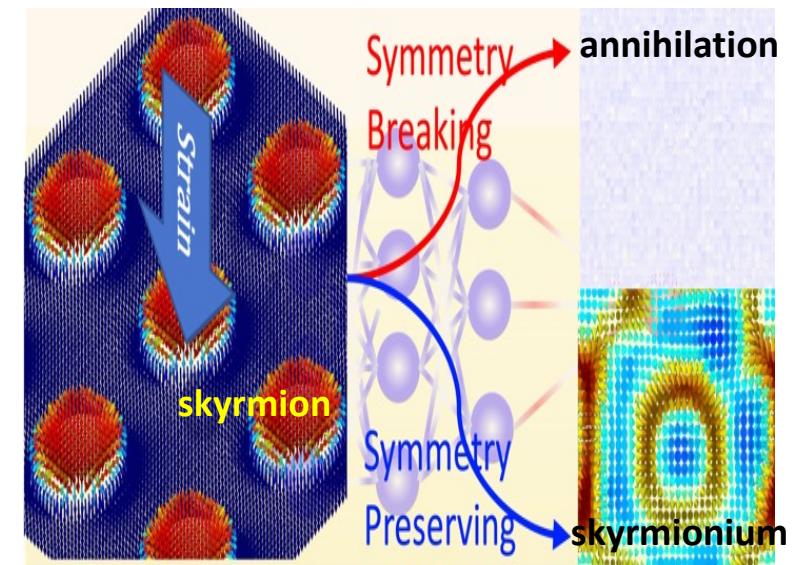
Linker et al., *J. Phys. Chem. Lett.* **13**, 11335 ('22); *Nano Lett.* **23**, 7456 ('23)

# Application: Ferroelectric Opto-Toptronics



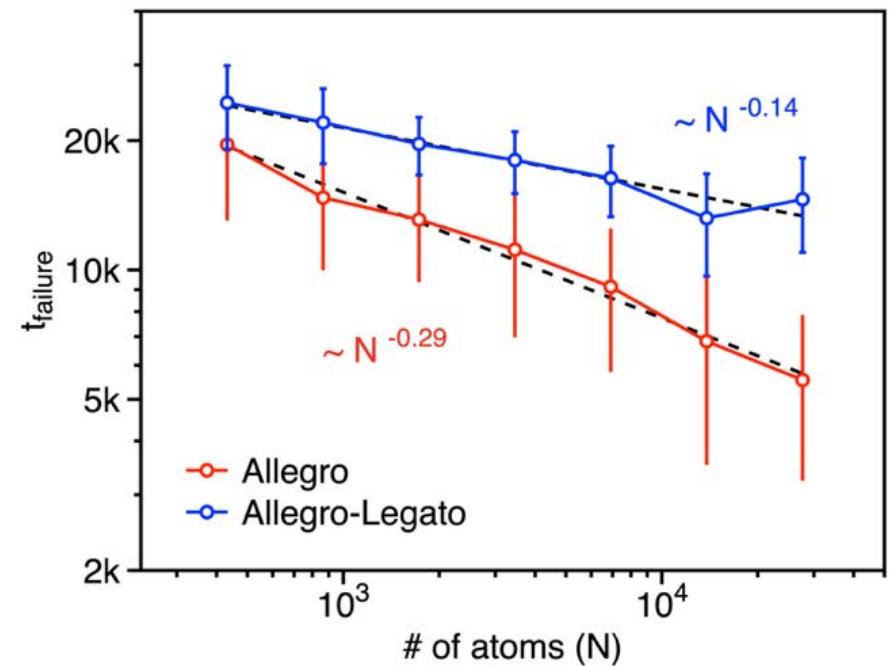
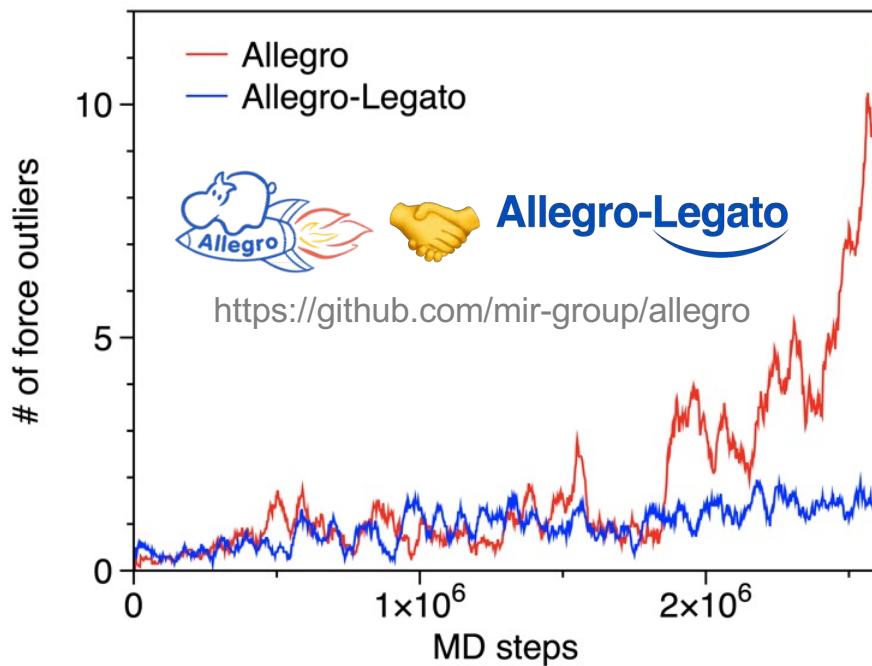
- Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications
- Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)
- Symmetry-controlled skyrmion-to-skyrmionium<sup>\*</sup> 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)



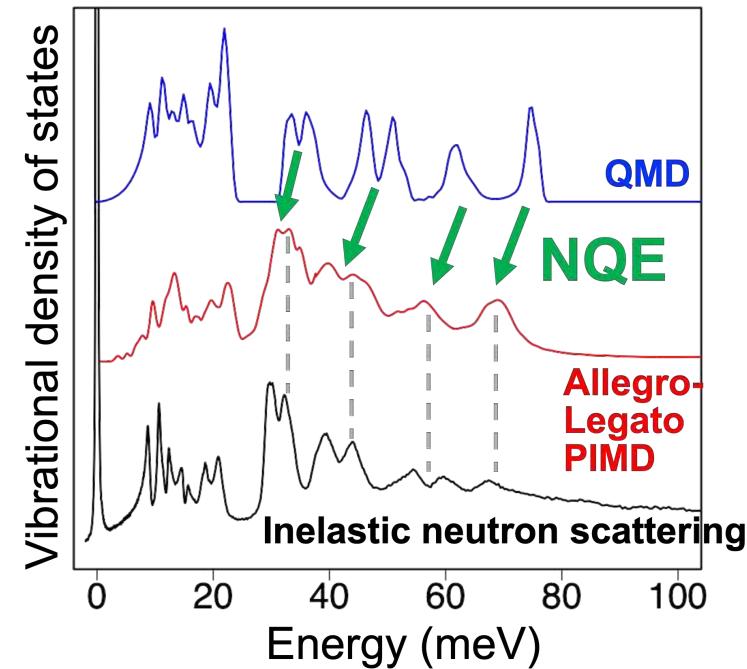
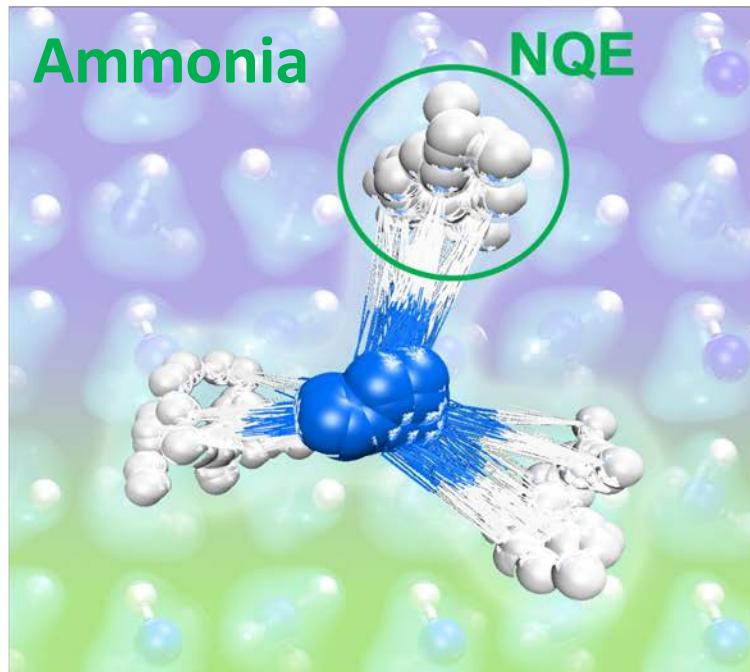
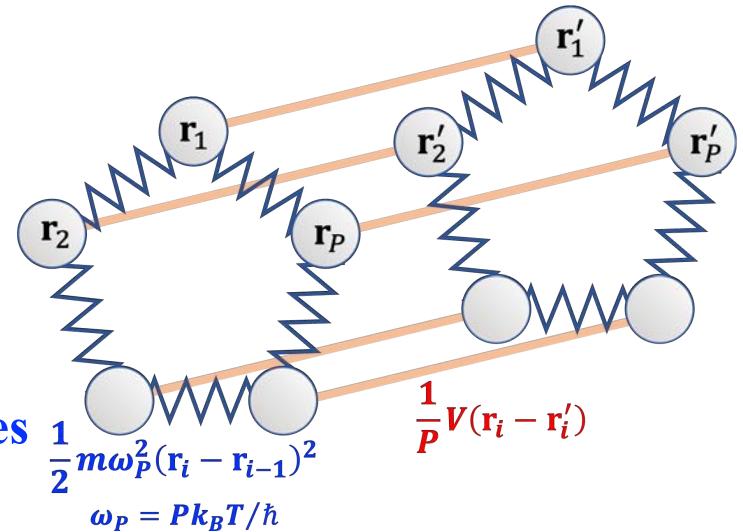
# Fast & Robust NNQMD: Allegro-Legato

- **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  
 $w_* = \operatorname{argmin}_w [L(w) + \max_{\|\epsilon\|_2 \leq \rho} \{L(w + \epsilon) - L(w)\}]$  ( $L$ : loss;  $w$ : 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



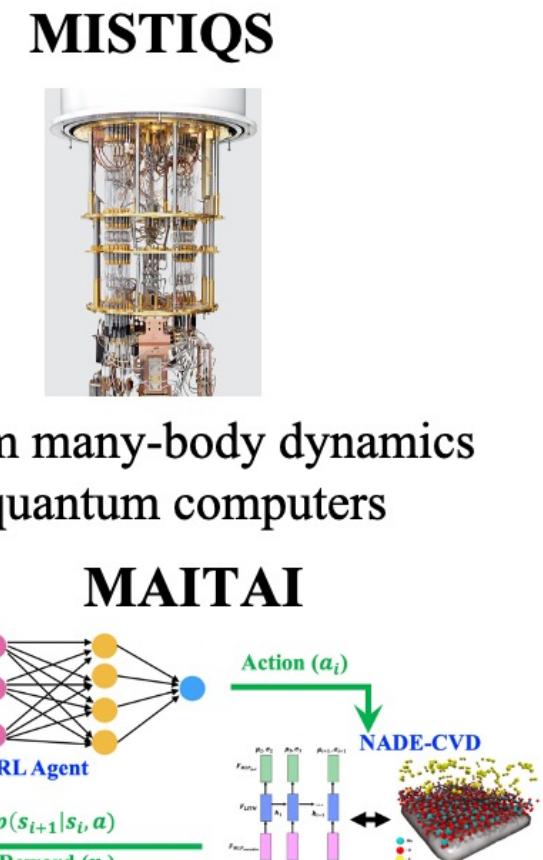
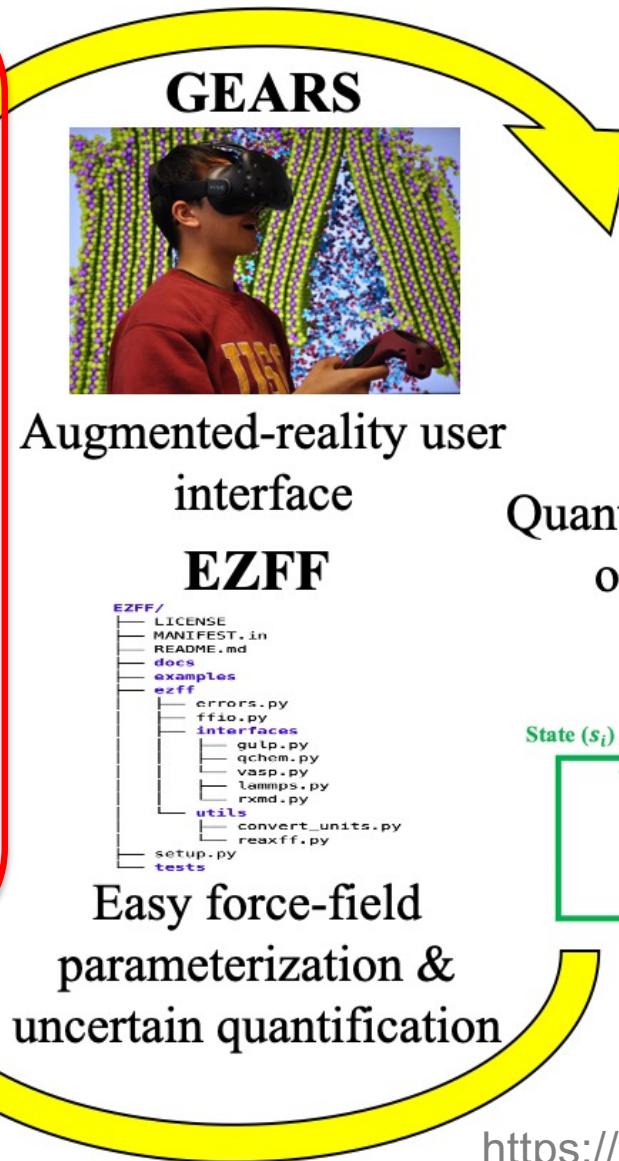
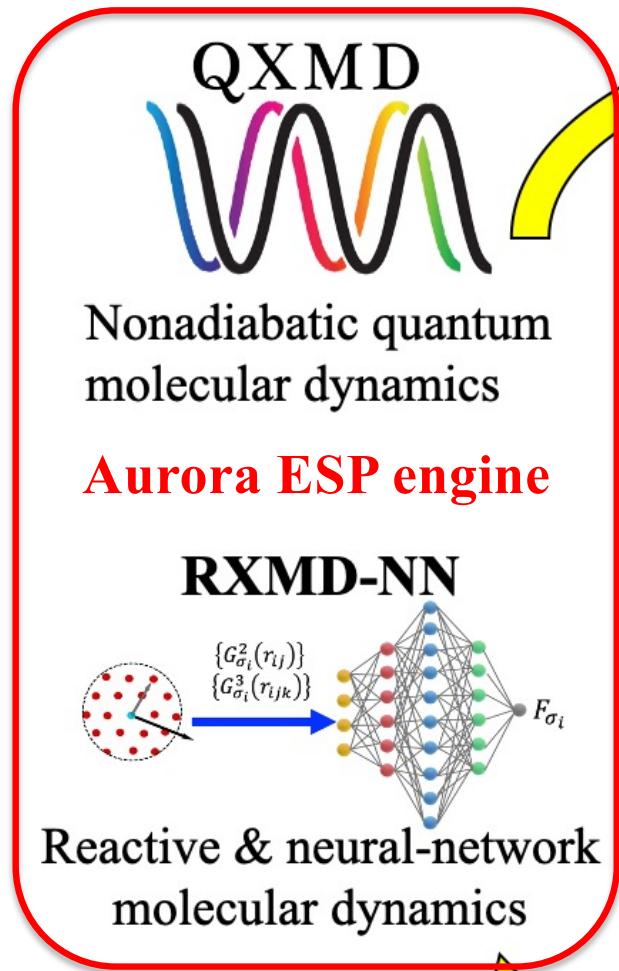
# Nuclear-Quantum NNQMD

- **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



# AIQ-XMaS Software Suite

## AI & Quantum-Computing Enabled Exa Quantum Materials Simulator

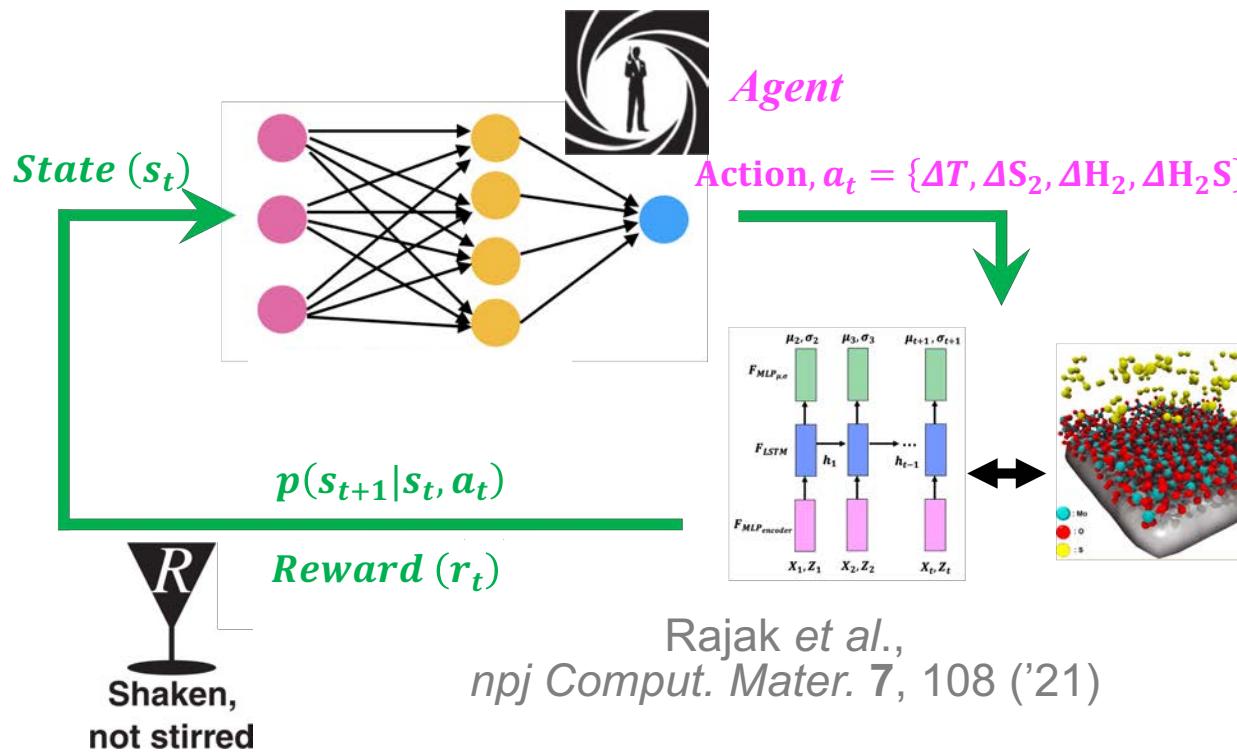


You will obtain hands-on training on QXMD

<https://cybermagics.netlify.app/software>

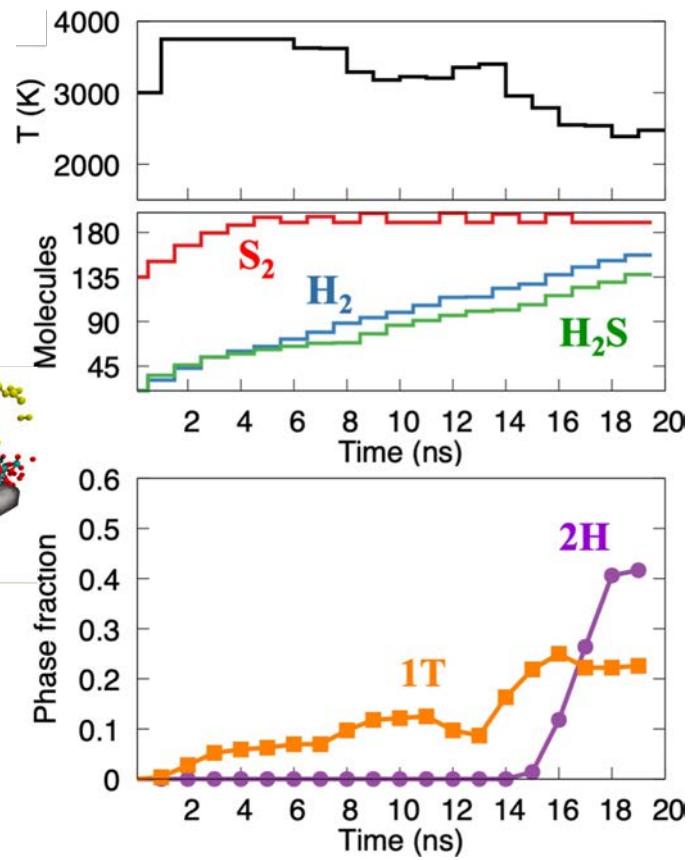
# Reinforcement Learning for Growth

- 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)

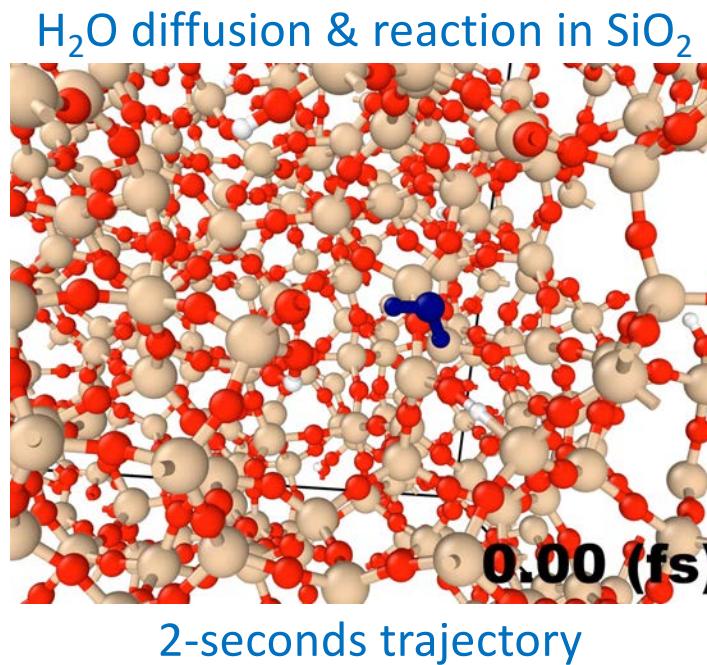
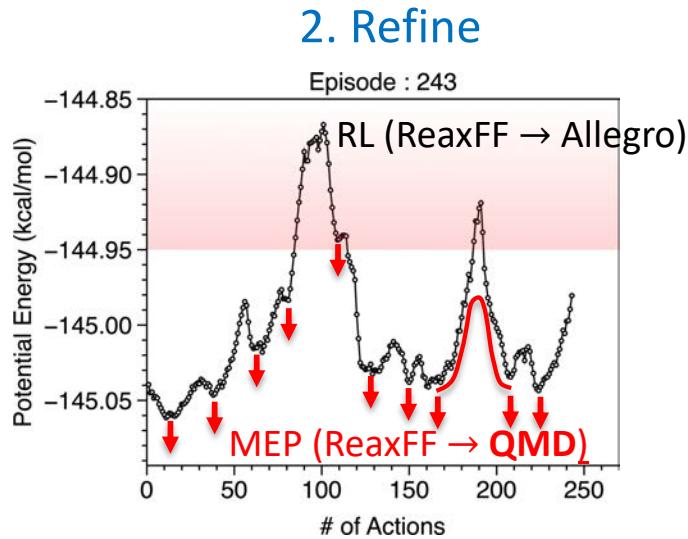
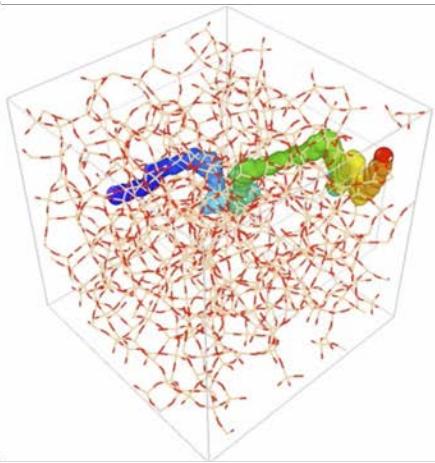


# RL to Enable Long-Time Dynamics

- 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

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

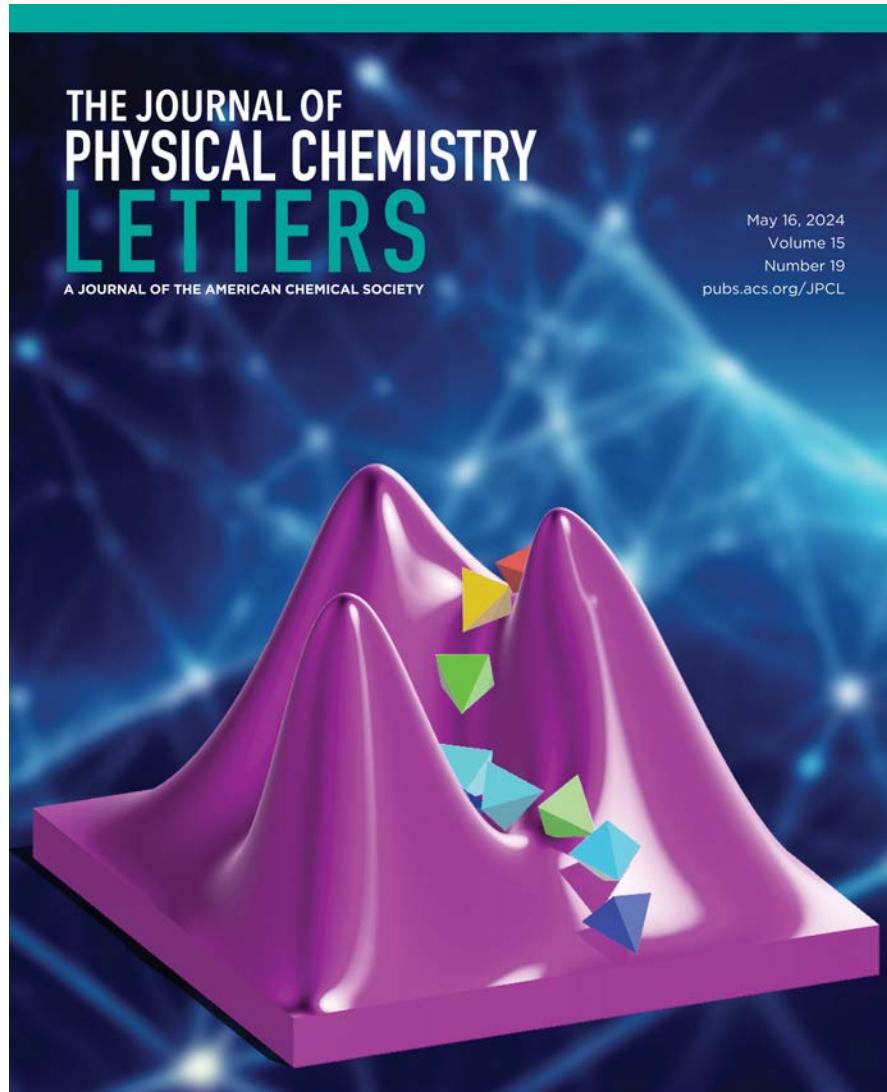
1. Explore



*AI for long time!*

# RL to Enable Long-Time Dynamics

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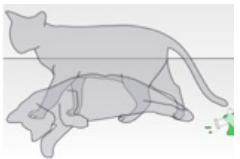


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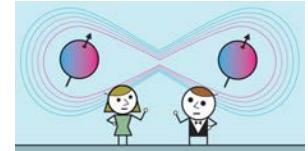
[www.acs.org](http://www.acs.org)

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

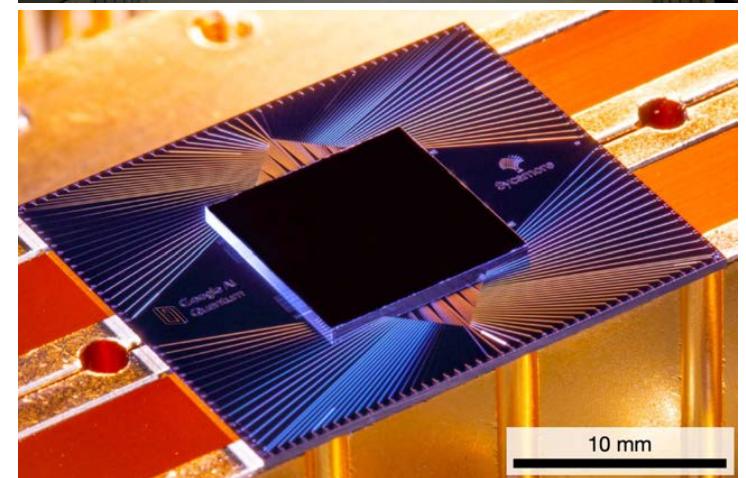
# Quantum Computing (QC) for Science



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



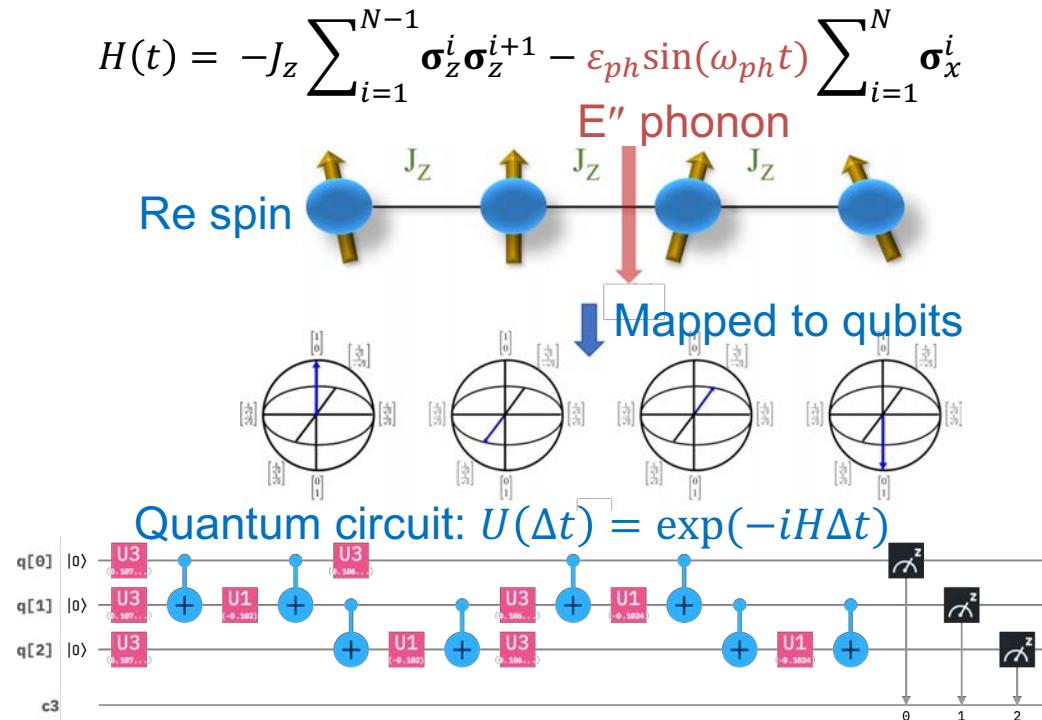
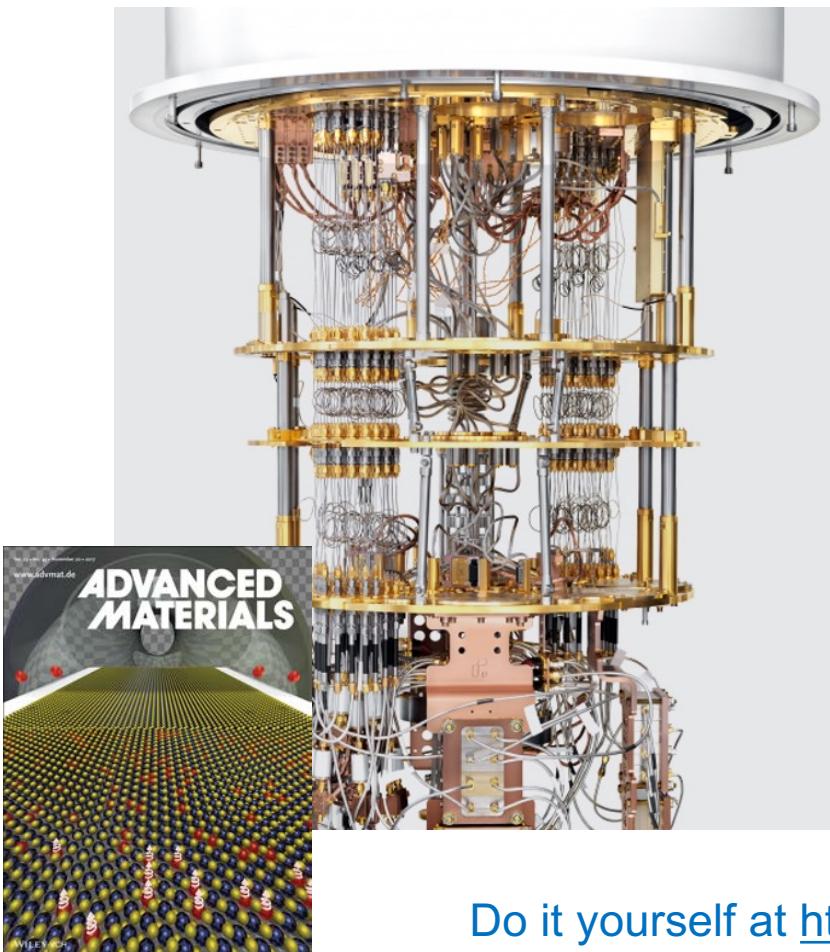
- U.S. Congress (Dec. 21, '18) signed National Quantum Initiative Act to ensure leadership in quantum computing & its applications
- Quantum supremacy demonstrated by Google  
F. Arute, *Nature* 574, 505 ('19)
- Quantum computing for science:  
*Universal simulator of quantum many-body systems*  
R. 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<sub>2</sub> monolayer to realize desired material properties on demand, thereby pushing the envelope of “quantum materials science”

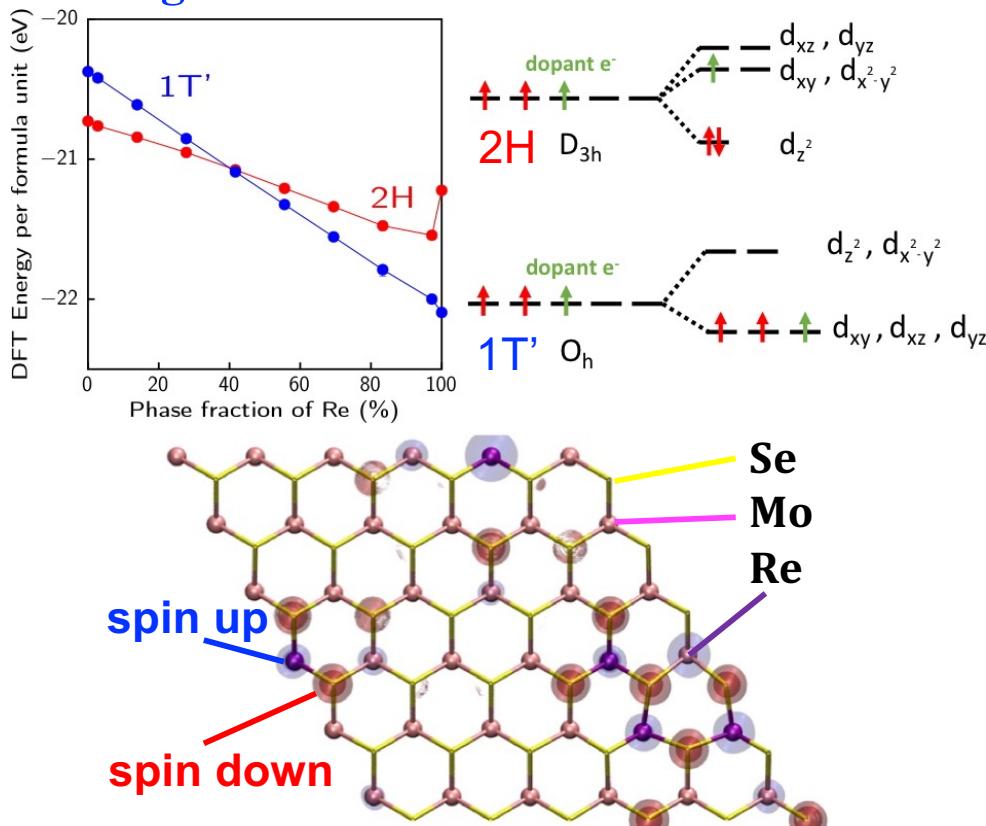


```
32 ...#define the two non-commuting terms that comprise the Hamiltonian-
33 ...Hz = PauliTerm("Z", 0, epsilon_0)-
34 ...Hy = PauliTerm("Y", 0, epsilon_ph*np.sin(w_ph*t))- 
35 ...#exponentiate the terms of the Hamiltonian for use in Trotter approx-
36 ...exp_Hz = exponential_map(Hz)(delta_t/(2.0*hbar))-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

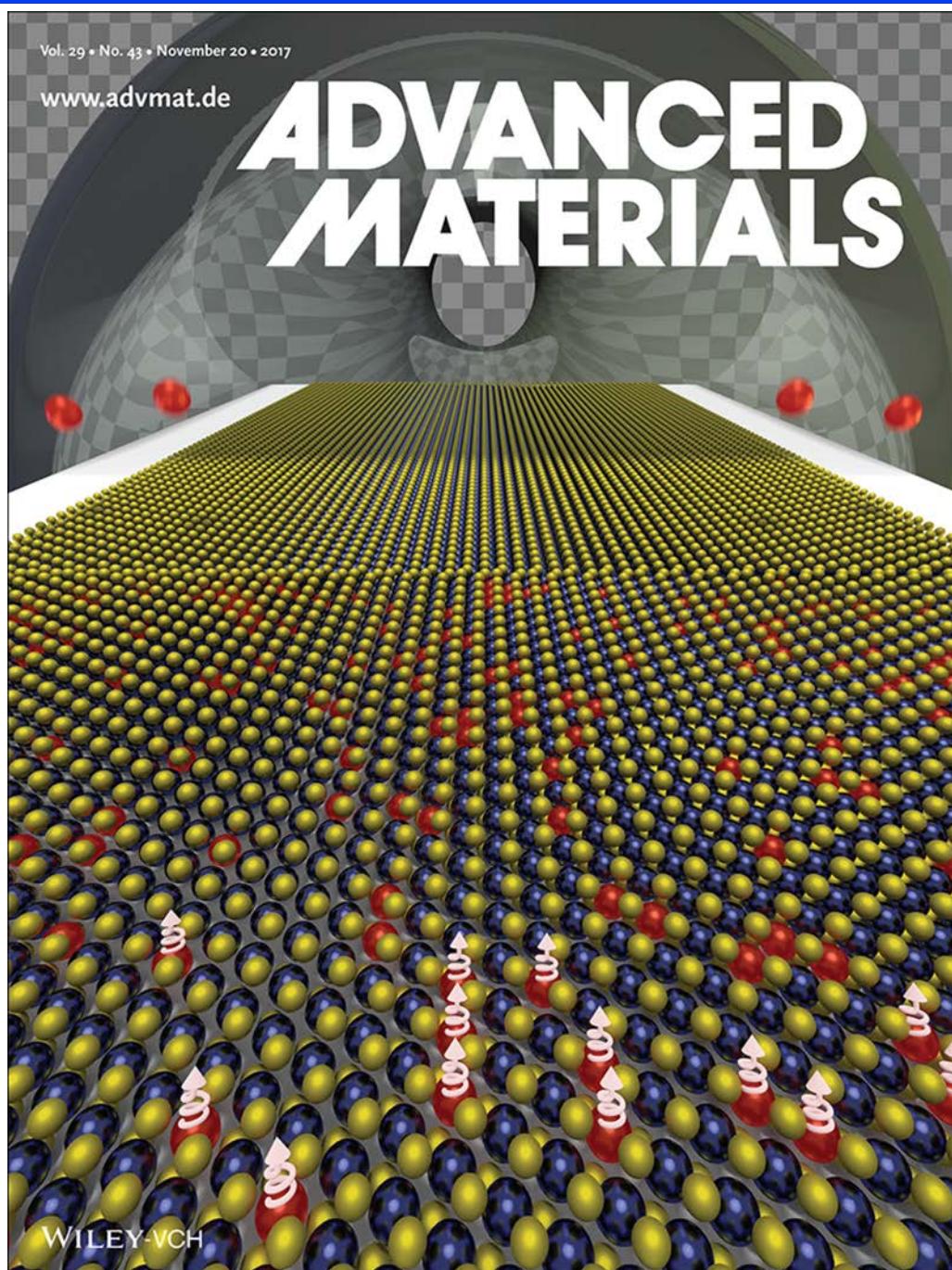
Do it yourself at <https://quantum-computing.ibm.com>

# Semiconductor-to-Metal Transition via Doping

- Experiment at Rice shows 2H-to-1T' phase transformation by alloying MoSe<sub>2</sub> 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)

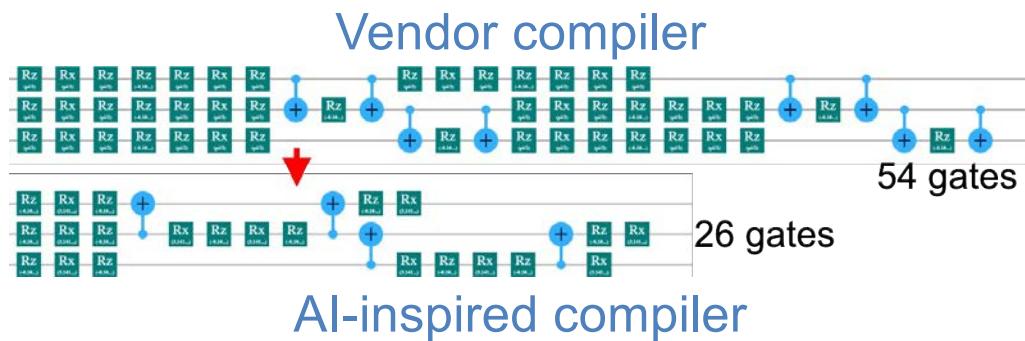


# Quantum Dynamics on Quantum Computers

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

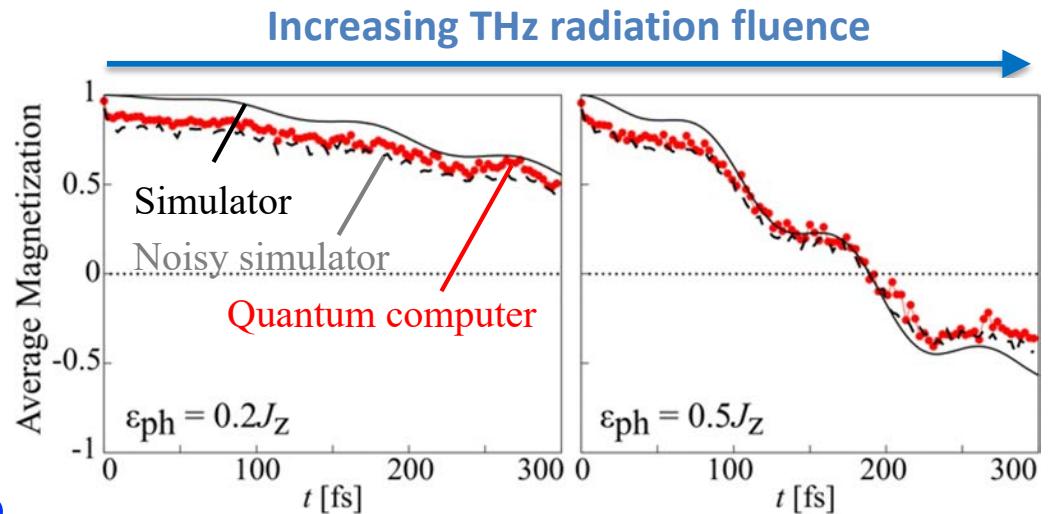
Bassman *et al.*,  
Phys. Rev. 101, 184305 ('20)

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

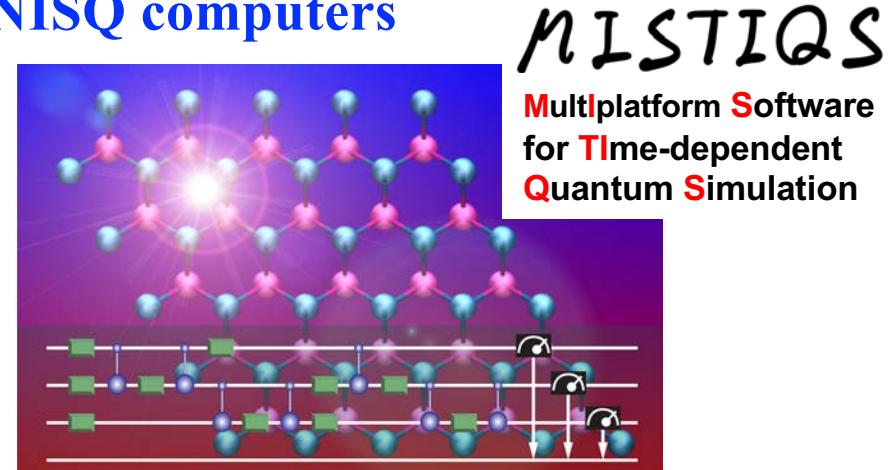


Bassman *et al.*,  
Quantum Sci. Tech. 6, 014007 ('21)

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

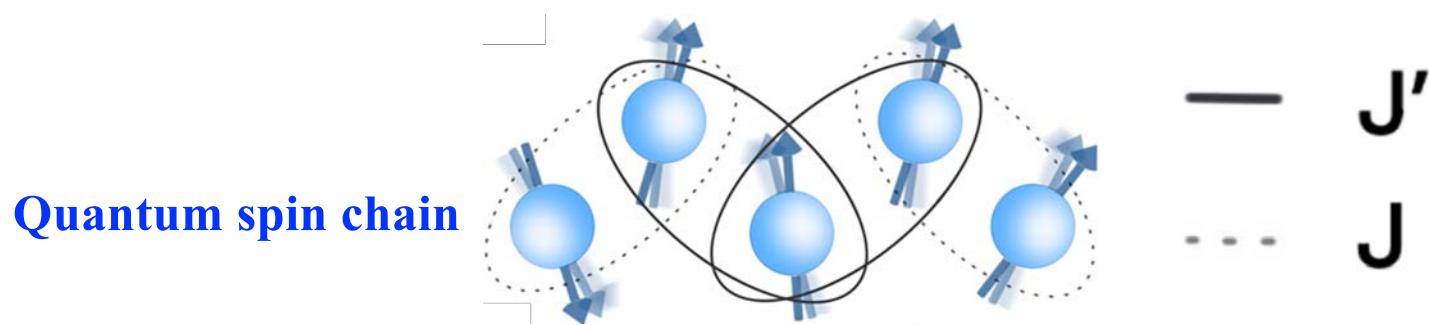


- Full-stack, cross-platform software for quantum dynamics simulations on NISQ computers



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

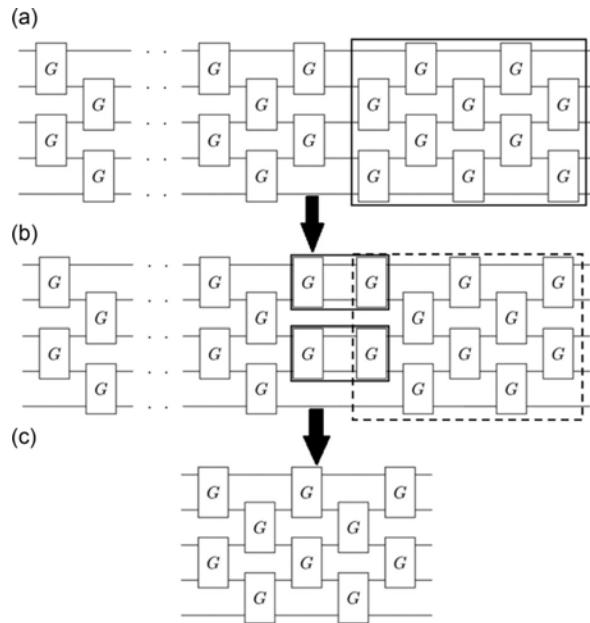
# Topological Quantum Dynamics



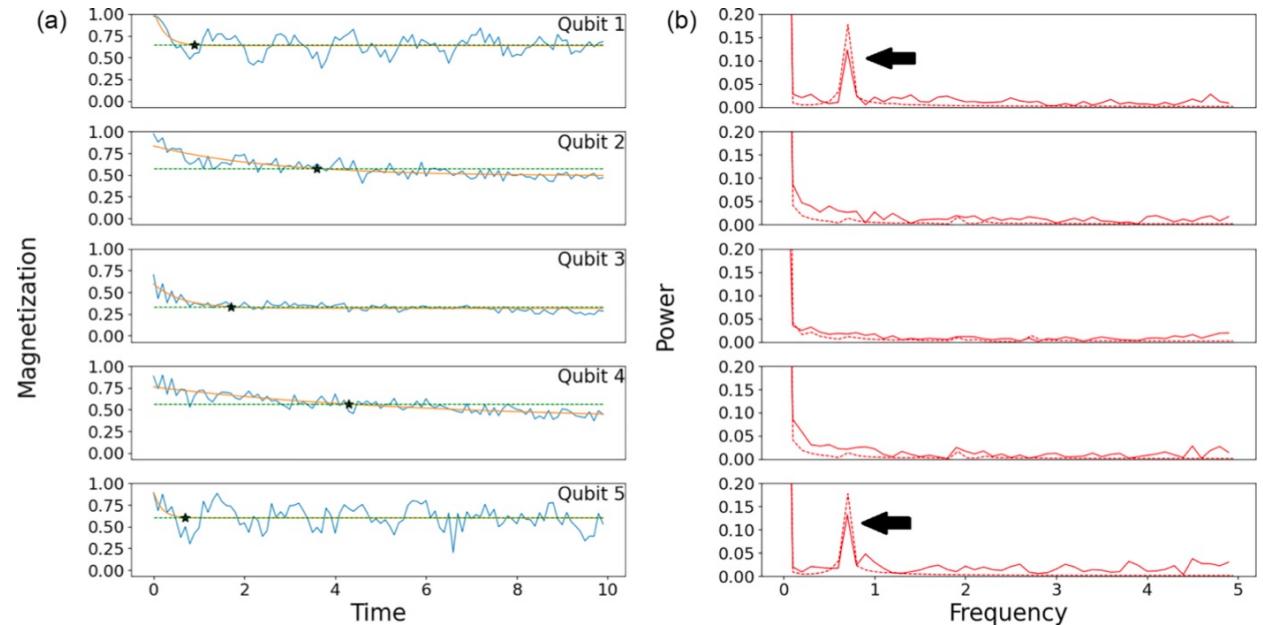
Quantum spin chain

—  $J'$   
- - -  $J$

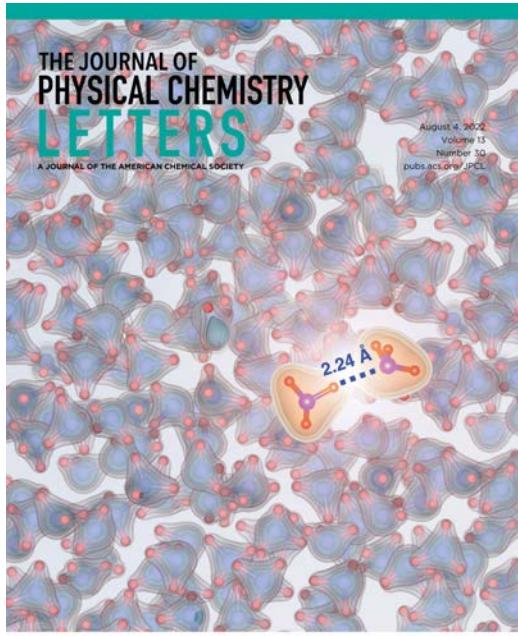
Constant-depth quantum circuit



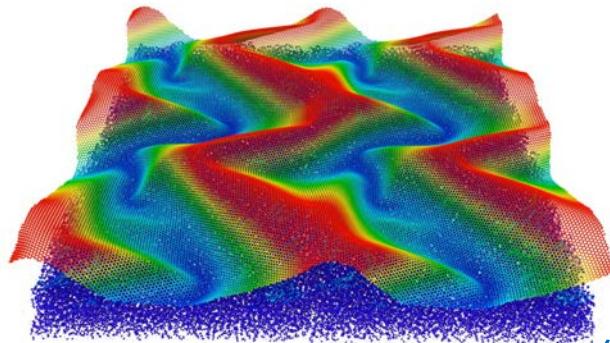
Topological surface mode



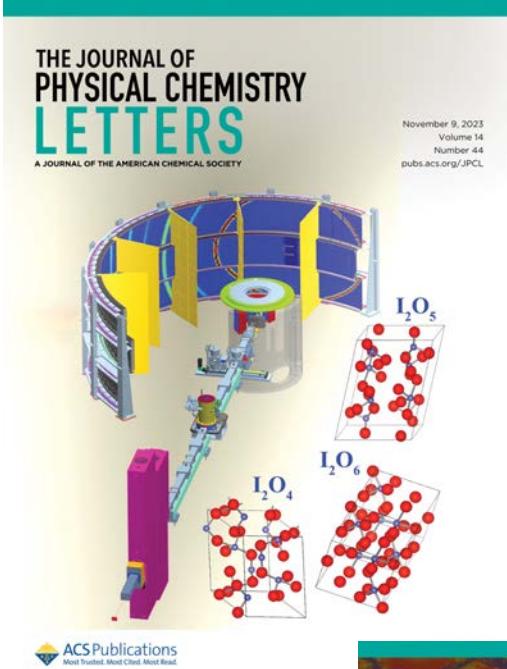
# Other Applications



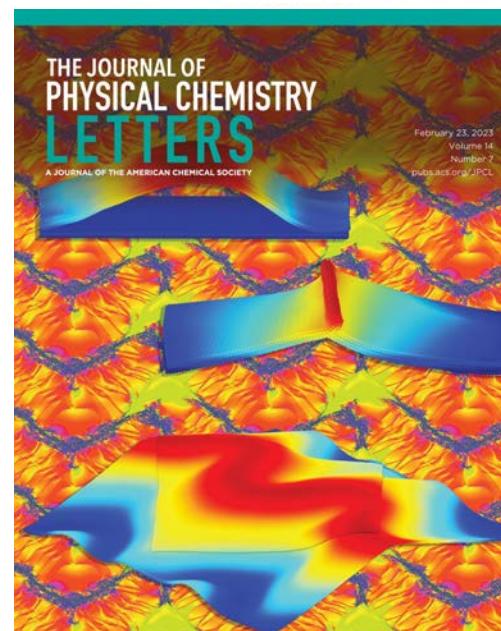
Green ammonia for  
renewable power  
(Aug. 4, '22)



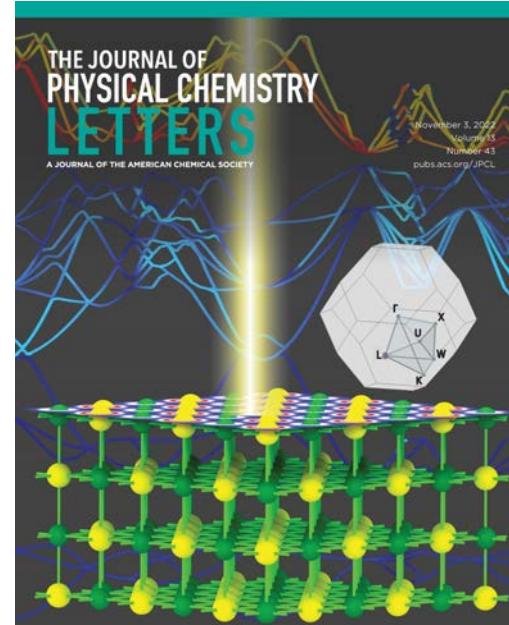
Strain self-  
assembly of  
2D  
metasurface  
(Feb. 23, '23)



Neutron  
scattering  
(Nov. '23)

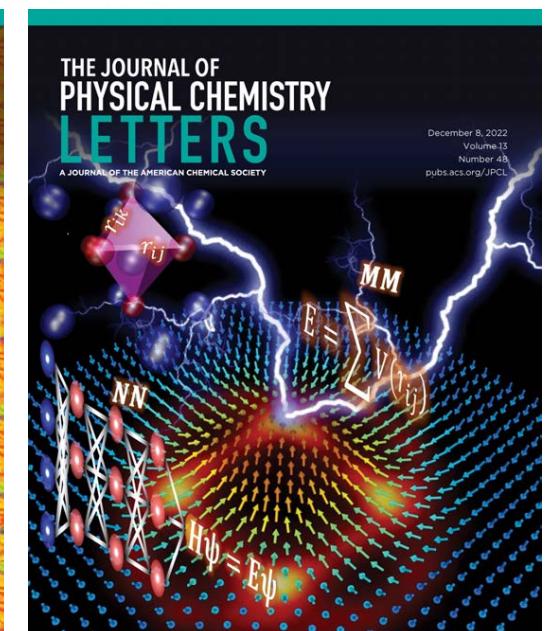


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Phase-change  
memory  
(Nov. 3, '22)

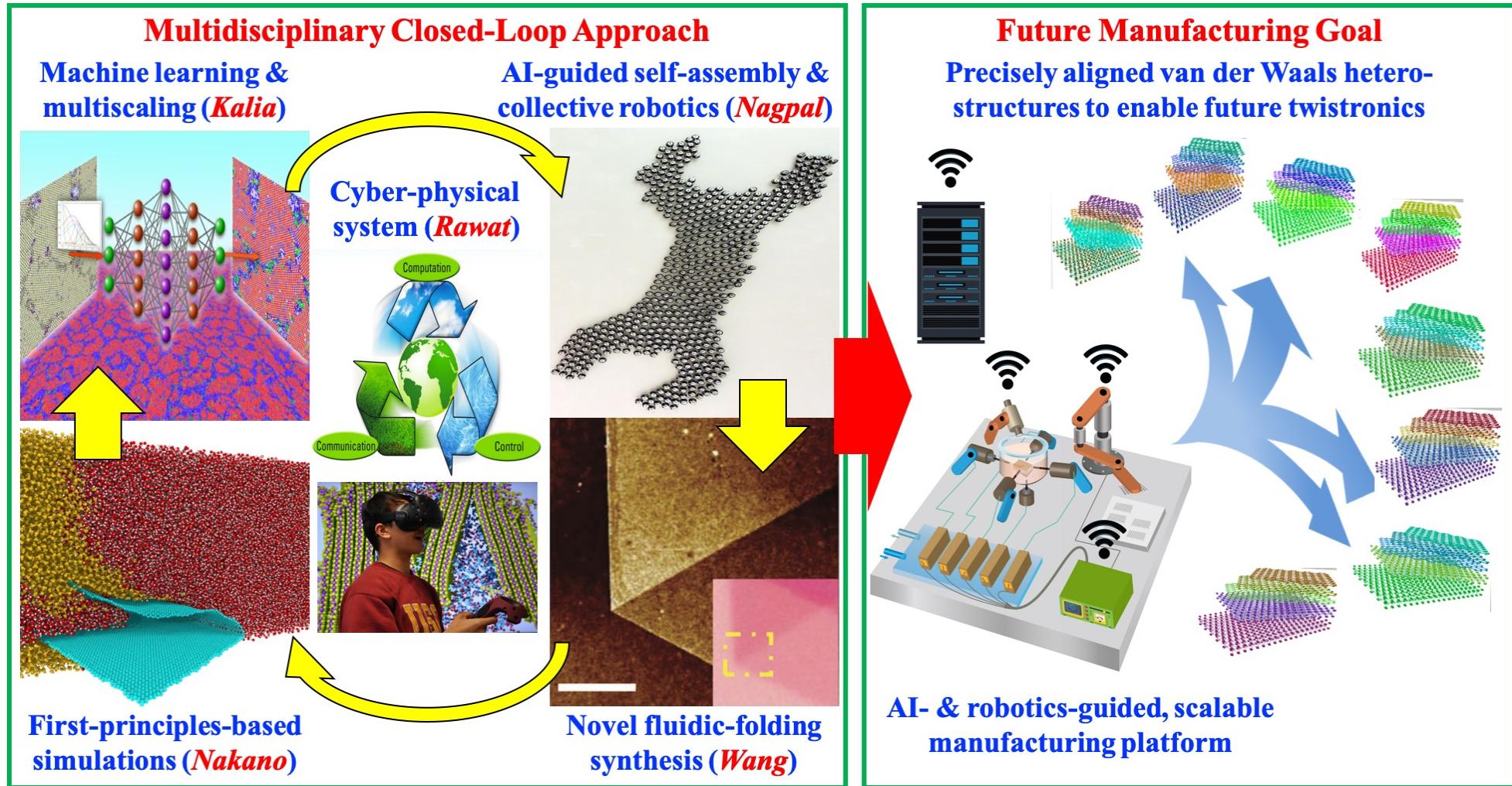
Topotronics  
(Dec. 8, '22)



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# 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)

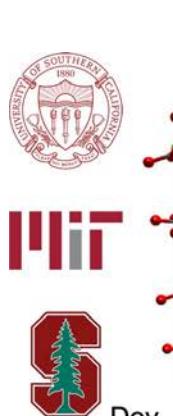
# NSF Future Semiconductors Project

- 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

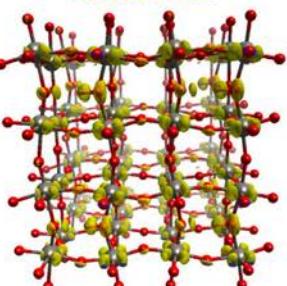
## Conductivity switch in H-doped WO<sub>3</sub>

### Protonic synapse (MIT)

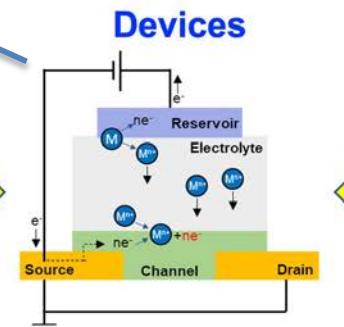
[Onen et al., *Science* 377, 539 ('22)]



#### Materials



#### Devices



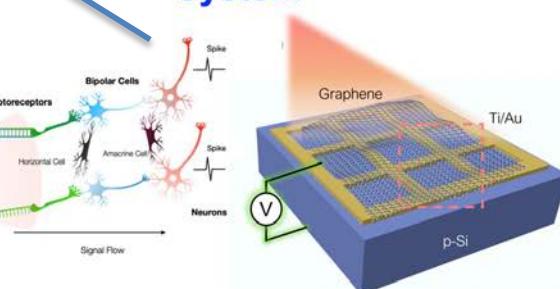
Dev, Krishnamoorthy, Kalia, Li

Wang, Yang, Zhang, Yildiz

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

Oscillatory retinal neural network (USC)  
[DOI: 10.21203/rs.3.rs-2935296/v1]

#### System

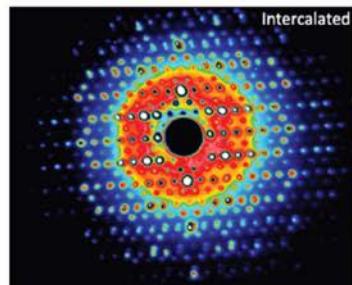


del Alamo, Kapadia, Lin, Raina



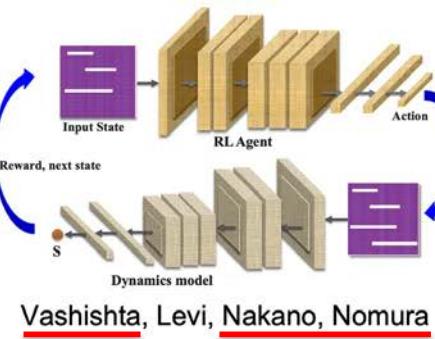
## UE/XS & AI/CS guided co-design and FeSe workforce support

### UE/XS



Lindenberg, Wei

### AI/CS



Vashishta, Levi, Nakano, Nomura

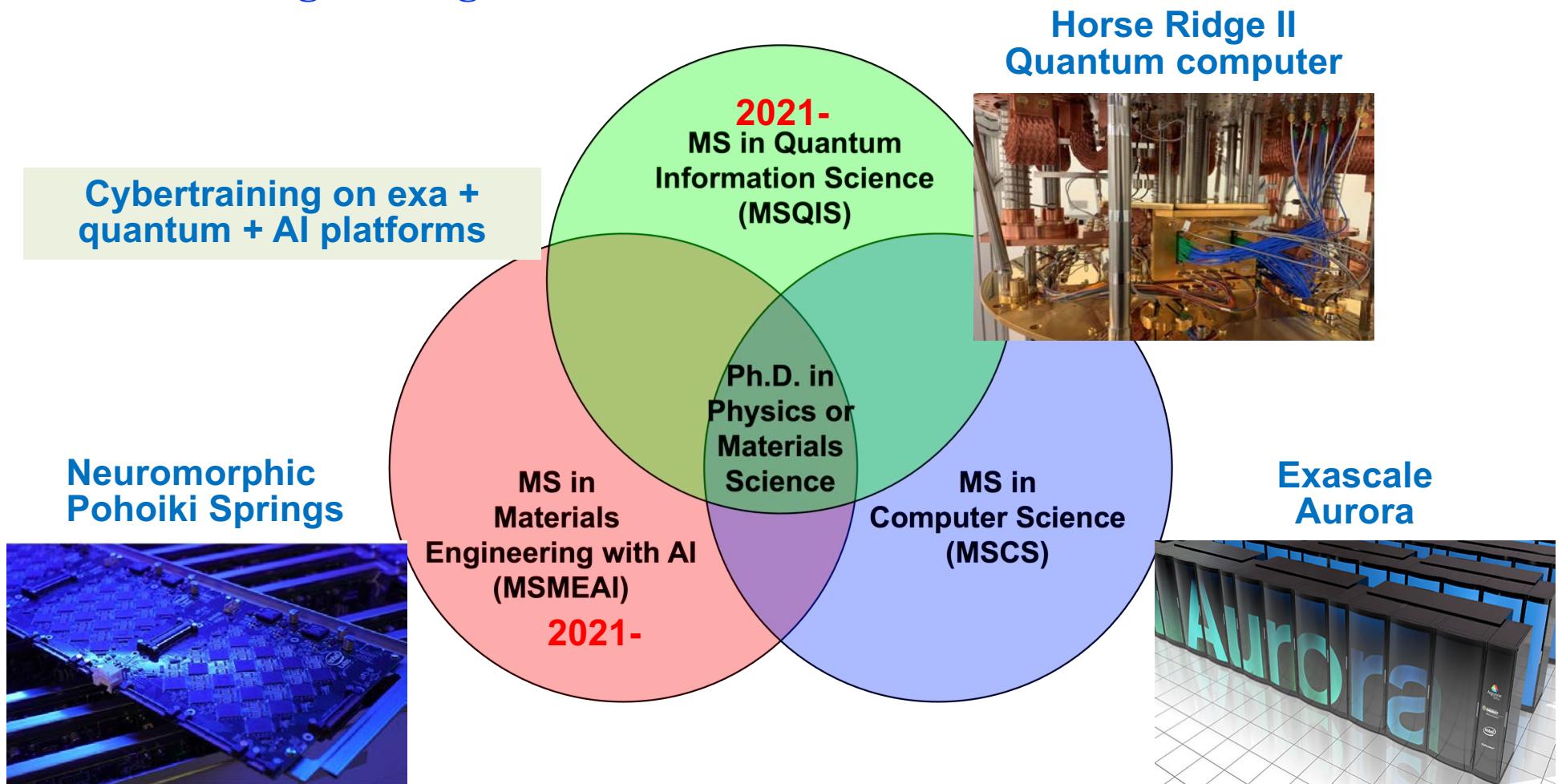
### FuSe Workforce Development



Nikias, Katehi, Raghavendra, Rawat

# Training Cyber-Science Workforce

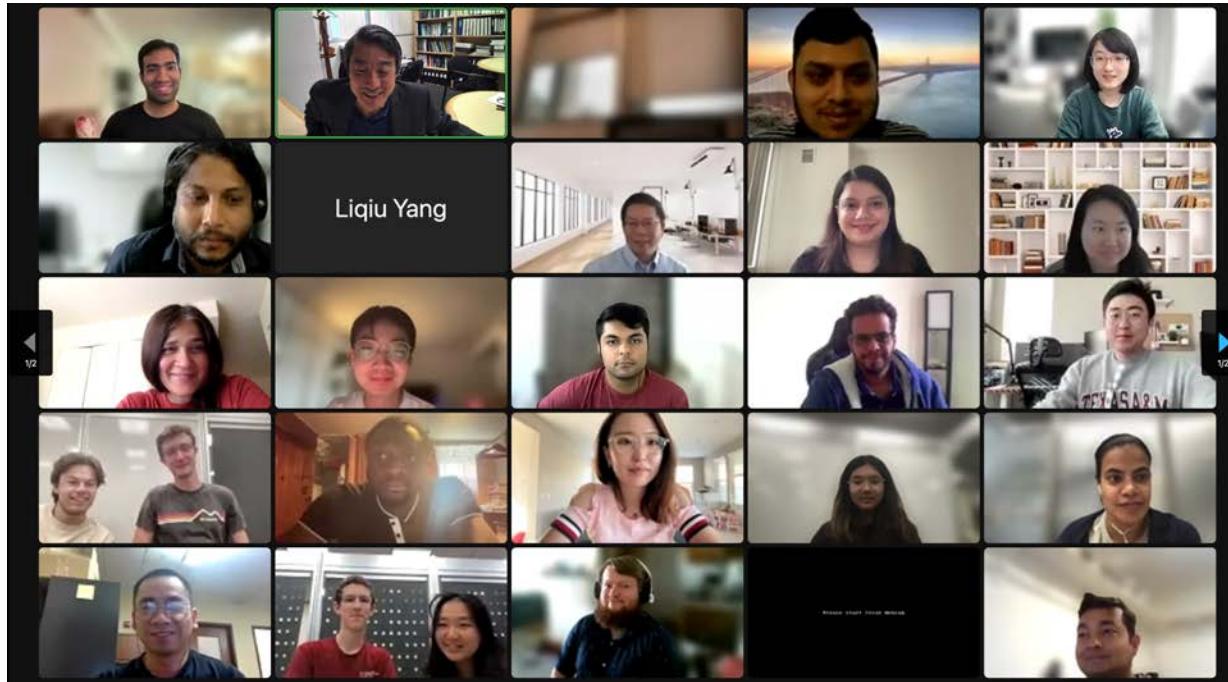
- New generation of computational scientists at the **nexus of exascale computing, quantum computing & AI**
- Unique dual-degree program: Ph.D. in materials science or physics, along with MS in computer science, MS in quantum information science or MS in materials engineering with AI



# 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

