

# 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

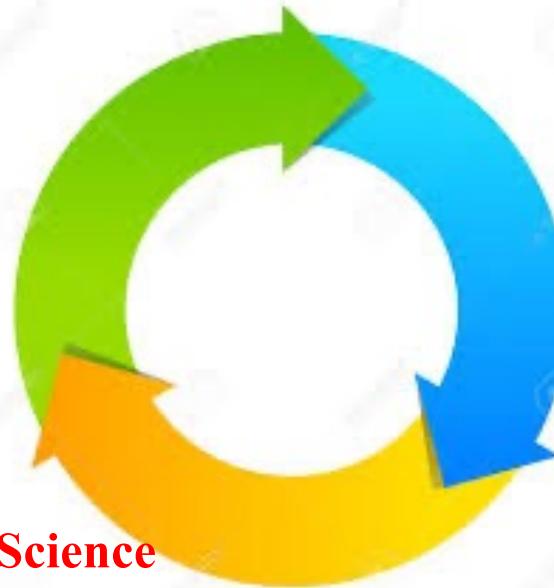


# Changing Computing Landscape for Science

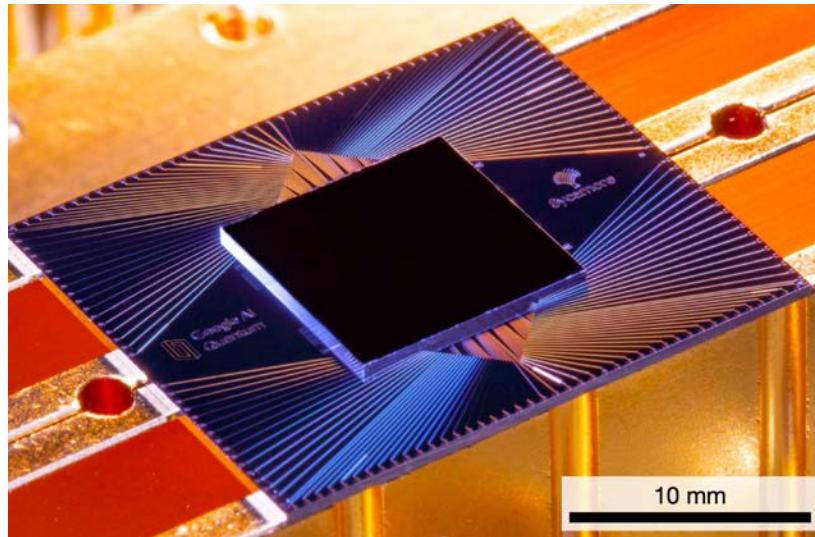
## Post-exascale Computing for Science



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>

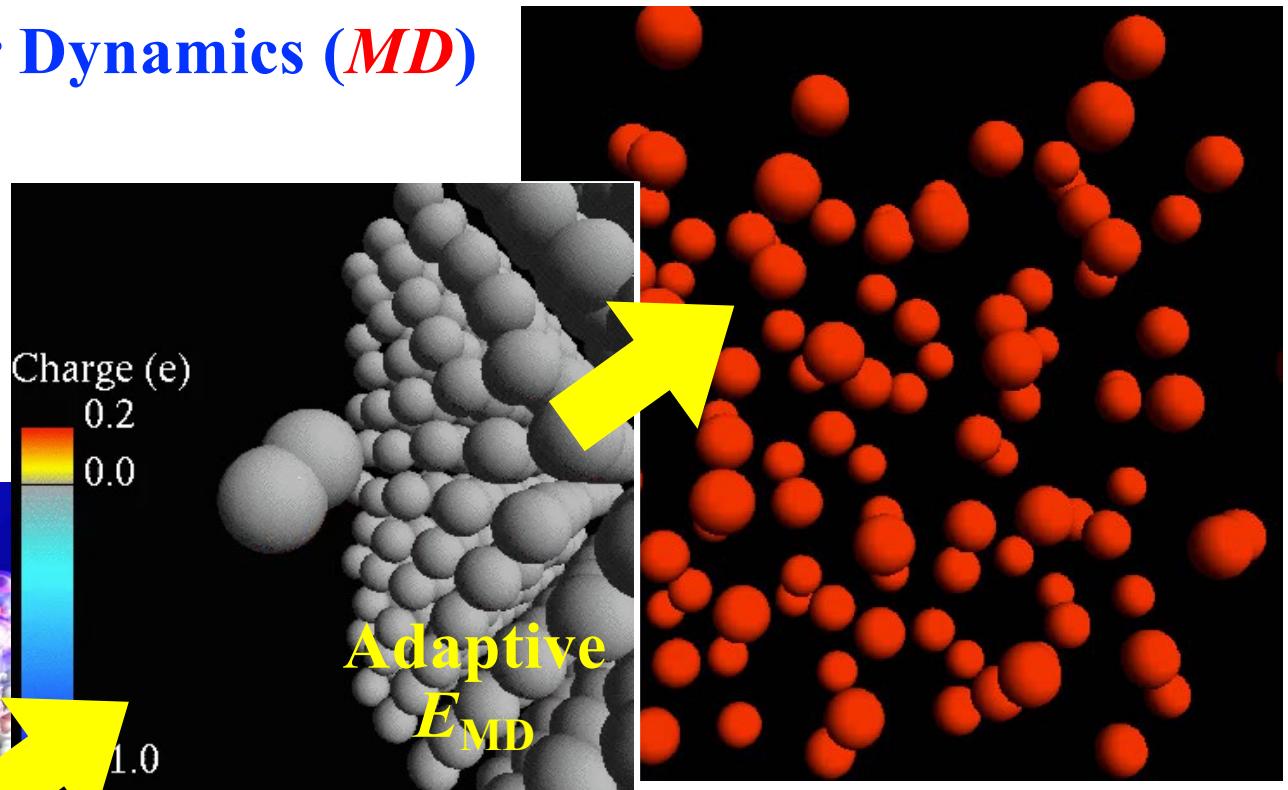
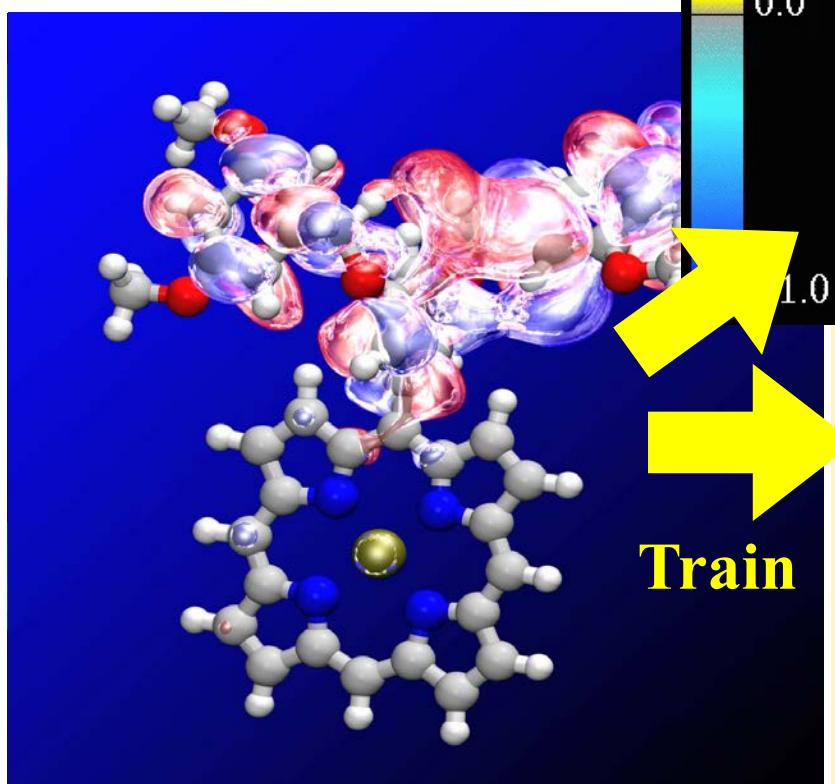
- New School of Advanced Computing: <https://sac.usc.edu>
- 30 senior & 60 junior & mid-level hires

# Molecular Dynamics & Machine Learning

## Molecular Dynamics (*MD*)

### Reactive MD (*RMD*)

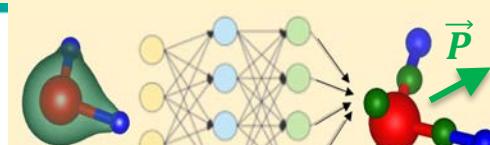
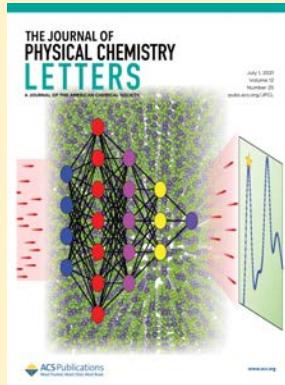
### Nonadiabatic quantum MD (*NAQMD*)



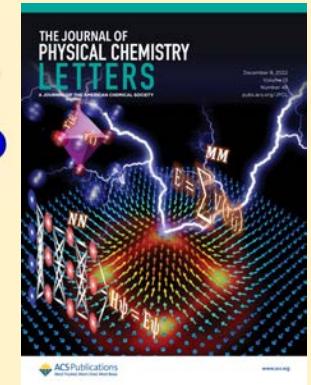
Adaptive  
 $E_{MD}$

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

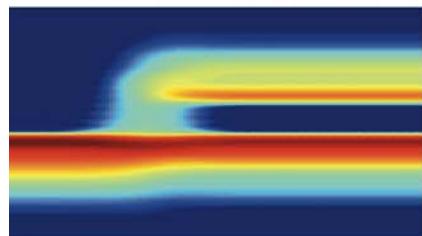
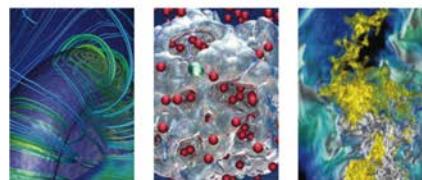
Train



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



# BES



# 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

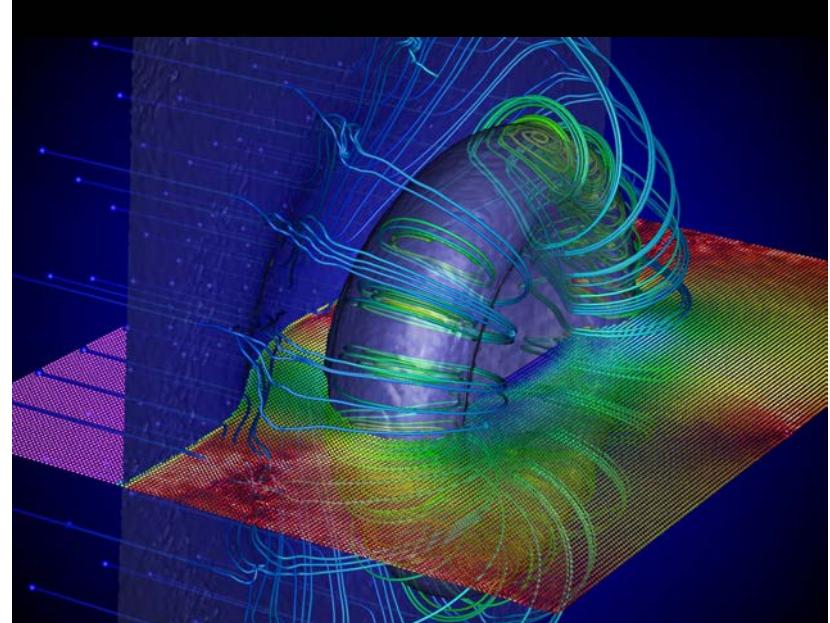
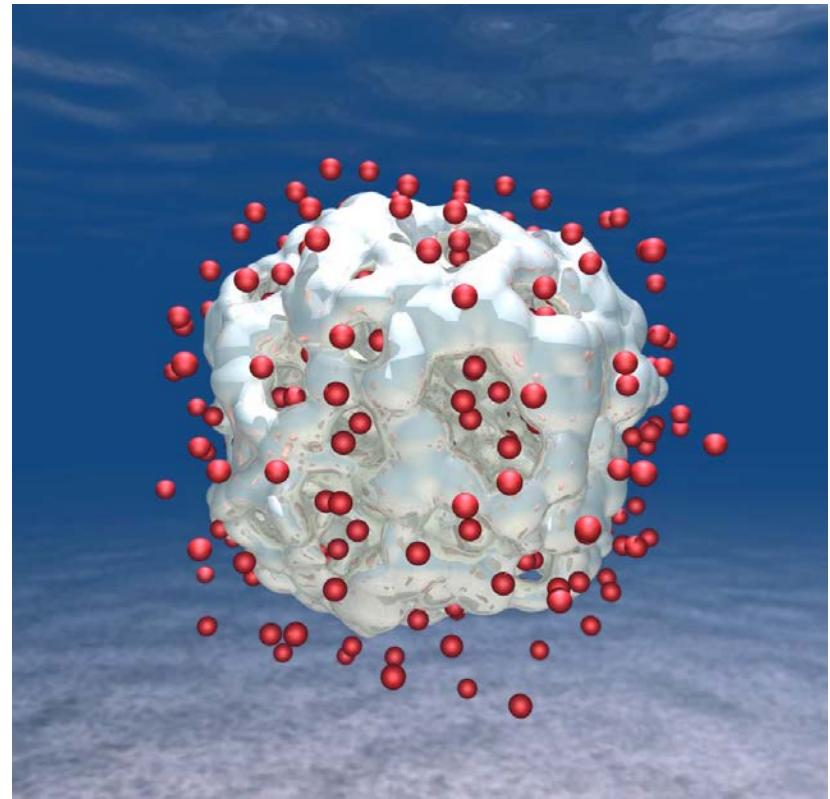
Shimamura *et al.*,  
*Nano Lett.*  
14, 4090 ('14)

*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)



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

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

Density functional theory (DFT)

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

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

$O(C^N)$   $\rightarrow$   $O(N^3)$   
1  $N$ -electron problem  $\rightarrow$   $N$  1-electron problems  
**intractable**  $\rightarrow$  **tractable**

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

## $O(N)$ 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

№ 20

## ANNALEN DER PHYSIK VIERTE FOLGE. BAND 84

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

# Academic Genealogy

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## 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

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



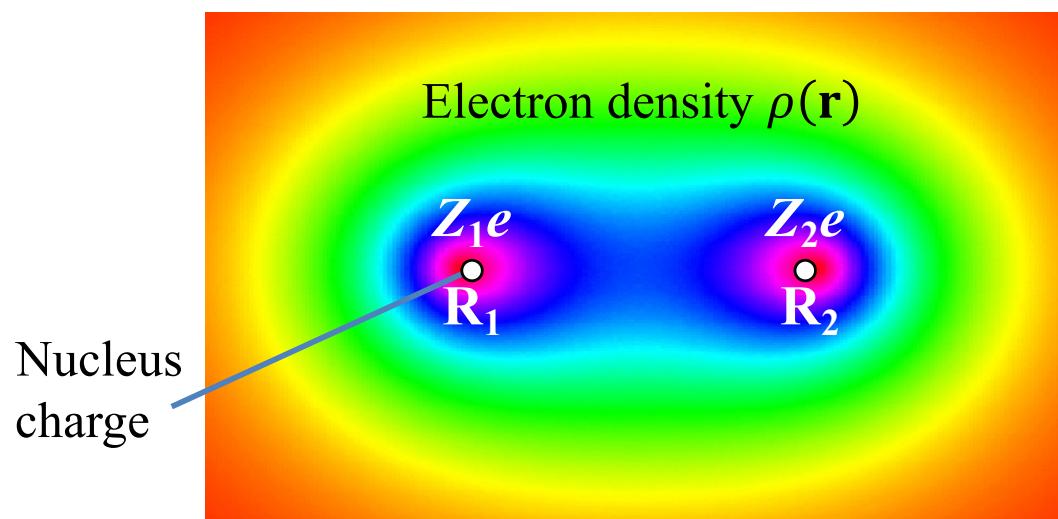
Sholl & Steckel, Density Functional Theory: A Practical Introduction ('23)

# 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**

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

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

exchange-correlation (xc) potential

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

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

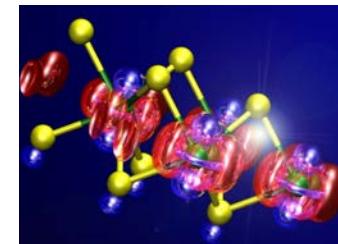
HSE: Heyd, Scuseria & Ernzerhof, *J. Chem. Phys.* **118**, 8207 ('03)

- Others supported by QXMD code: Select an appropriate functional for the material system & purpose

- DFT+U method for transition metals

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

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

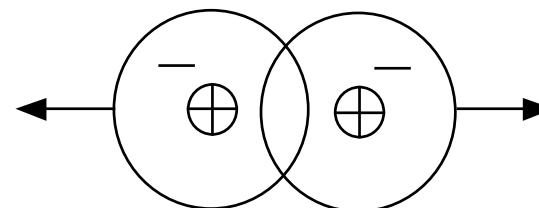


Localized  
d-electrons

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

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

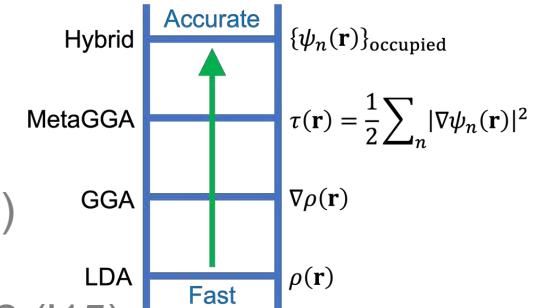
Grimme, *J. Chem. Phys.* **132**, 154104 ('10)



- vdW: Nonlocal correlation functional

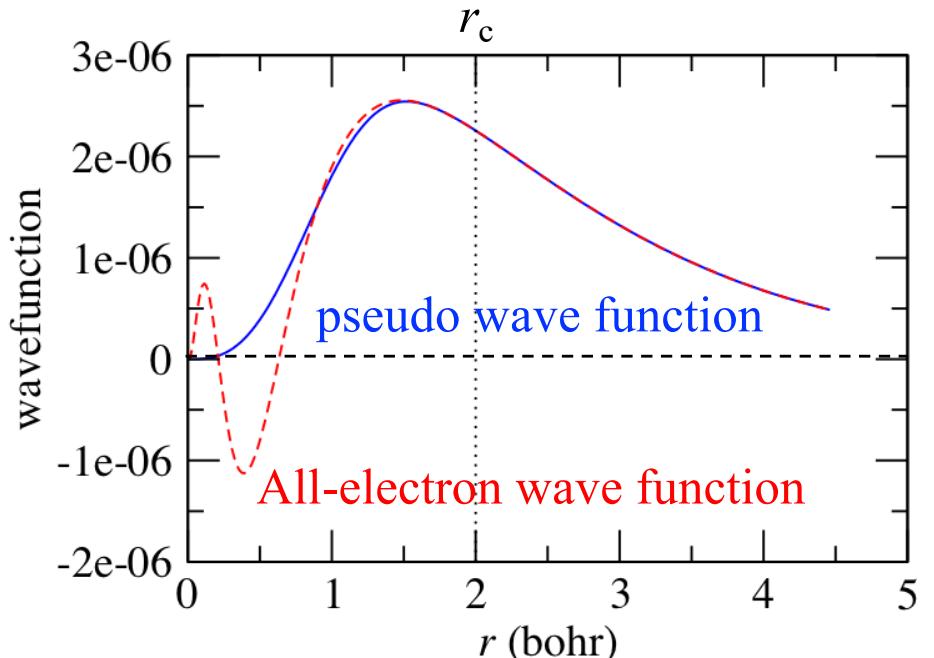
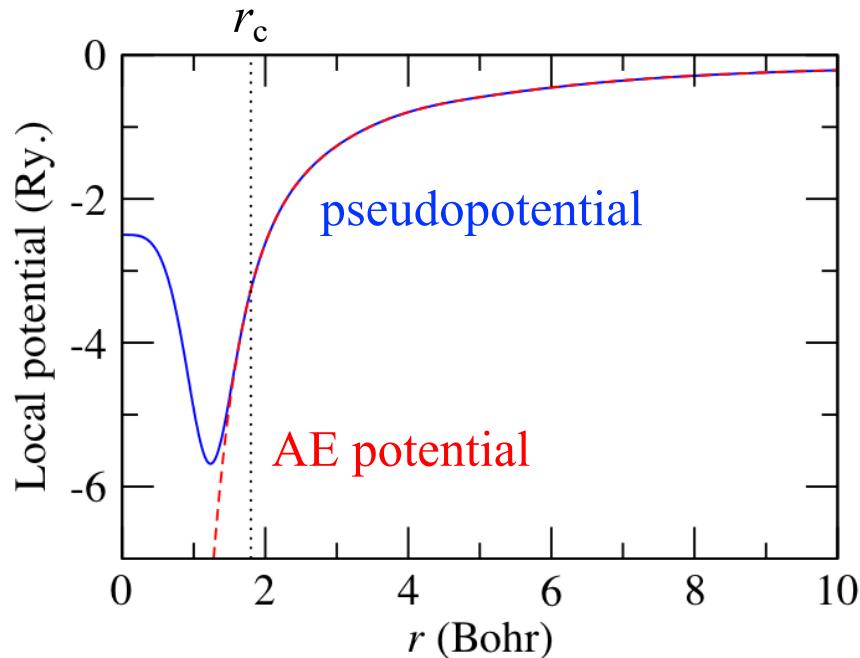
$$E_c^{\text{nl}} = \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \rho(\mathbf{r}) \phi(\mathbf{r}, \mathbf{r}') \rho(\mathbf{r}')$$

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



# Abstraction: Pseudopotential

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



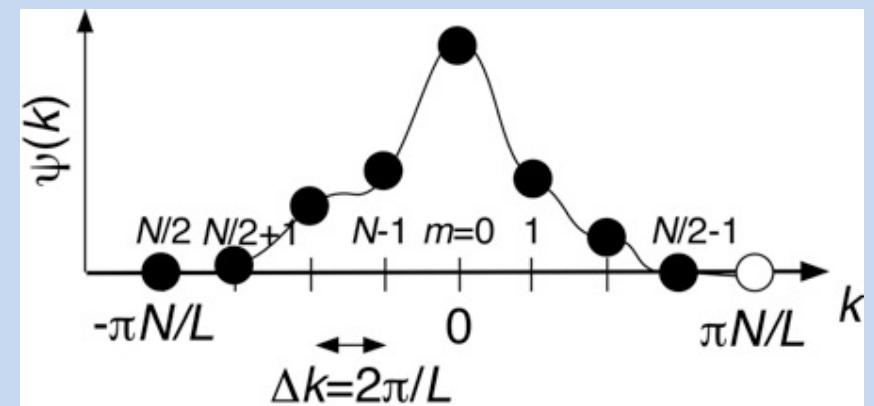
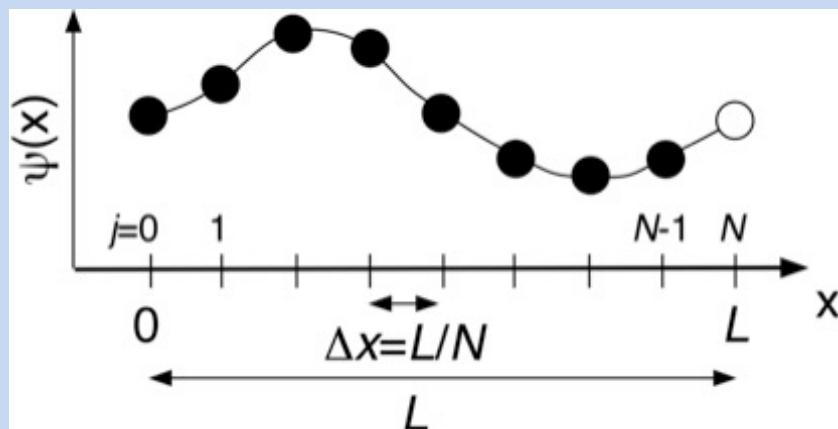
- Commonly used pseudopotentials
  - > Norm-conserving: Troullier & Martins, *Phys. Rev. B* **41**, 1993 ('91)
  - > Ultrasoft: Vanderbilt, *Phys. Rev. B* **41**, 7892 ('90)
  - > Projector augmented wave (PAW): Blochl, *Phys. Rev. B* **50**, 17953 ('94)

# 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

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

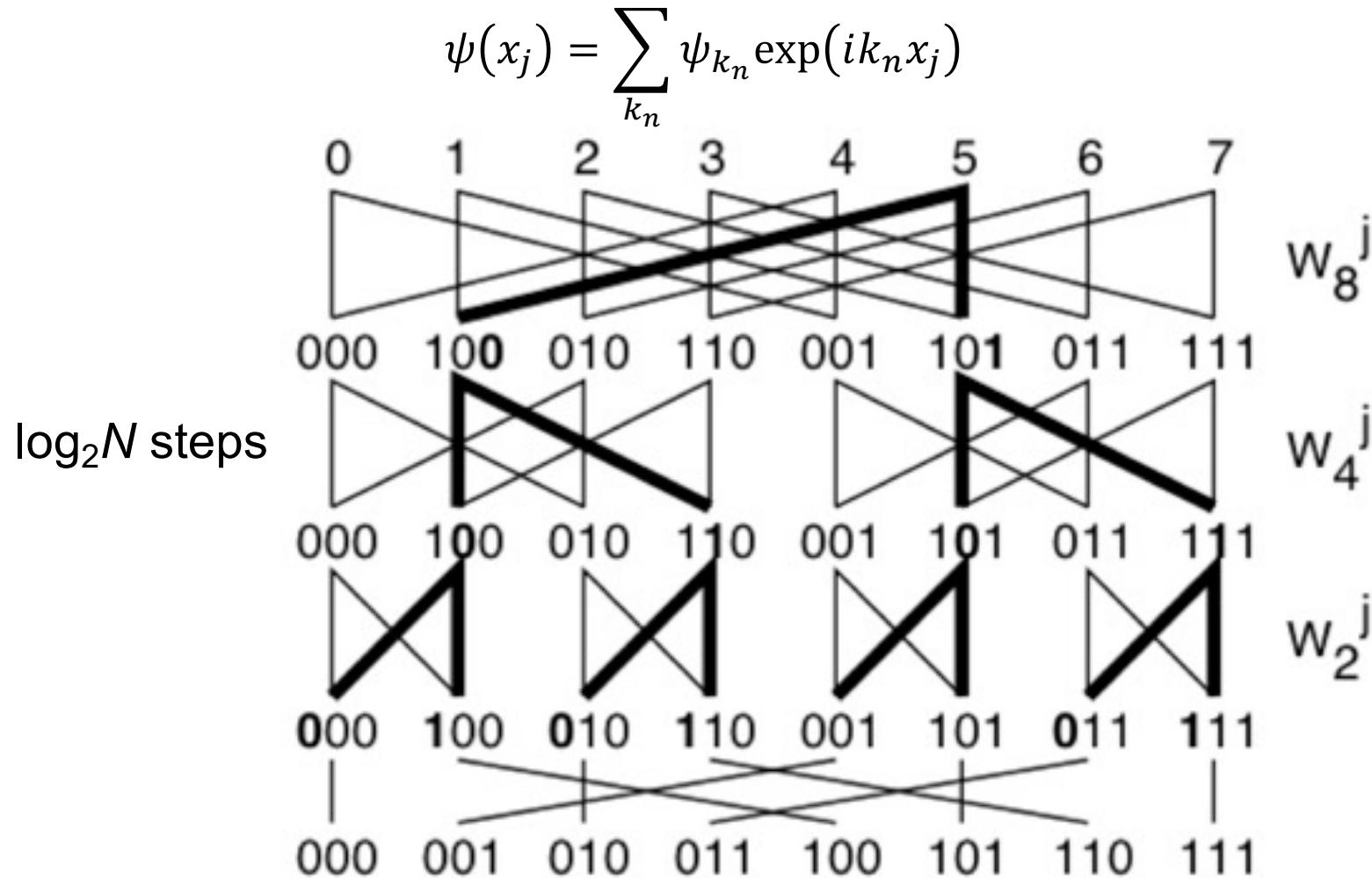
1D example



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

# Numerics: Fast Fourier Transform

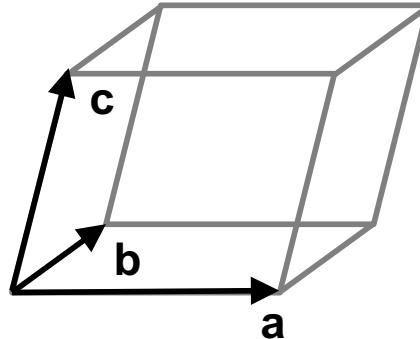
- $O(N \log N)$  fast Fourier-transform (FFT) algorithm is typically used to perform Fourier transform



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

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

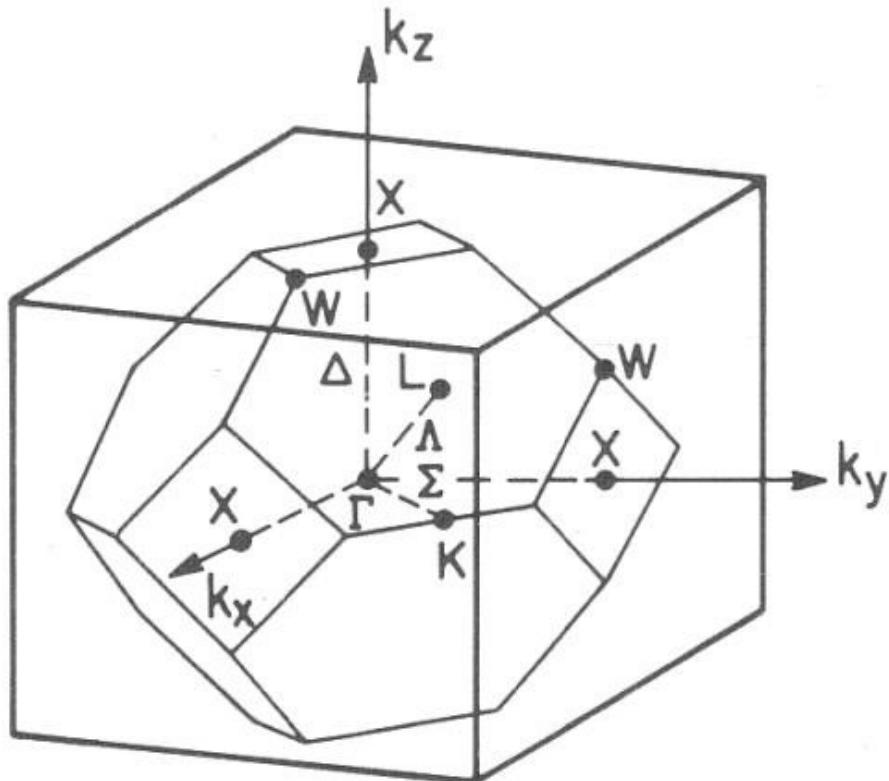
$\mathbf{k} \in$  first Brillouin zone in the reciprocal space

# Electronic Bands: Infinite Lattice

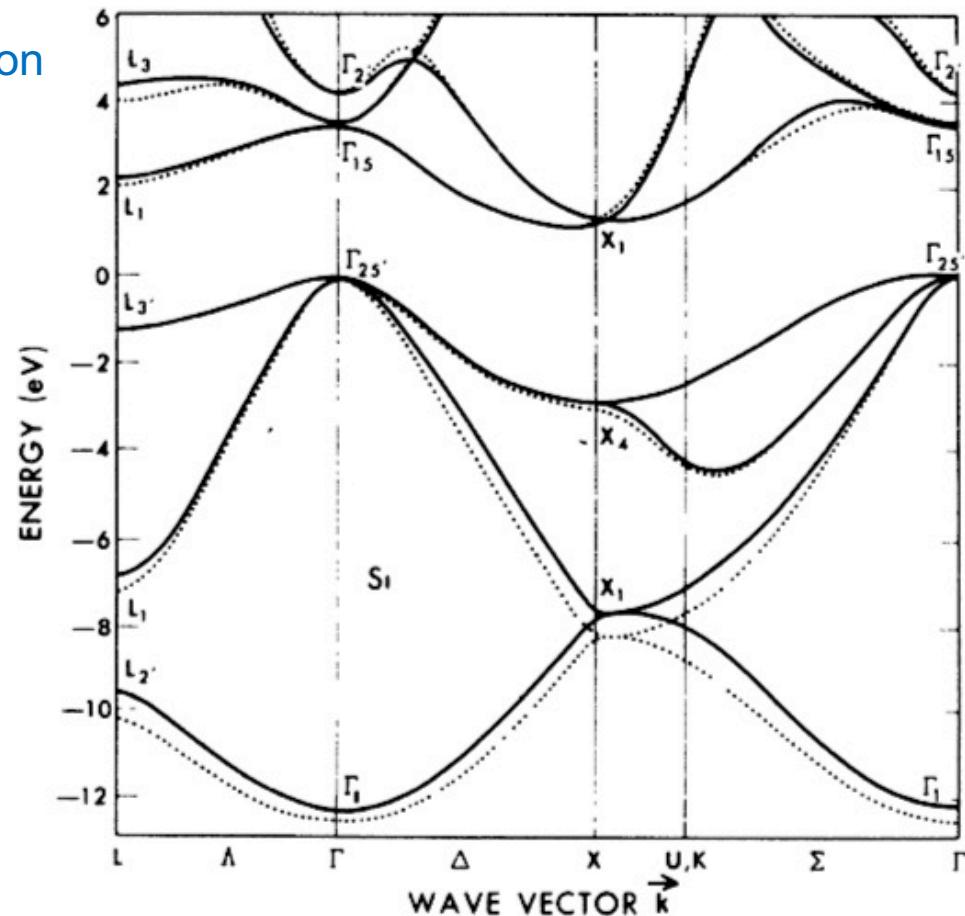
- **Bloch theorem:**  $\psi_{n\mathbf{k}}(\mathbf{r}) = \exp(i\mathbf{k} \cdot \mathbf{r})u_{n,\mathbf{k}}(\mathbf{r})$

## 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

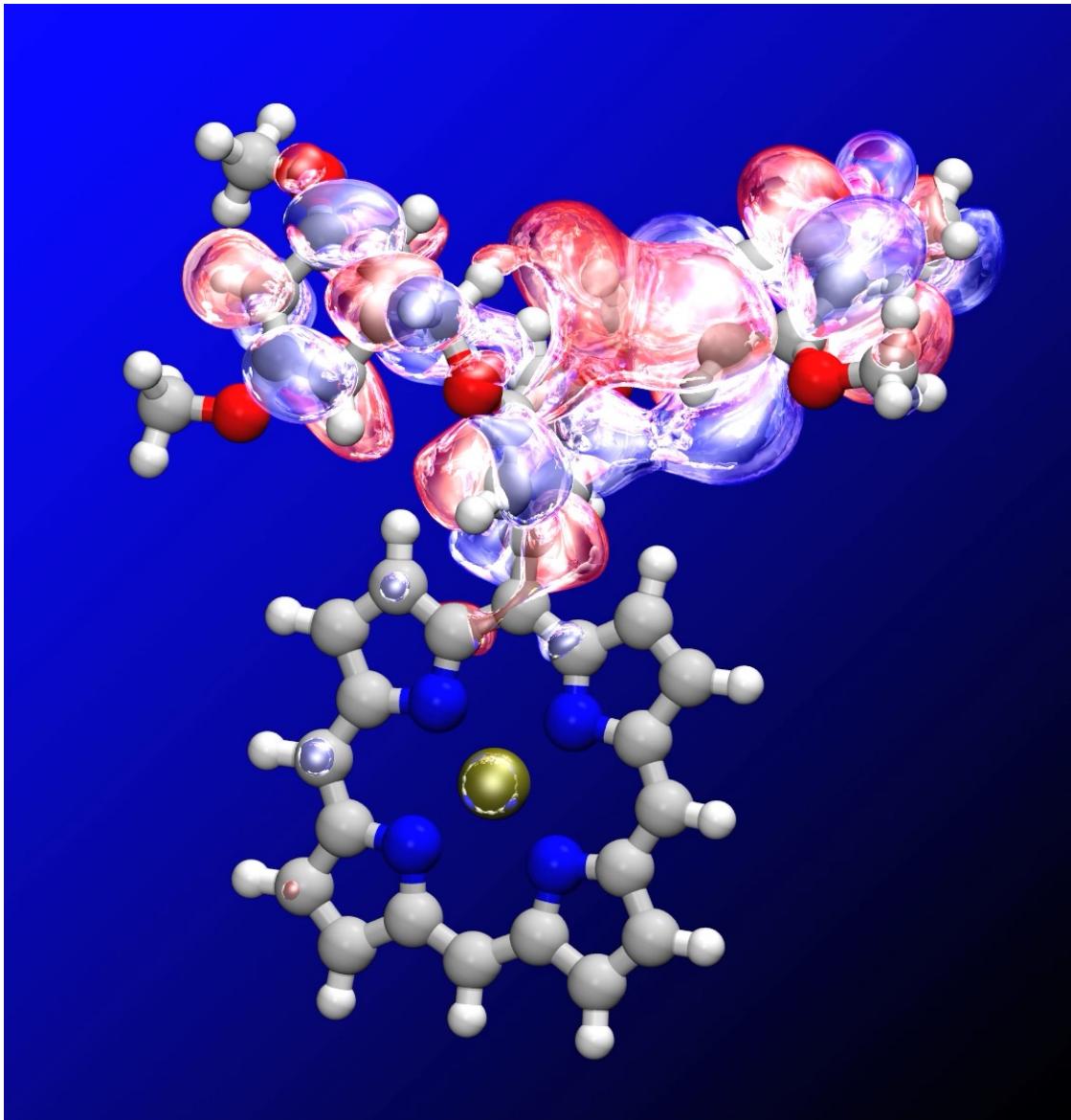
Given  $\{\psi_n, \epsilon_n\}$ ,  
determine  $\mu$  and  
compute  $\rho(\mathbf{r})$

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

Chemical potential

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

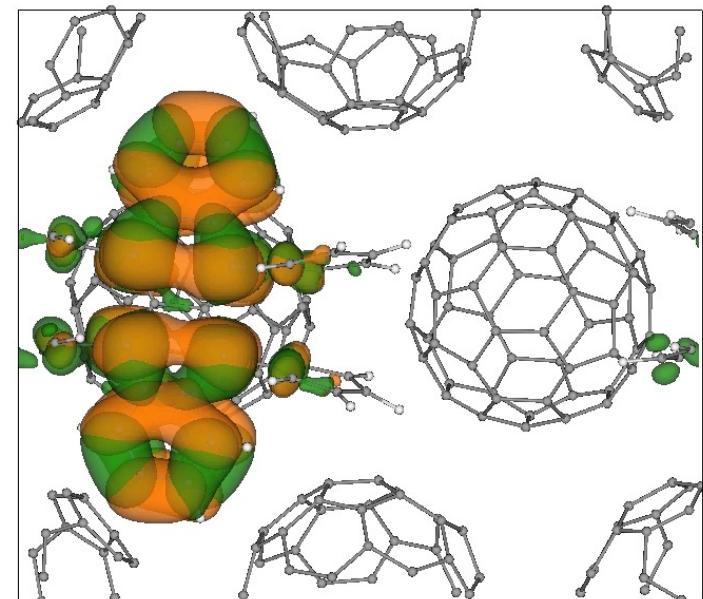
# Nonadiabatic Quantum Molecular Dynamics



*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>



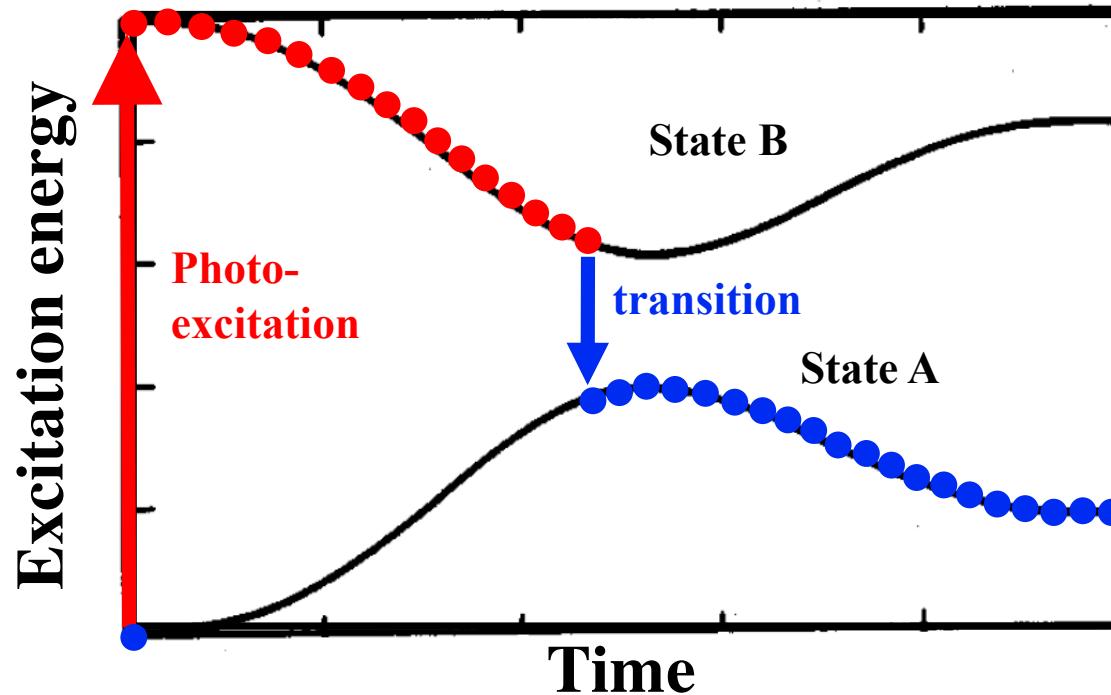
quasi-electron; quasi-hole

- **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

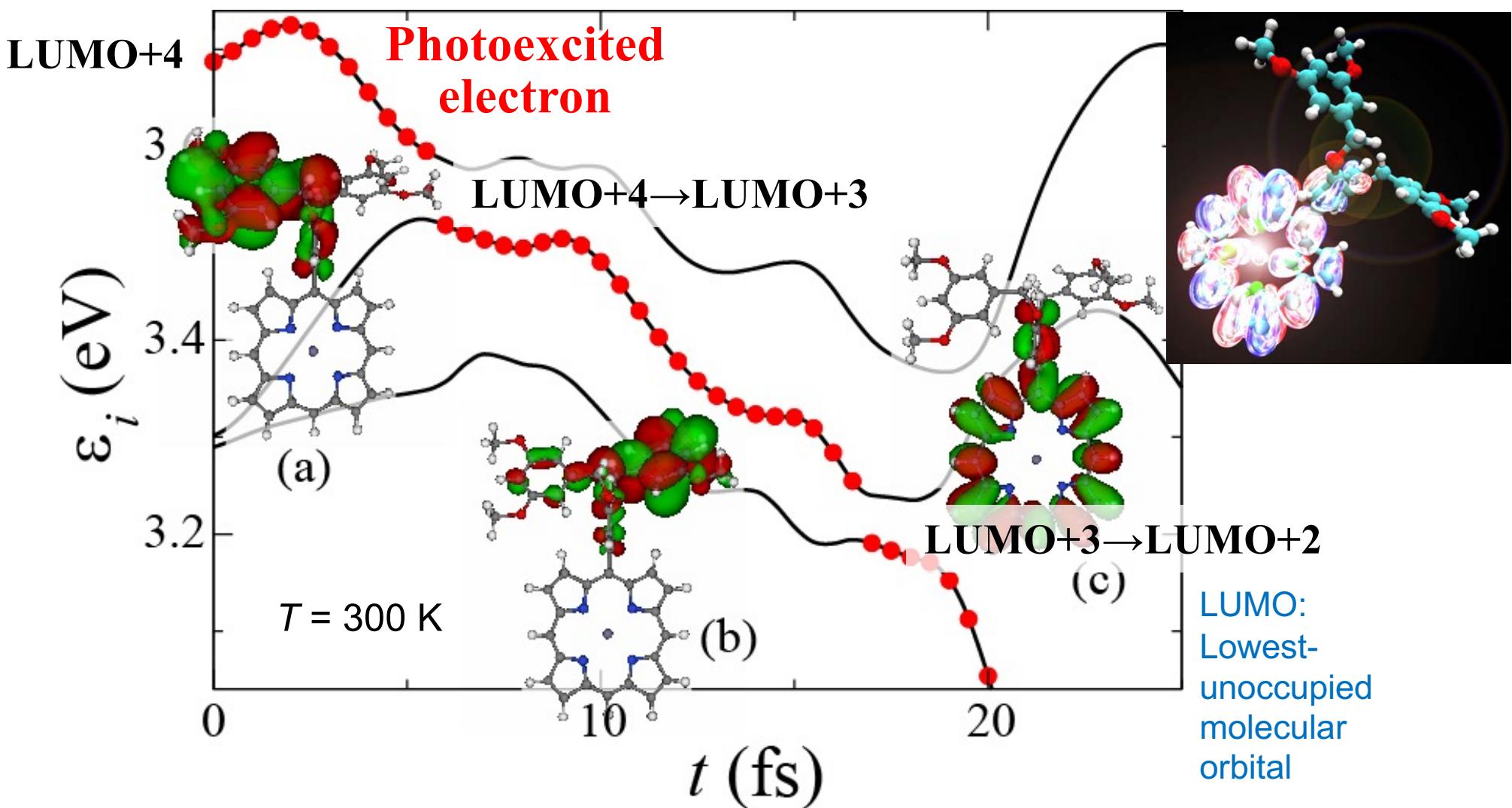
K-th excitation frequency

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

J-th adiabatic excited state

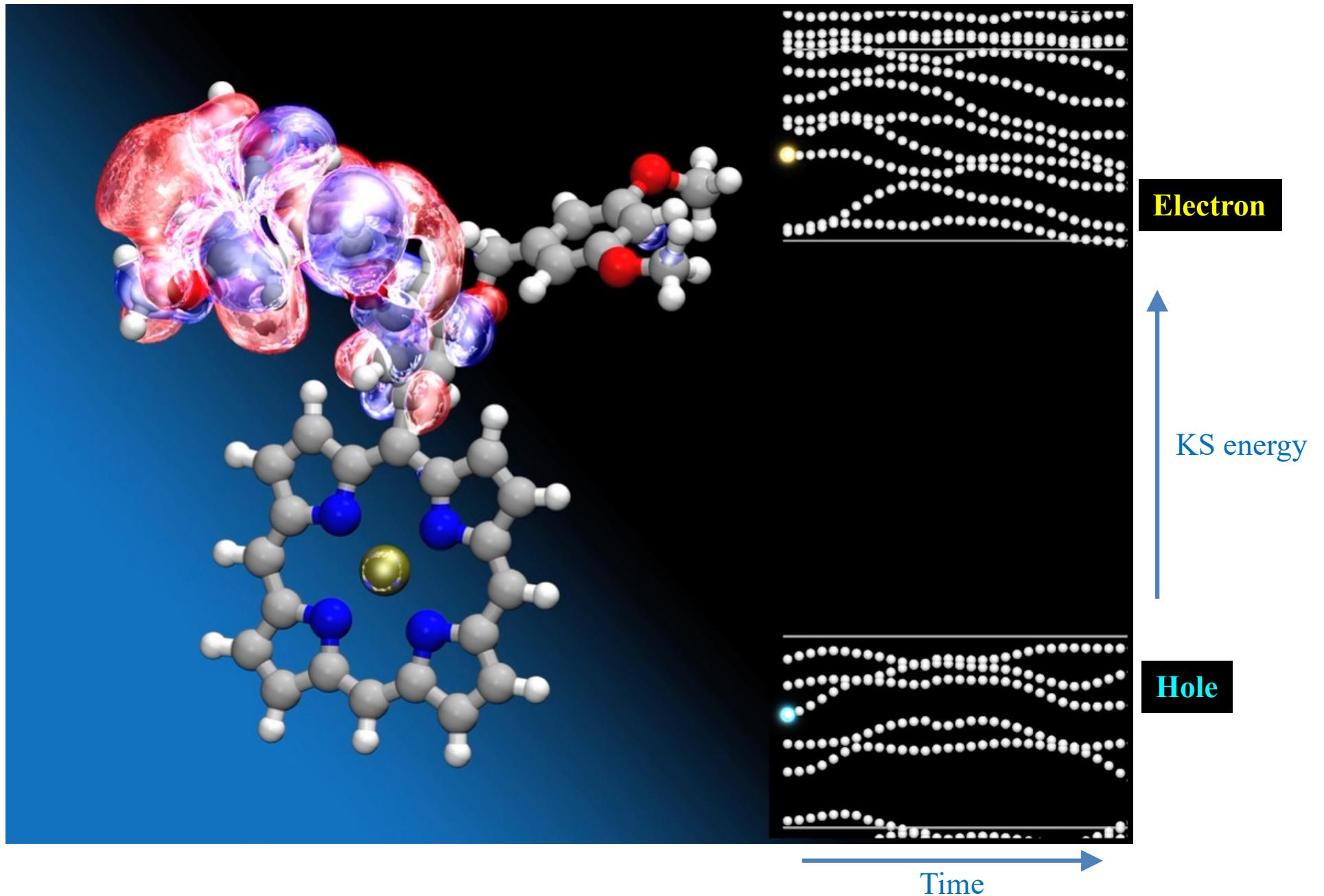
Electronic transition assisted by nuclei motion

# 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]**

$$\delta V(t) = \delta v_{kl\tau}(t) \hat{a}_{k\tau}^+ \hat{a}_{l\tau} \xrightarrow{\hspace{10em}} \delta P_{ij\sigma}(t) = \delta \langle \Phi(t) | \hat{a}_{i\sigma}^+ \hat{a}_{j\sigma} | \Phi(t) \rangle$$

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

electron      hole

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

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

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

***I*-th excitation energy**

## ***I-th excitation energy***

## Kohn-Sham energy

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

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

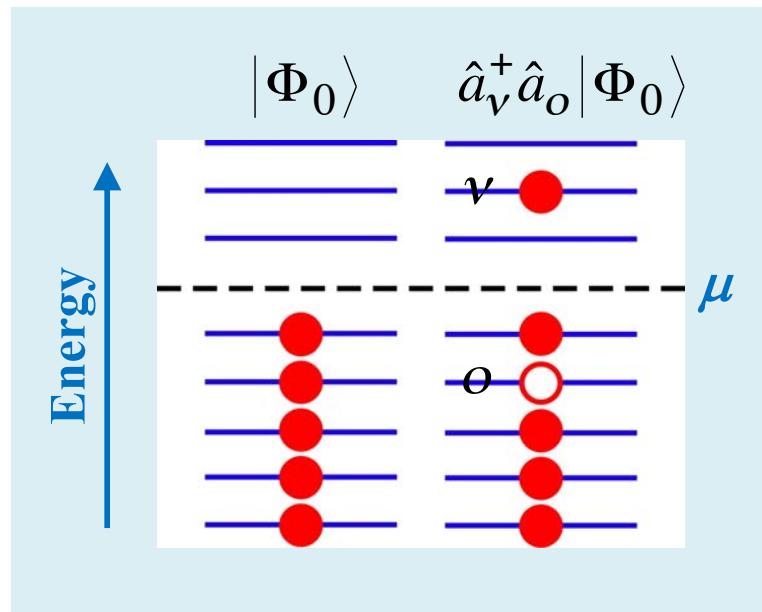
Coulomb & exchange-correlation interaction matrix elements

# 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://cybermagics.netlify.app/software>

# Current & Future Supercomputing

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



- Atomistic simulations on million cores (pre-exascale)

Innovative & Novel Computational Impact on Theory & Experiment

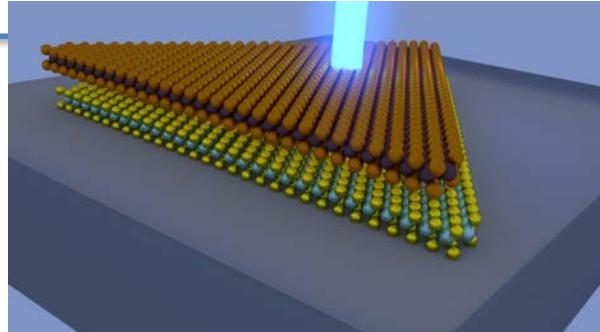
Title: AI-Guided Exascale Simulations of Quantum Materials Manufacturing and Control

PI and Co-PIs: Aiichiro Nakano—PI, Rajiv K. Kalia, Ken-ichi Nomura, Priya Vasishta



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

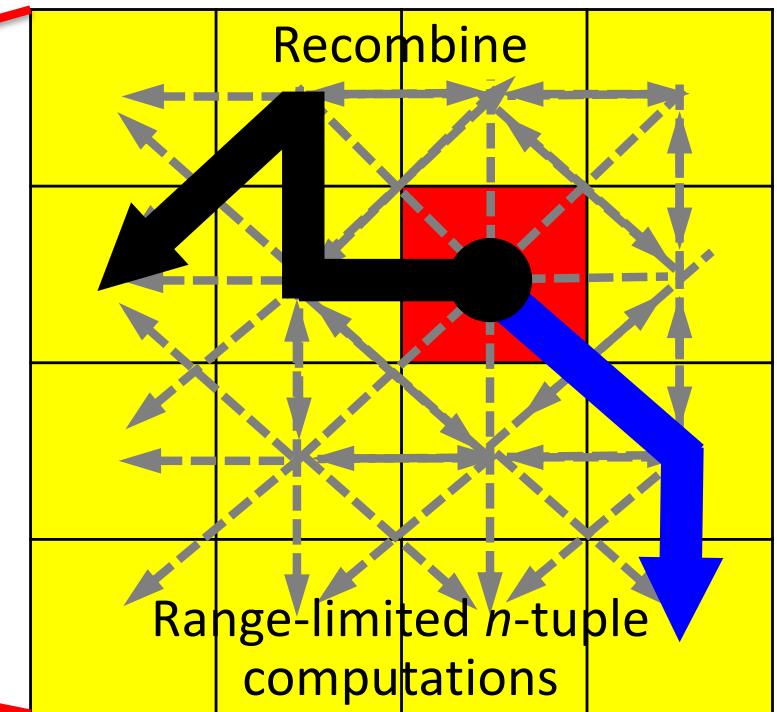
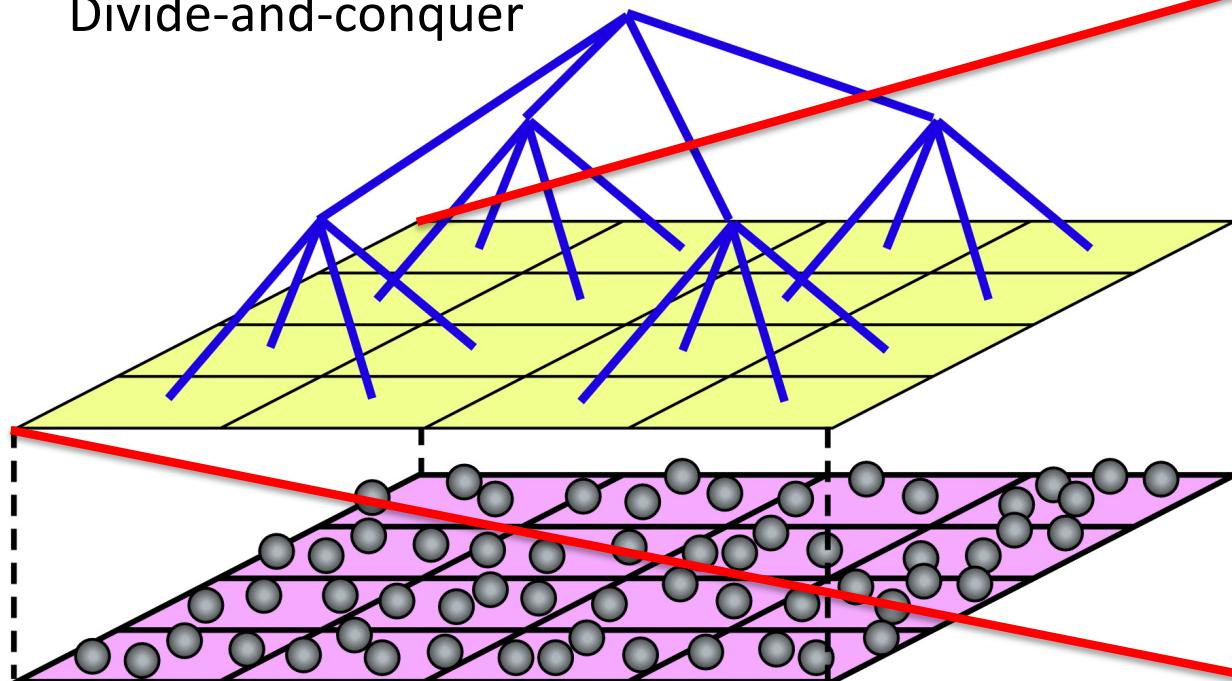
Michael Feldman | November 15, 2017 04:34 CET

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

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

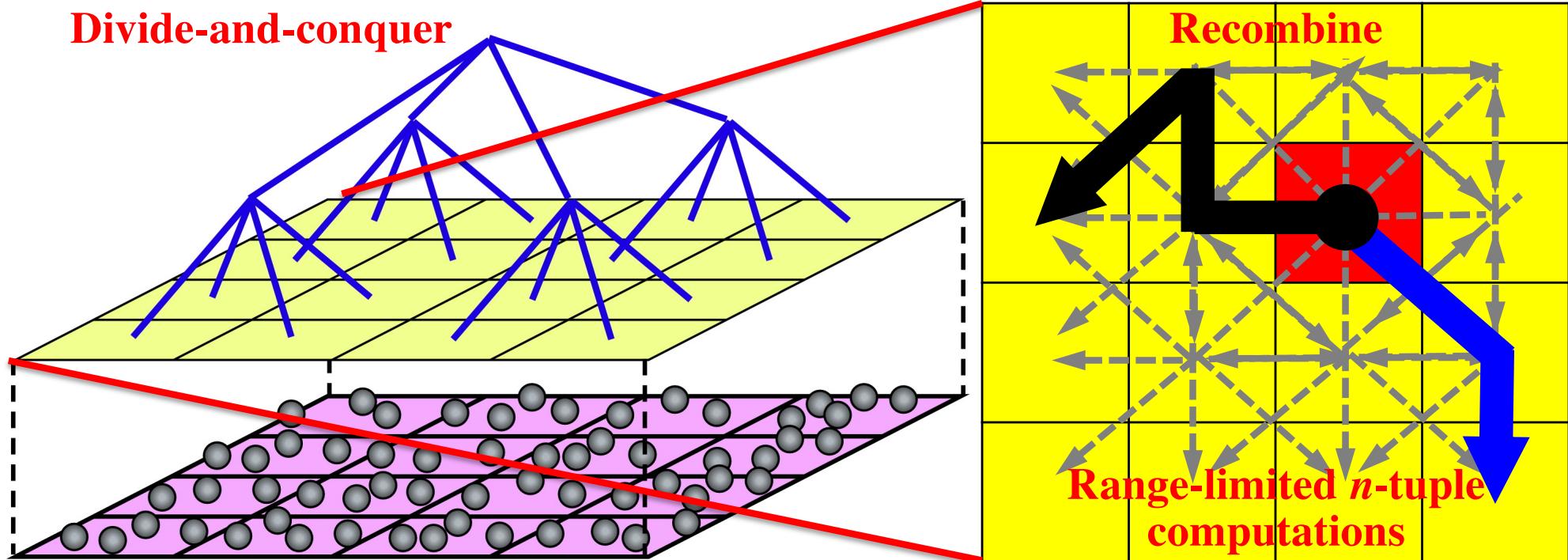
F. Shimojo et al., *J. Chem. Phys.* **140**, 18A529 ('14);  
K. Nomura et al., *ACM/IEEE SC14* ('14)

Divide-and-conquer



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

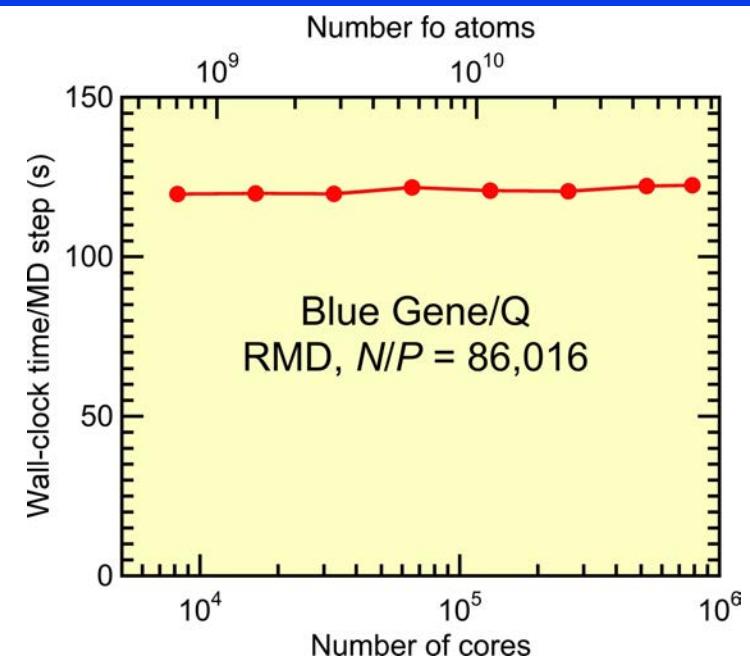
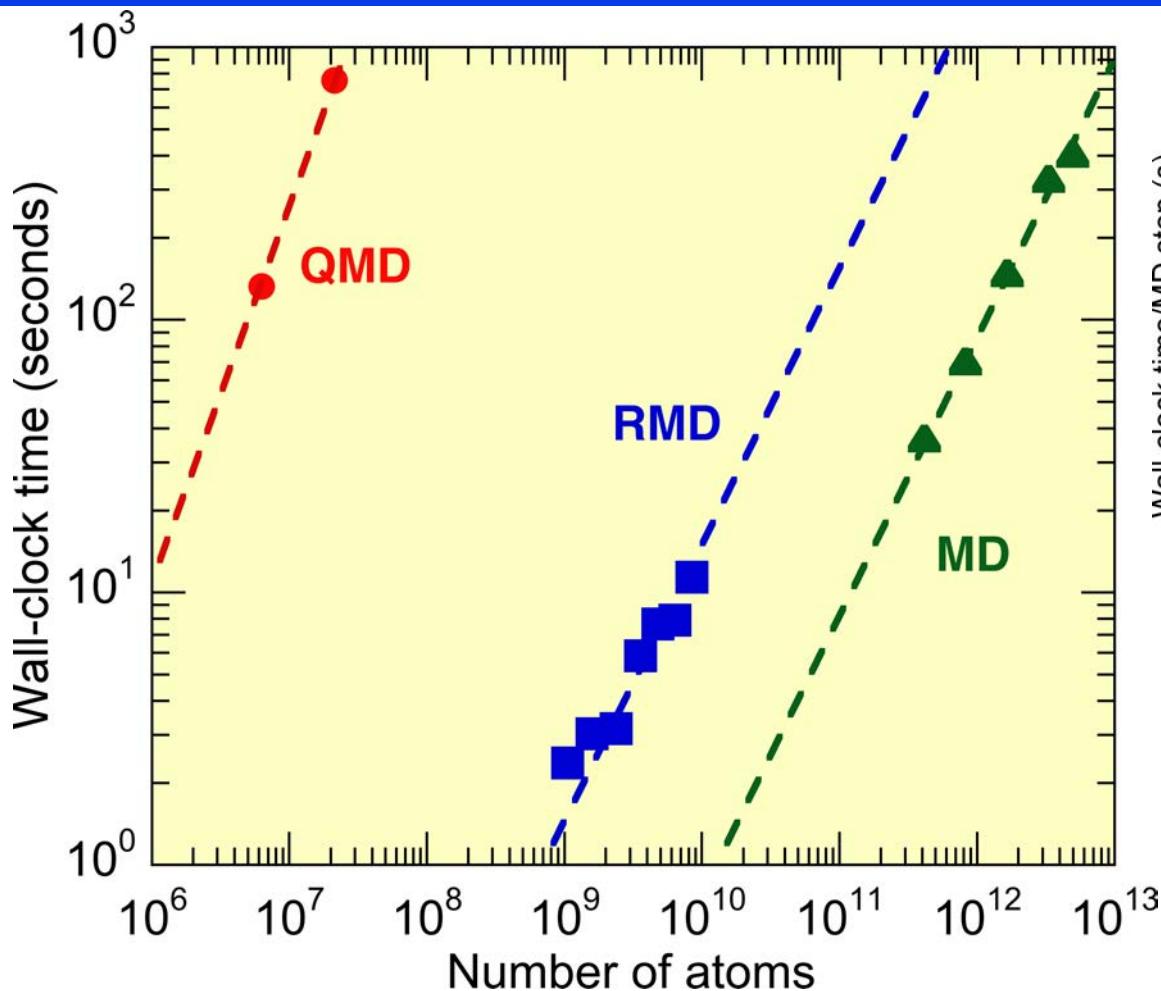
# Divide-Conquer-Recombine (DCR) Engines



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



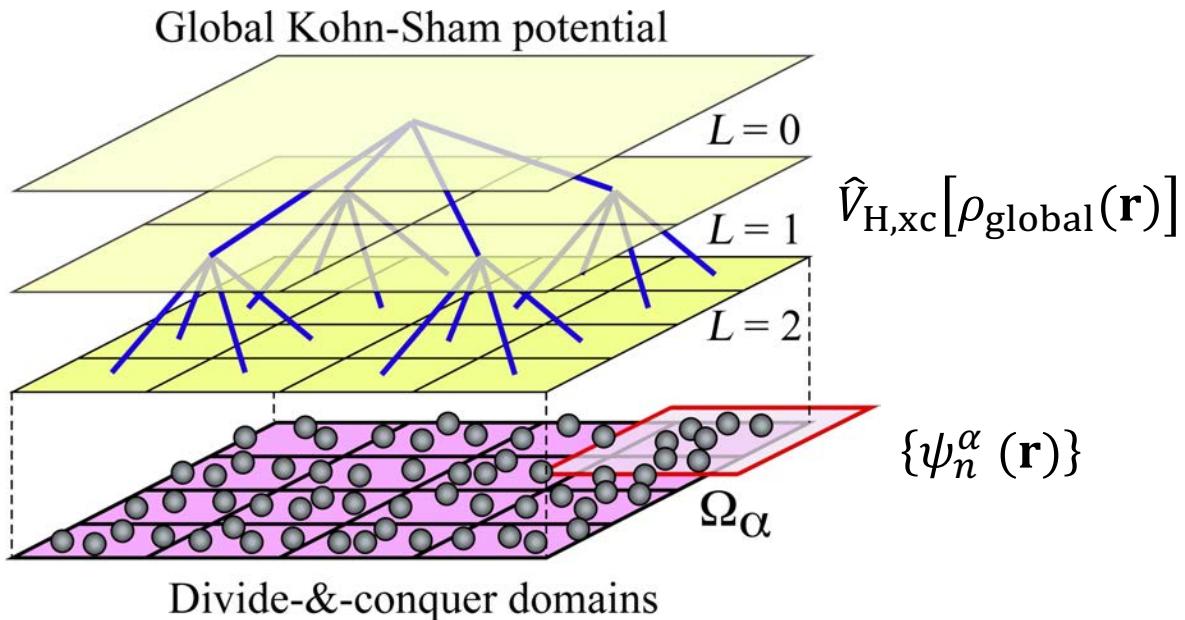
**QMD (quantum molecular dynamics): DC-DFT**

**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

$$\rho_{\text{global}}(\mathbf{r}) = \sum_\alpha p_\alpha(\mathbf{r}) \rho_\alpha(\mathbf{r})$$

Domain support function

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

Global chemical potential

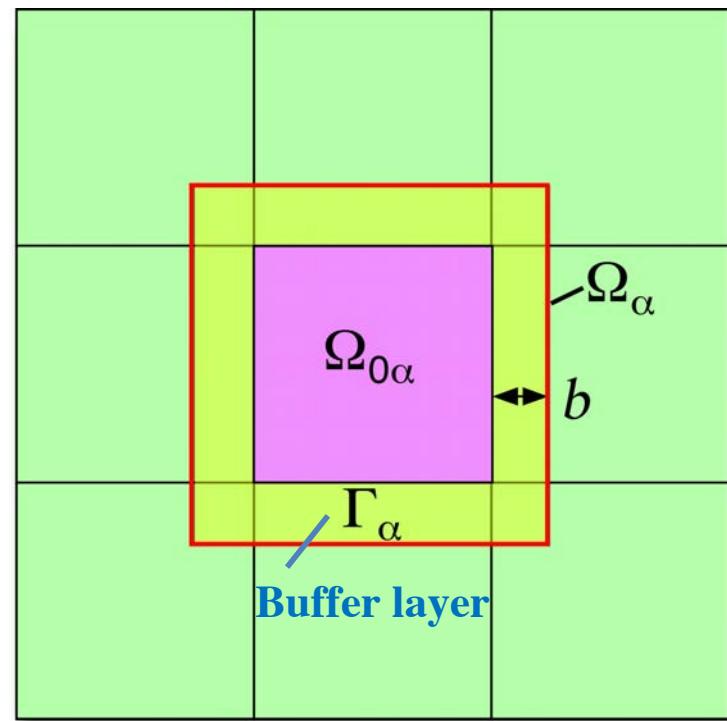
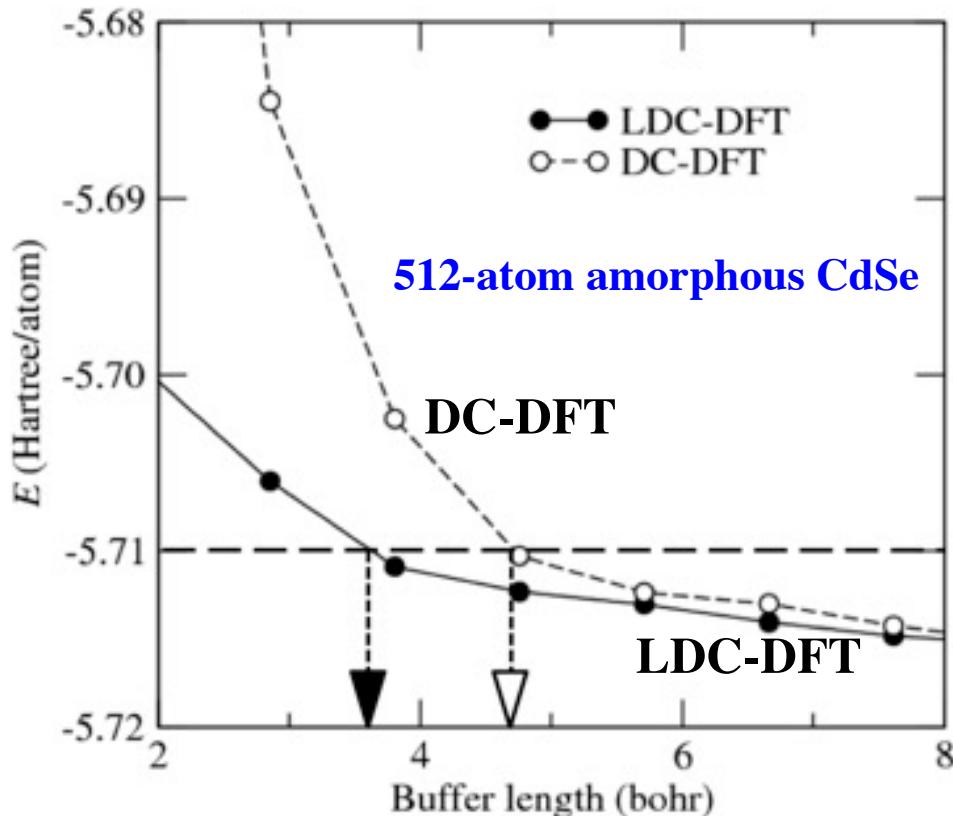
$$N = \int d\mathbf{r} \rho_{\text{global}}(\mathbf{r})$$

# Lean Divide-&-Conquer (LDC) DFT

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

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

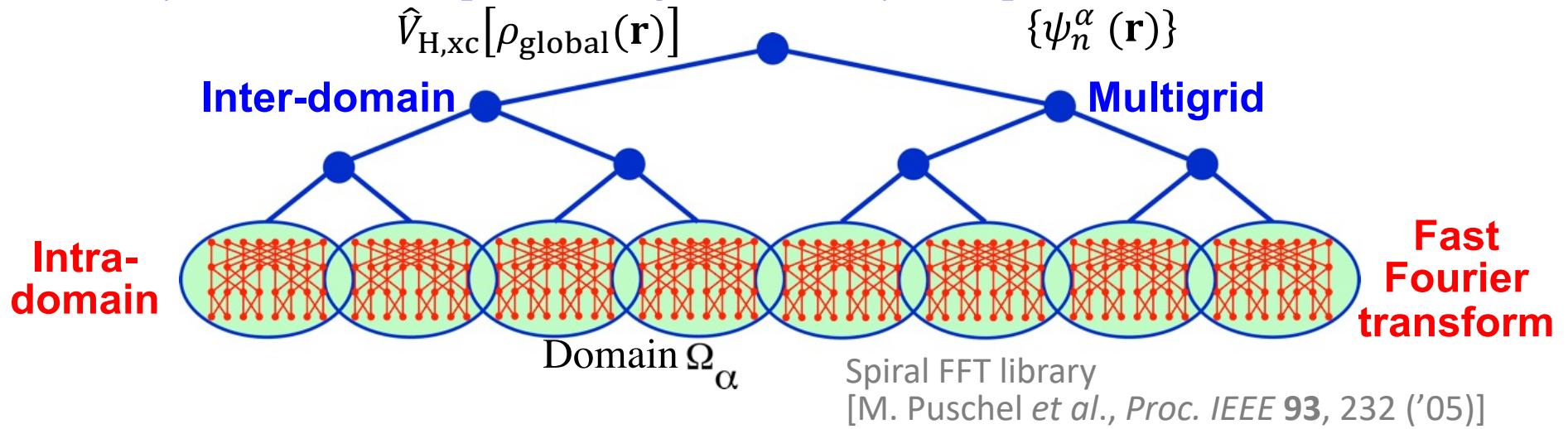
- More rapid energy convergence of LDC-DFT compared with nonadaptive DC-DFT



- 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:  $n^{\nu}$ )

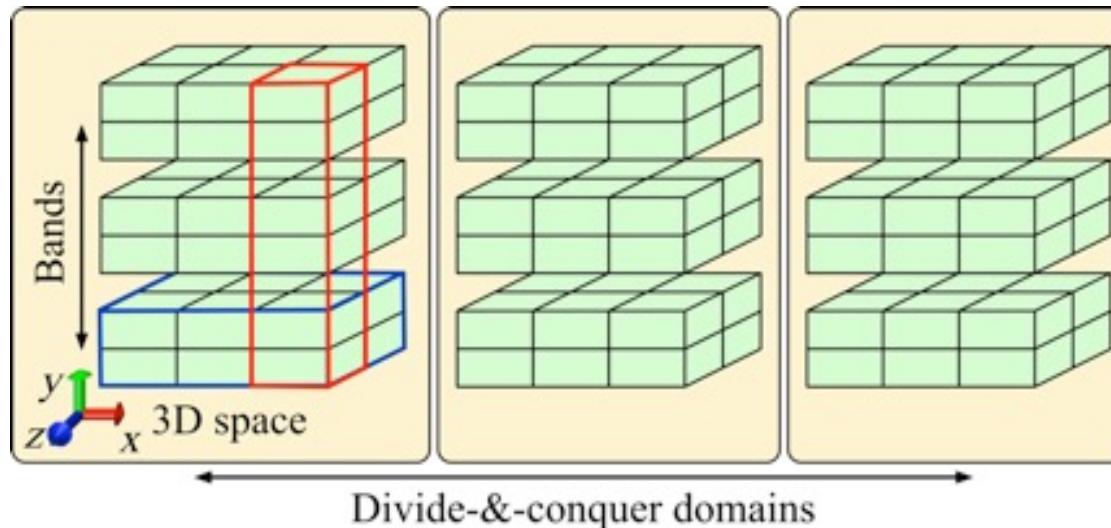
# Hierarchical Computing

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



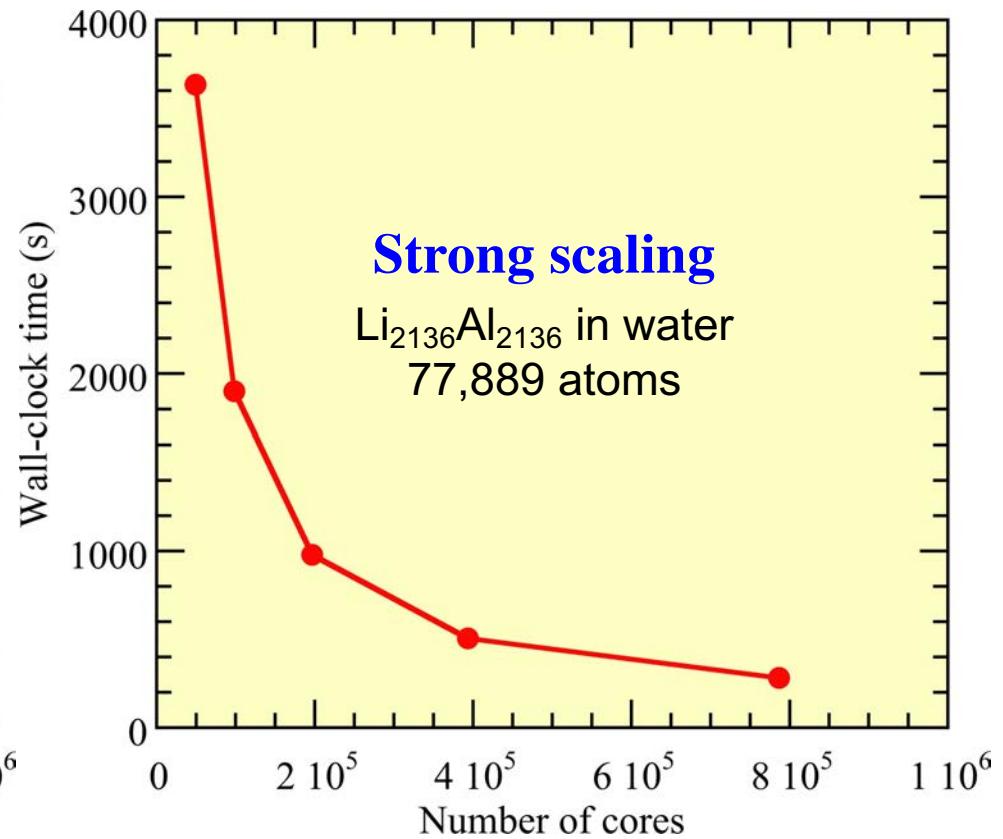
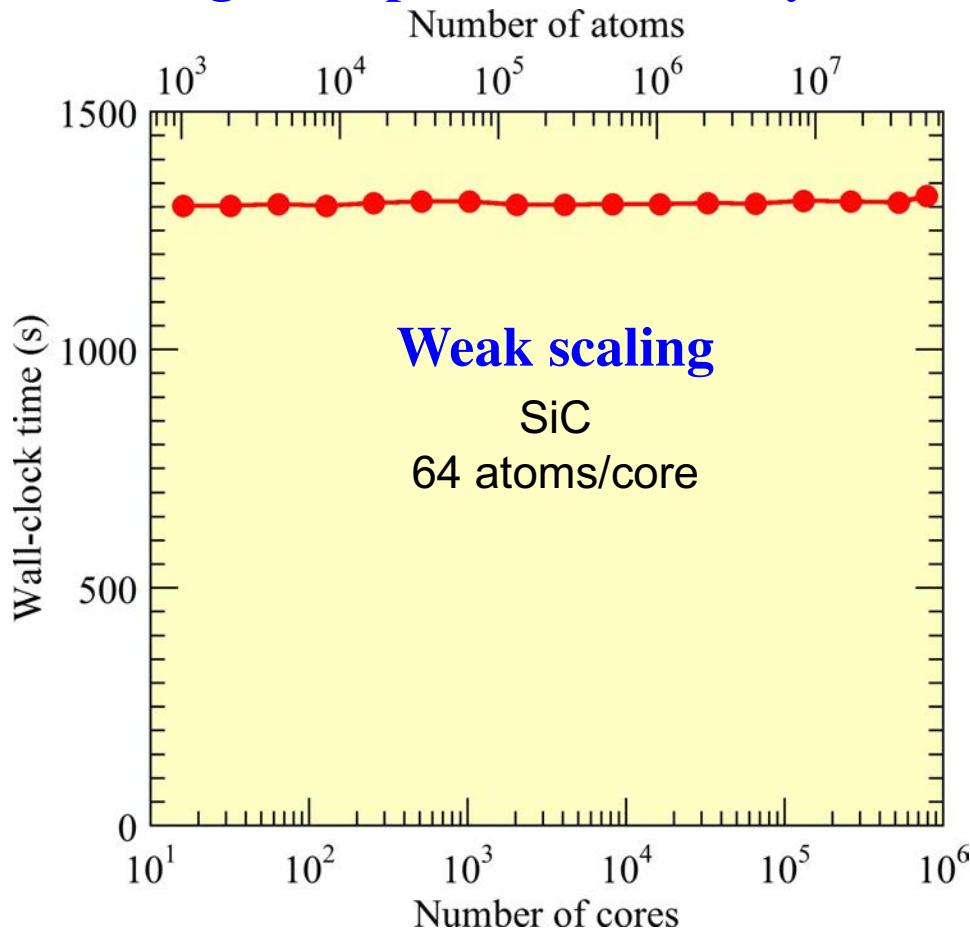
cf. globally- sparse-yet-locally-dense eigensolver [J. H. Lam *et al.*, *Nature Commun.* **15**, 3479 ('24)]

- 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)

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



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

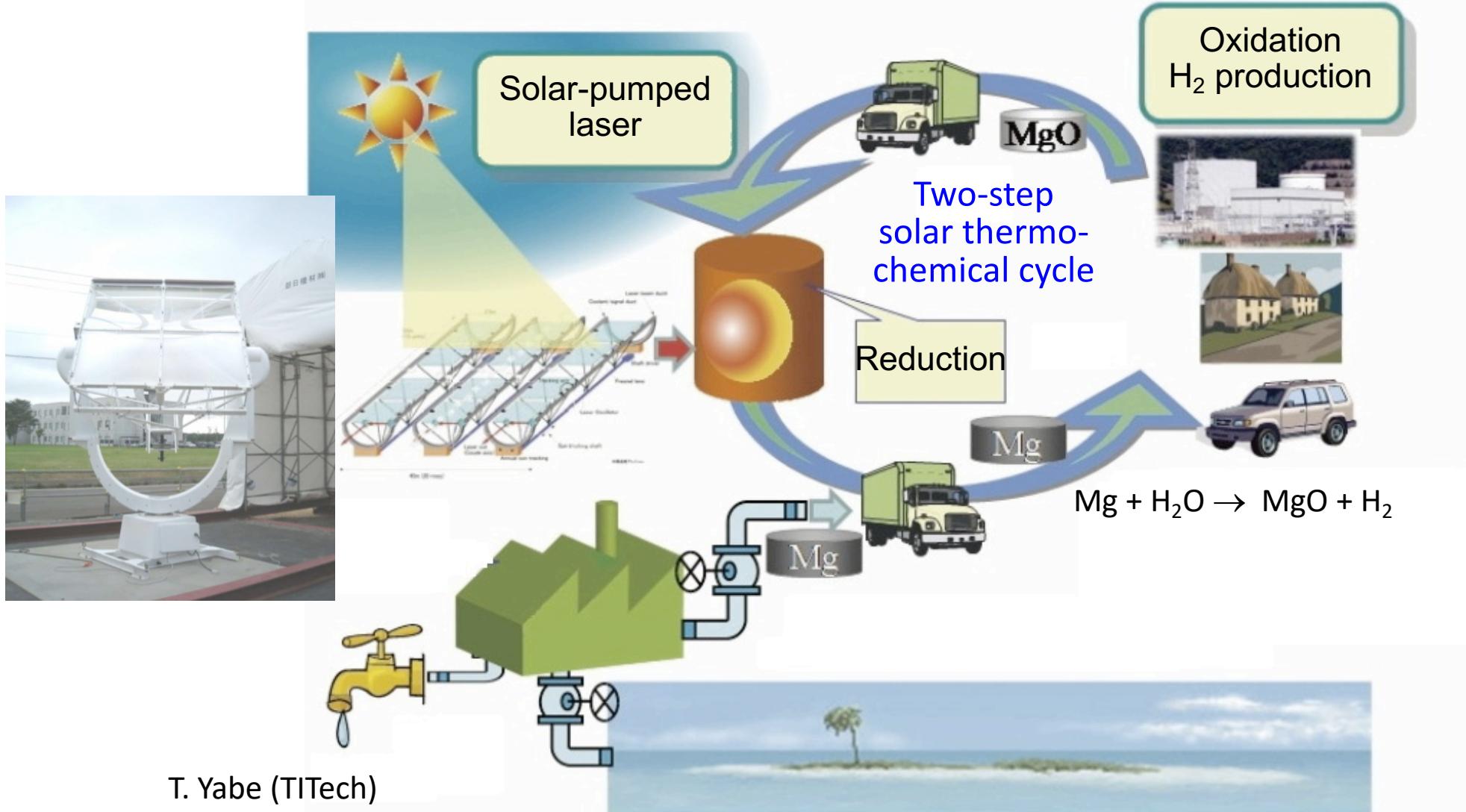
$$\hat{v}_{\text{nl}}\Psi = \sum_{i,j}^L \tilde{\mathbf{B}}(i)\tilde{\mathbf{D}}(i,j)\tilde{\mathbf{B}}(j)^T$$

- **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)

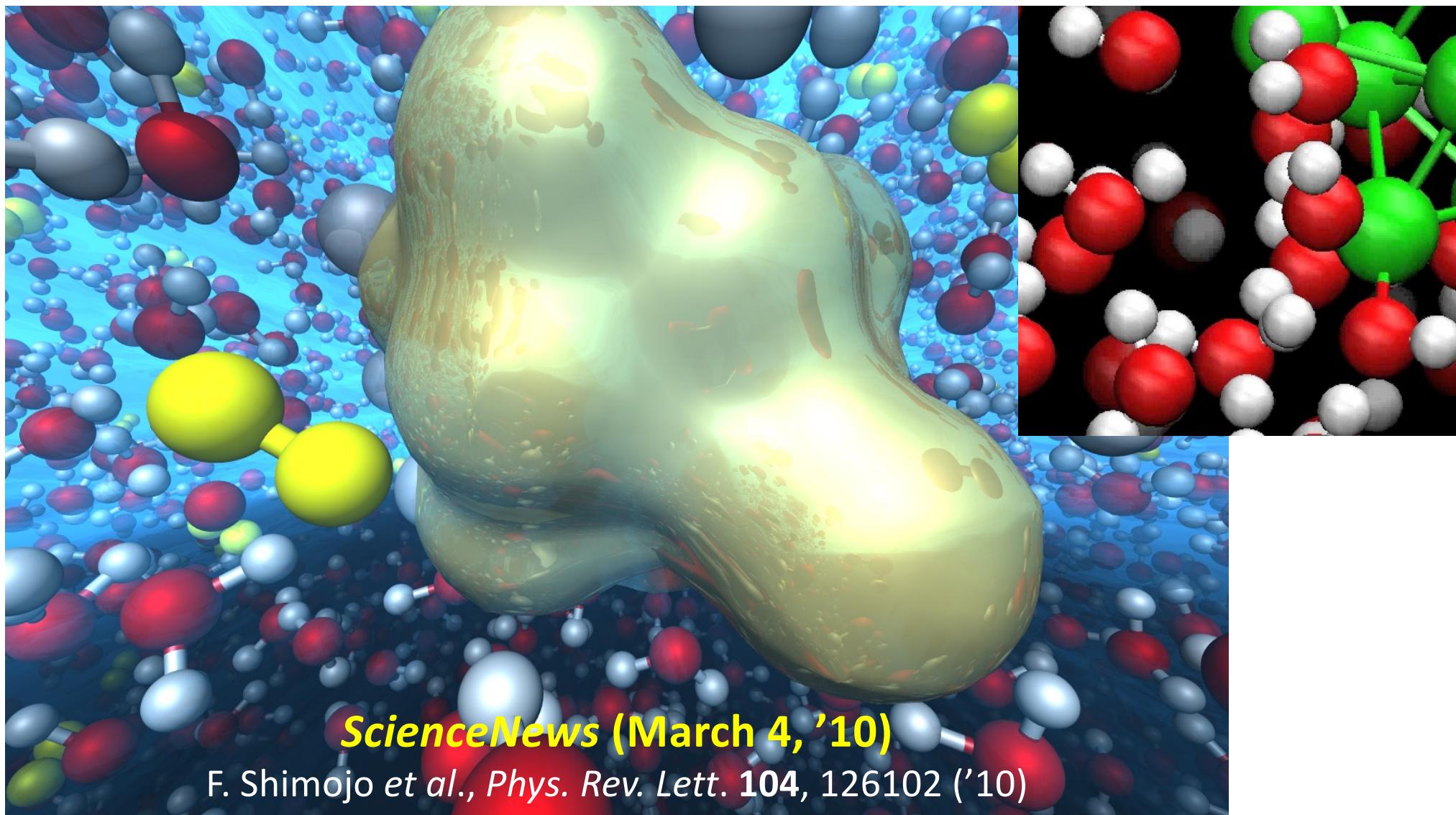
S. C. Tiwari *et al.*, *HPC Asia ('20)* best paper

# Renewal Energy Cycle by Metal Carriers



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

# Nanotechnology Solution



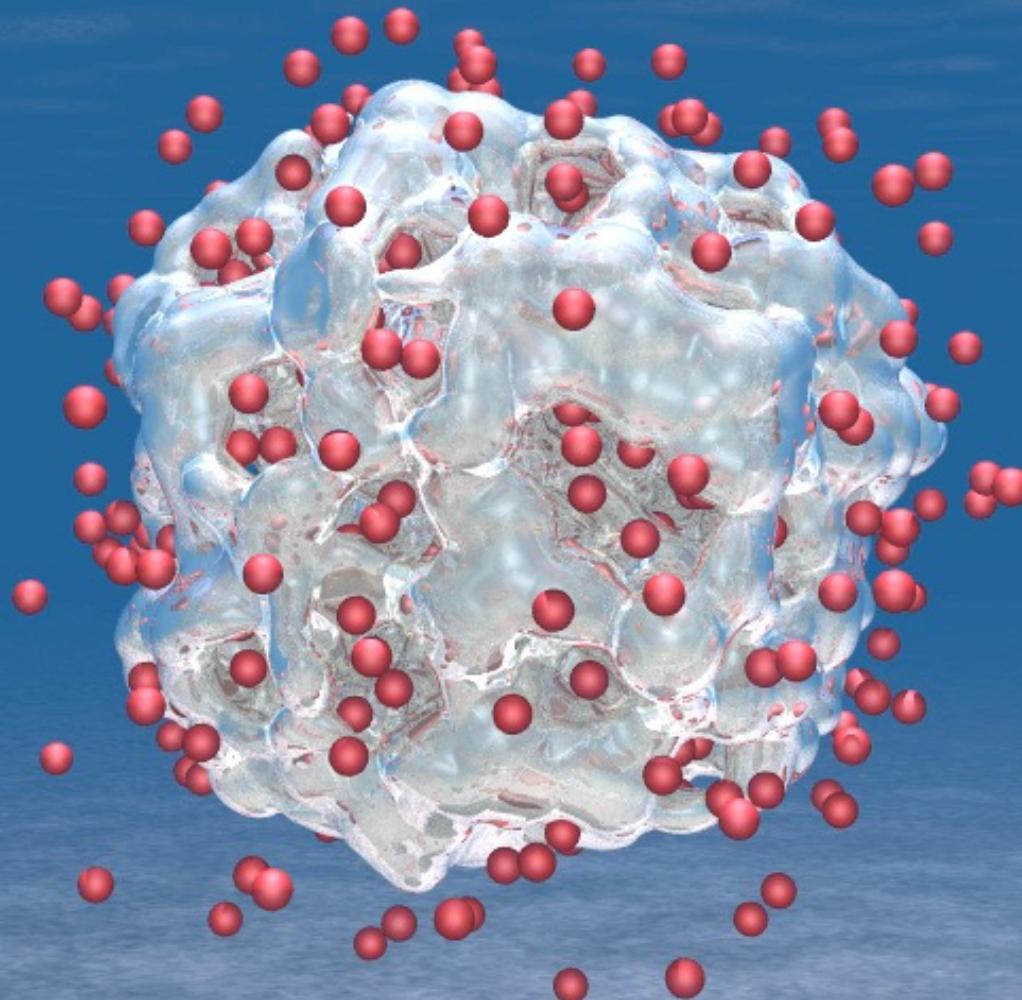
- QMD simulation shows rapid H<sub>2</sub> production from water by a superatom\* (Al<sub>17</sub>), but the technology is not scalable to larger particle sizes

\*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

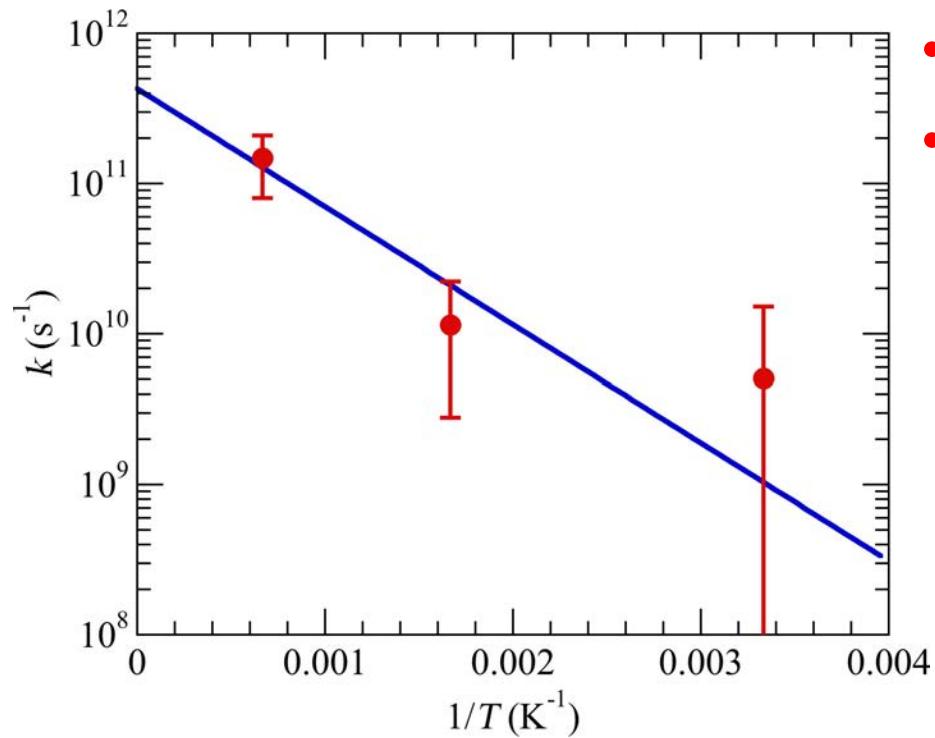
K. Shimamura *et al.*,  
*Nano Lett.* **14**, 4090 ('14)



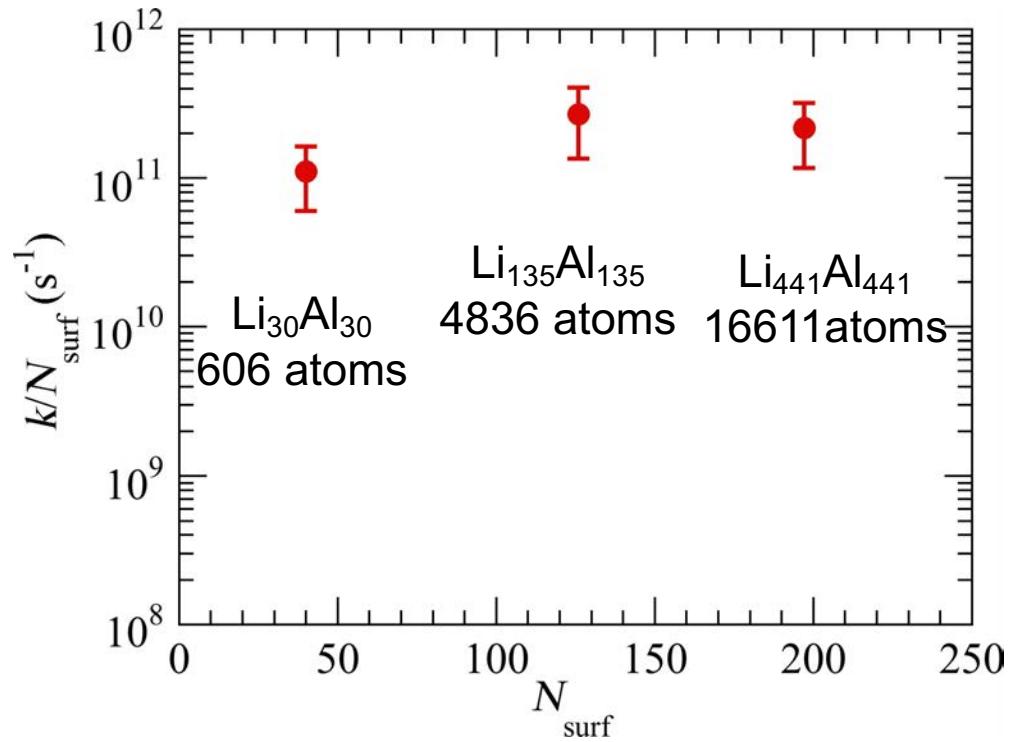
21,140 time steps (129,208 self-consistent-field iterations)

# 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

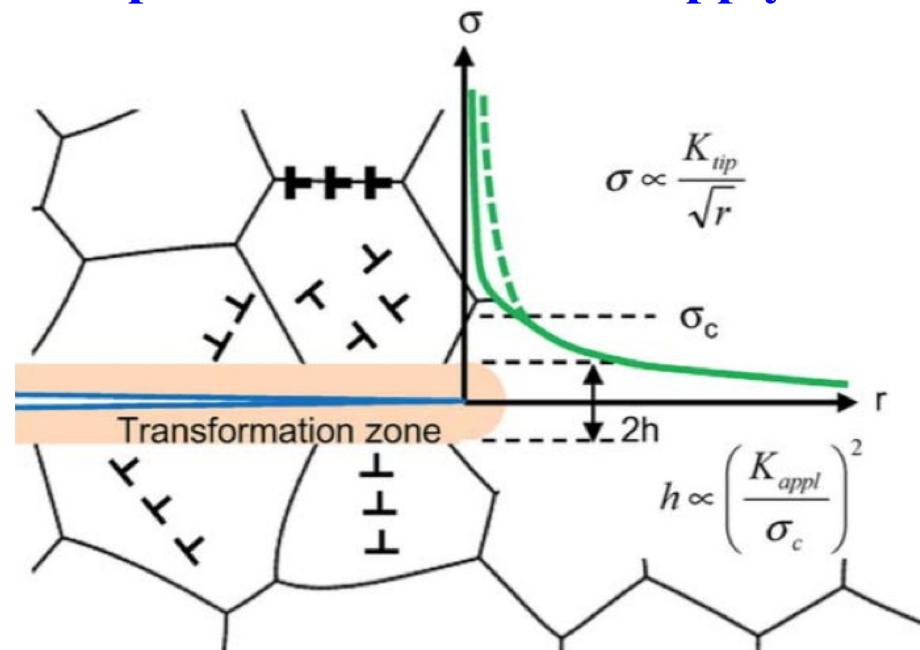
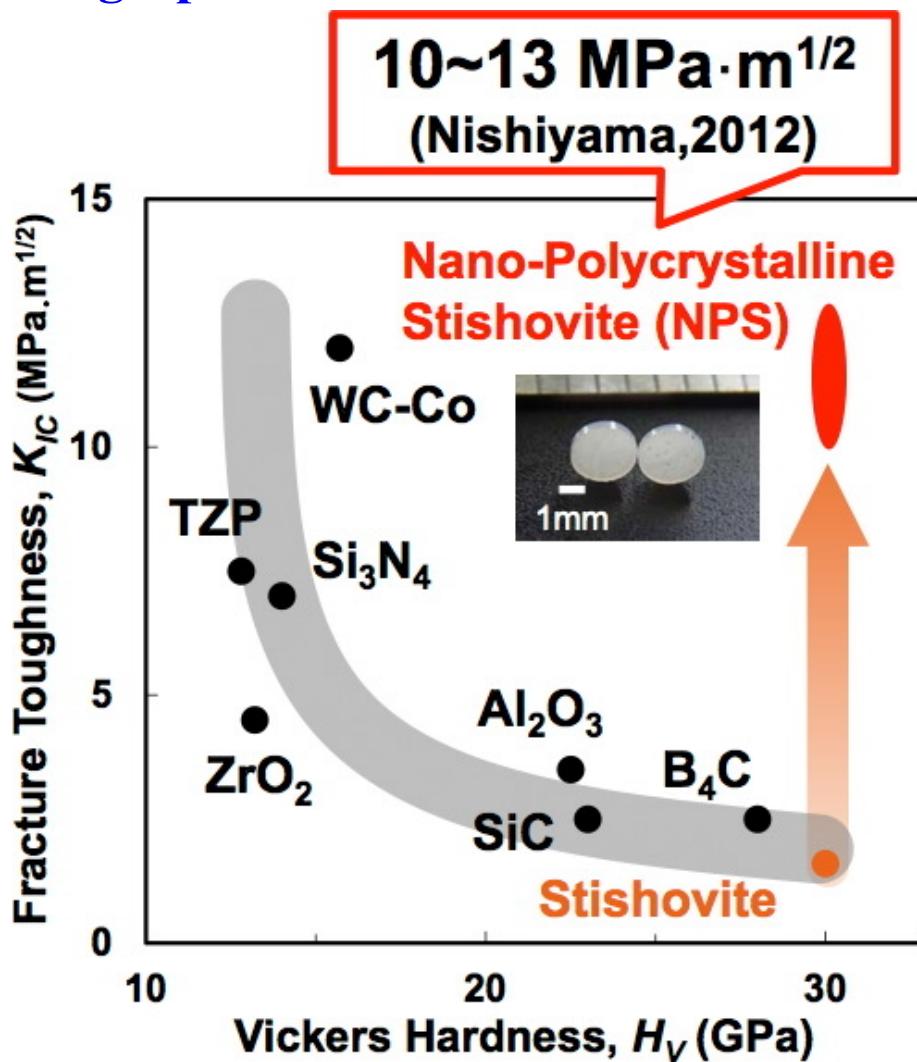


- Reaction rate does not decrease for larger particles → industrial scalability

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

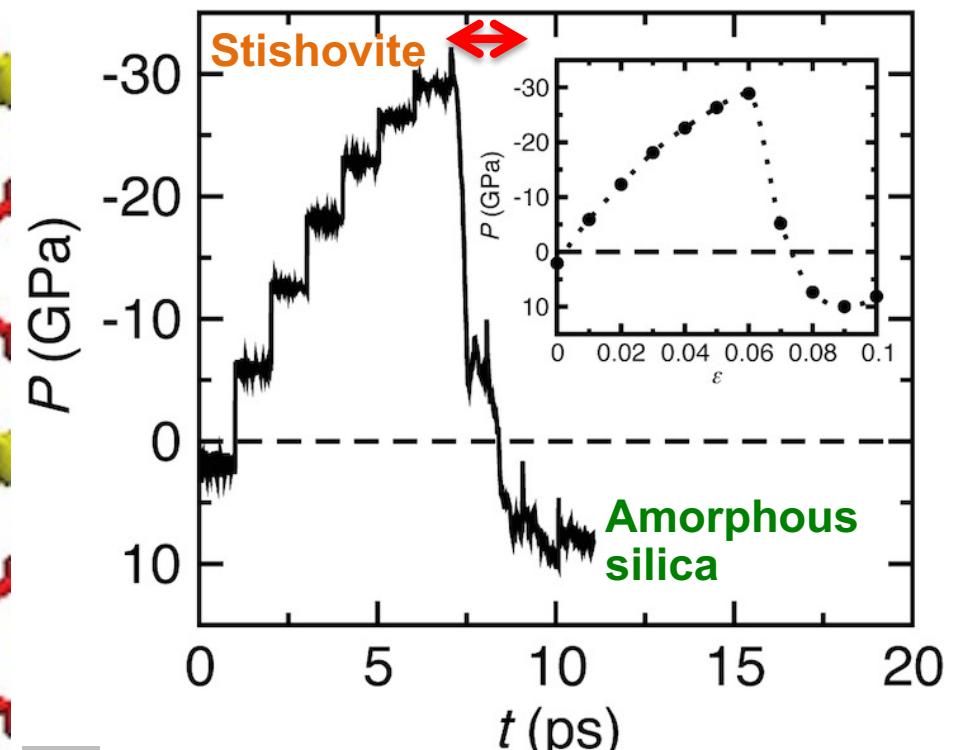
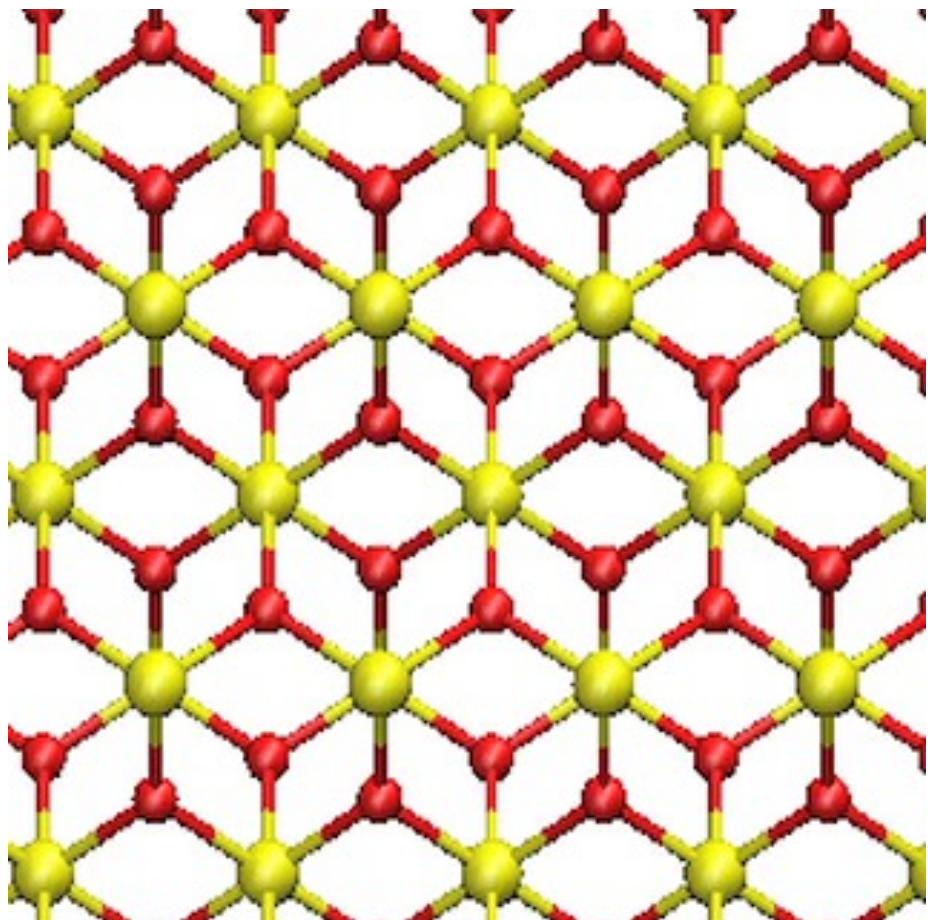


K. Yoshida *et al.*, *Sci. Rep.* **5**, 10993 ('15);  
*Acta Mater.* **124**, 316 ('17)

- 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

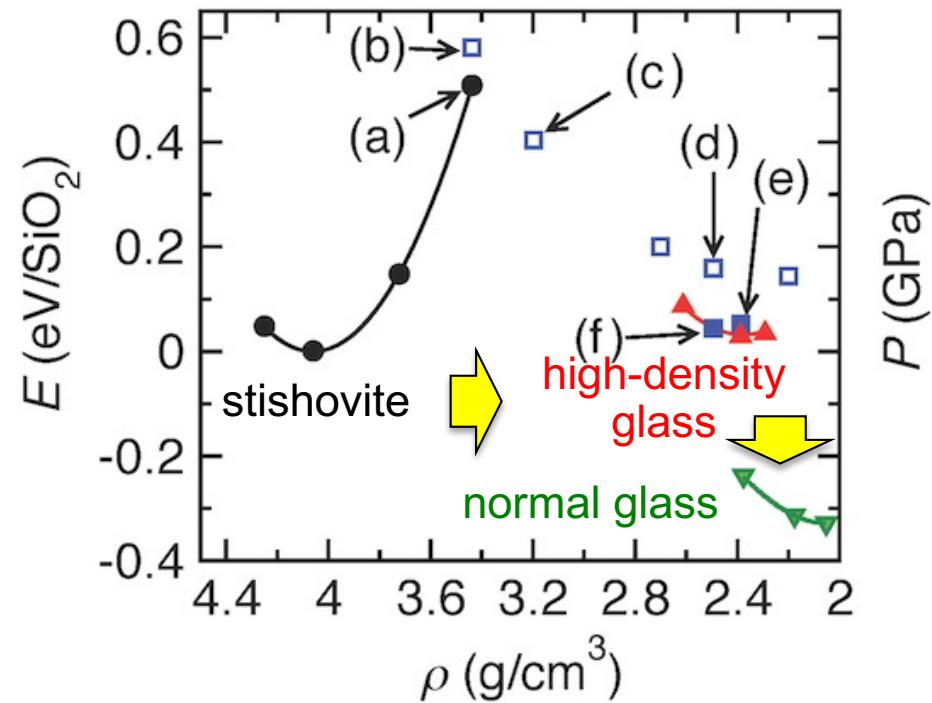
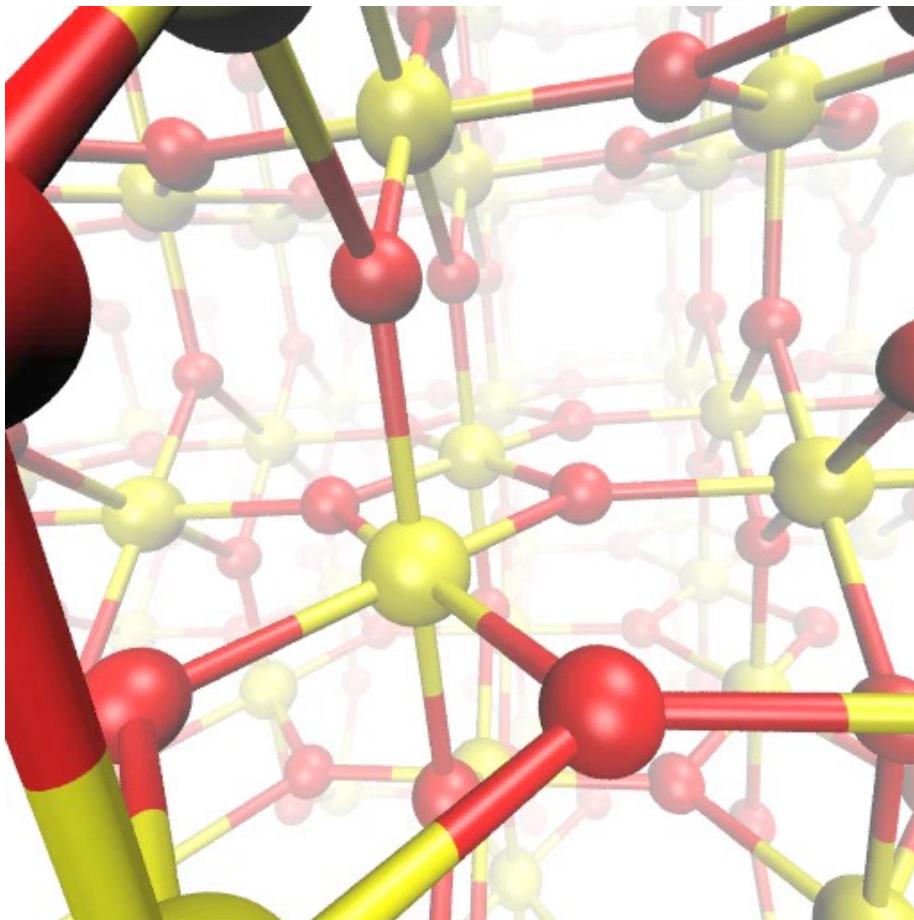


$$\text{Volume}_{\text{amorphous silica}} \sim 2 \times \text{Volume}_{\text{stishovite}}$$

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

# Rapid Amorphization Mechanism

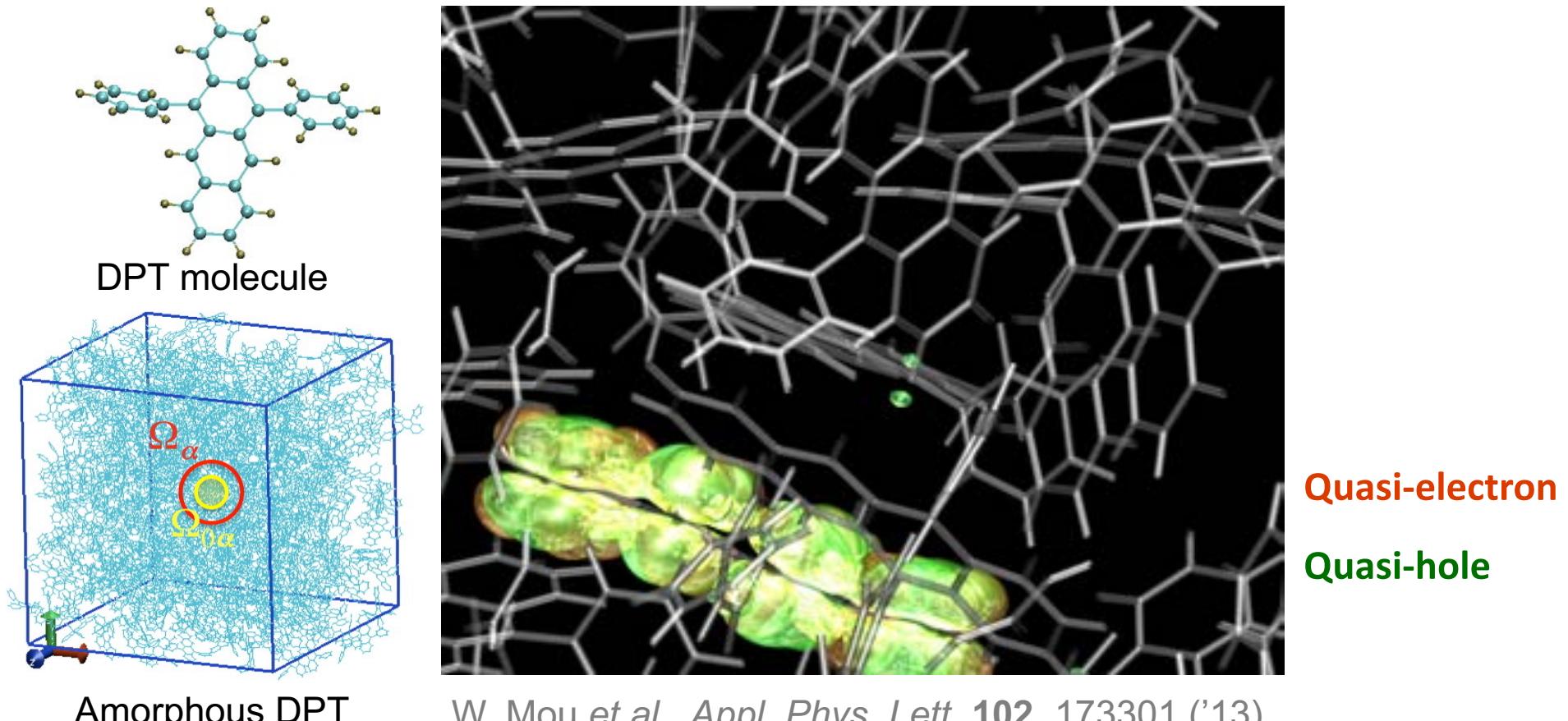
- Found a displacive amorphization mechanism that only involves short-distance collective motions of atoms, thereby facilitating the rapid transformation



- 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

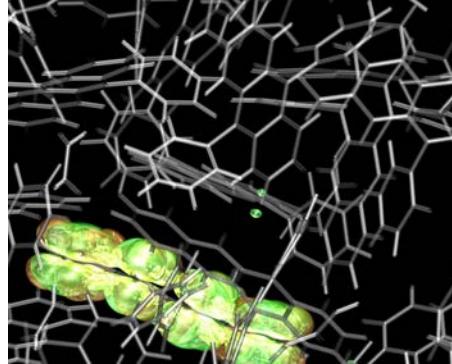
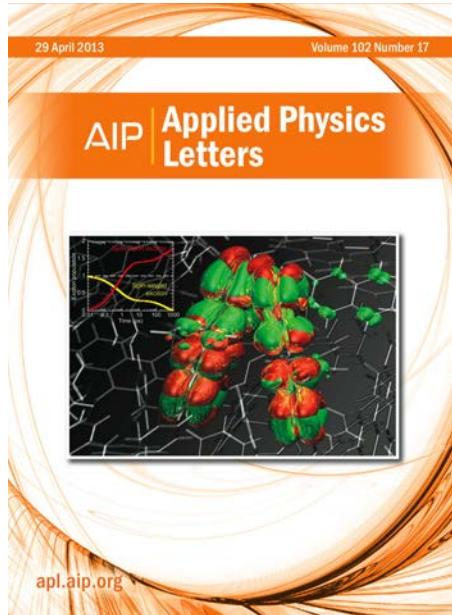
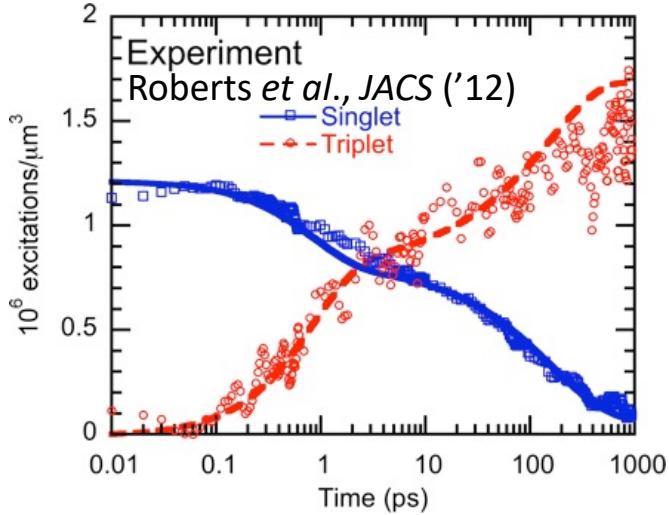
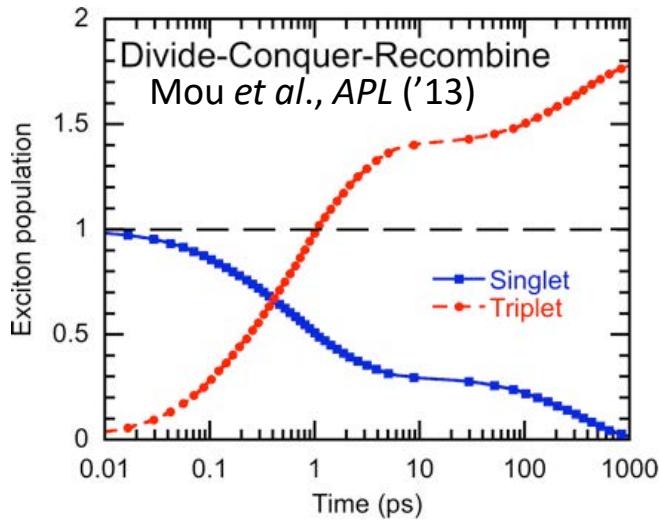
- Photo-current doubling by splitting a singlet exciton into 2 triplet excitons
- Singlet fission in mass-produced disordered organic solid → efficient low-cost solar cells
- Experimental breakthrough: SF found in amorphous diphenyl tetracene (DPT)



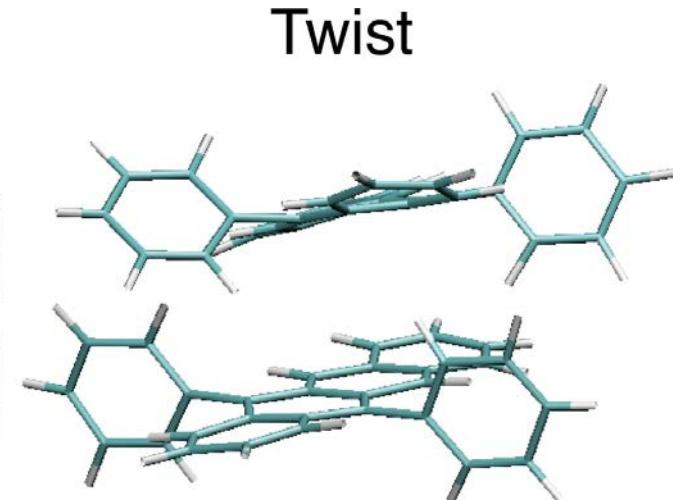
- Divide-conquer-recombine nonadiabatic QMD (phonon-assisted exciton dynamics) + time-dependent perturbation theory (singlet-fission rate) + kinetic Monte Carlo calculations of exciton population dynamics in 6,400-atom amorphous DPT

# Singlet-Fission Hot Spot

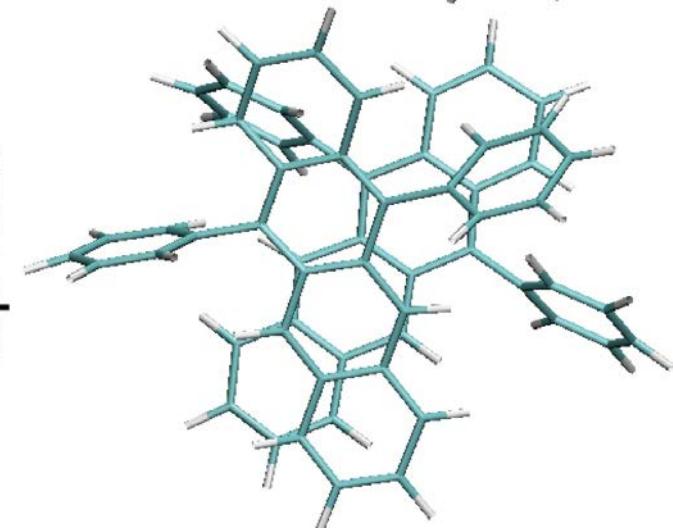
- Nonadiabatic quantum molecular dynamics simulations not only reproduced experimentally measured exciton population dynamics but also revealed unknown molecular geometry of singlet fission hot spots



Side view



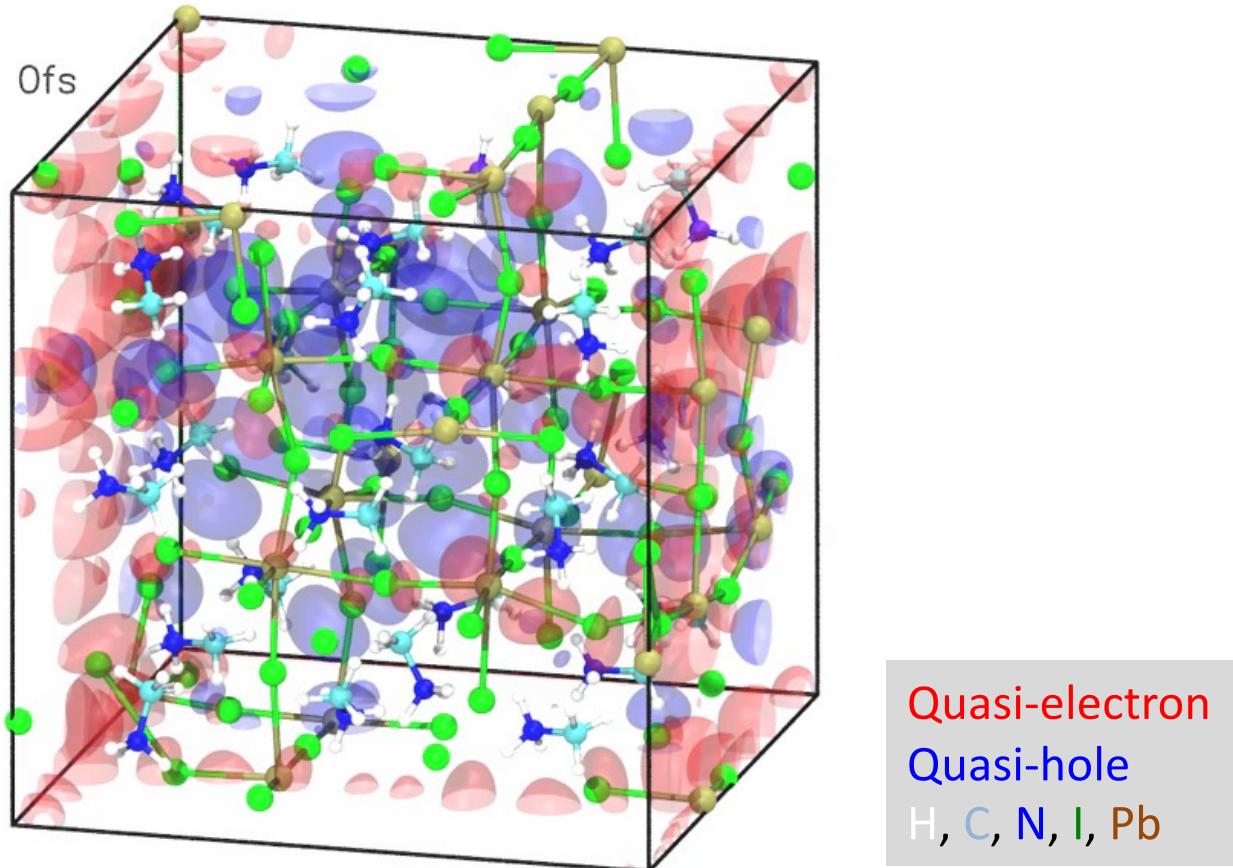
Top view



# 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)]

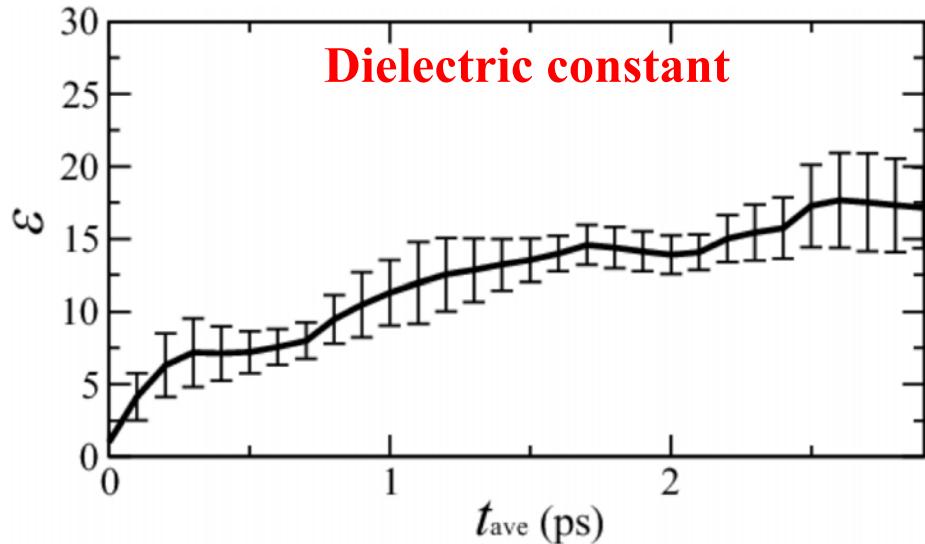


- Nonadiabatic QMD simulation

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

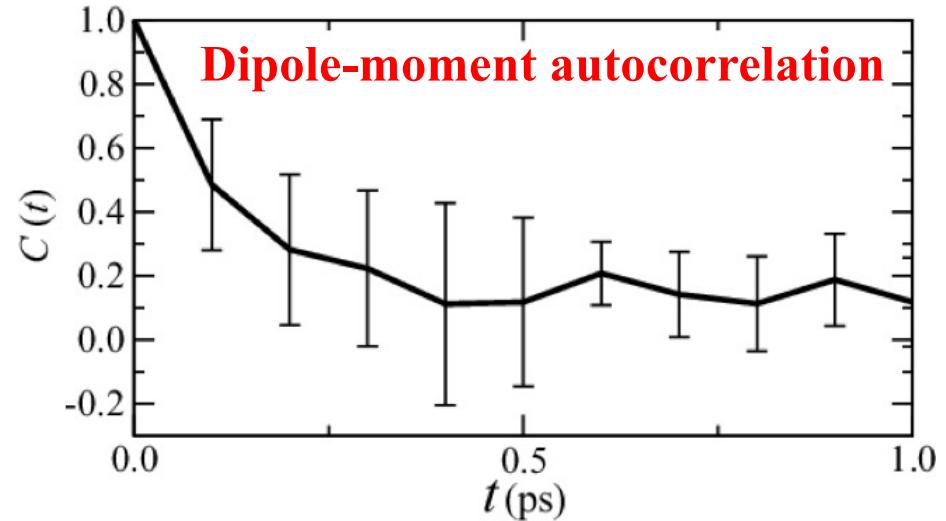


$$\epsilon = 1 + \frac{4\pi}{3k_BTV} (\langle \mathbf{M}^2 \rangle - \langle \mathbf{M} \rangle^2)$$

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)

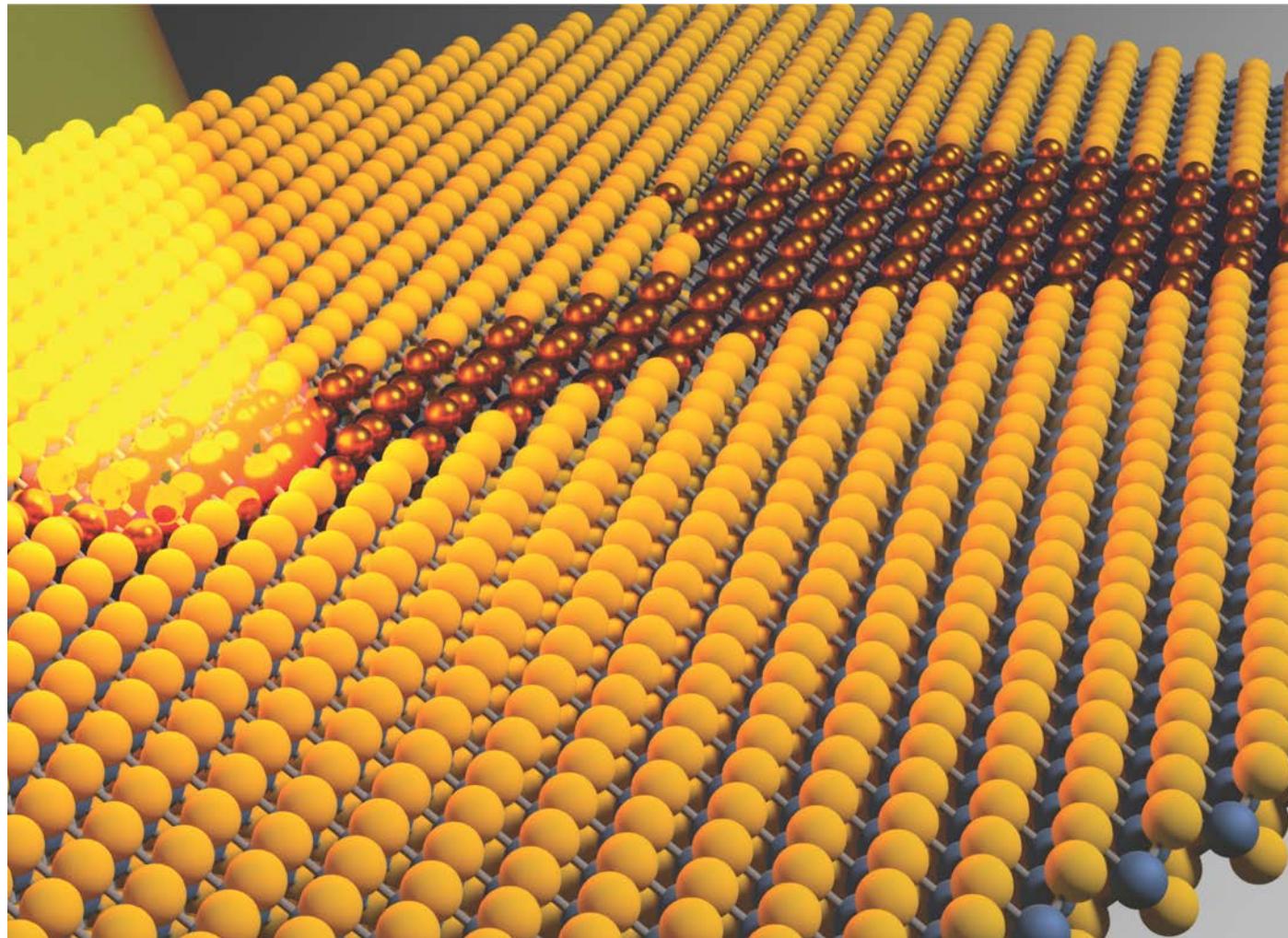
- Large dielectric constant of MA sublattice causes small exciton binding energy, **0.012 ± 0.009 eV (experimental upper bound = 0.05 eV [D'Innocenzo *et al.*, *Nat. Commun.* **5**, 3586 ('14)])**
- MA sublattice quickly screens out electrostatic electron-hole attraction to unbind an exciton & generate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

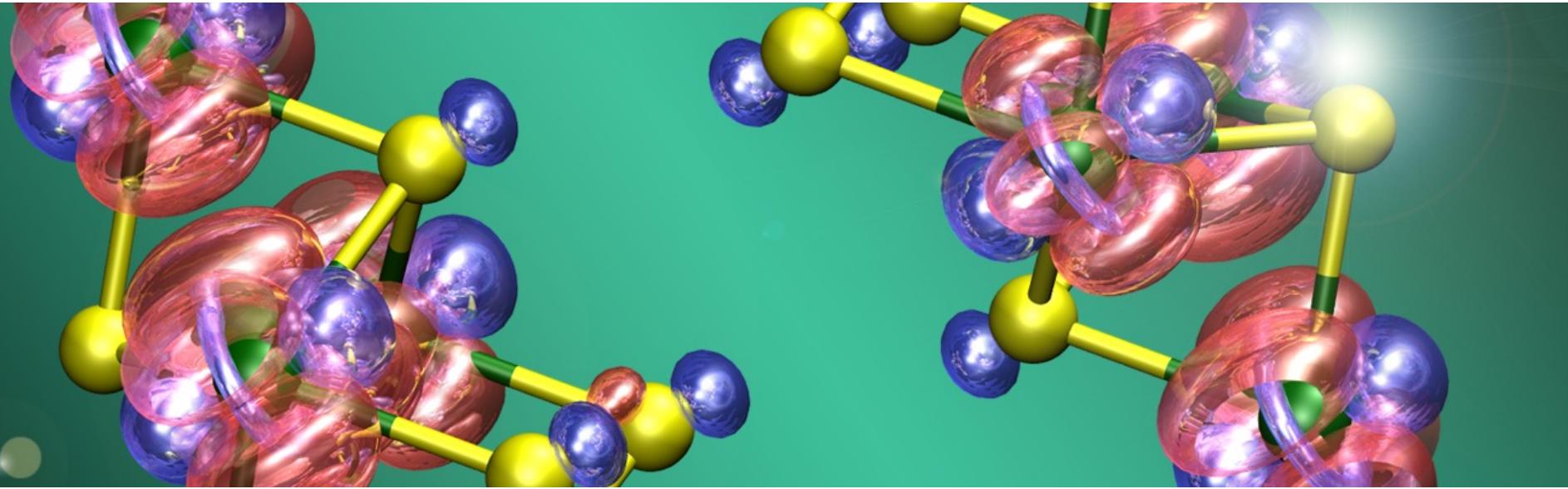
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**Goal:** Use ultrafast laser pulses to transform material structures & properties  
(e.g., semiconductor-to-metal) on demand

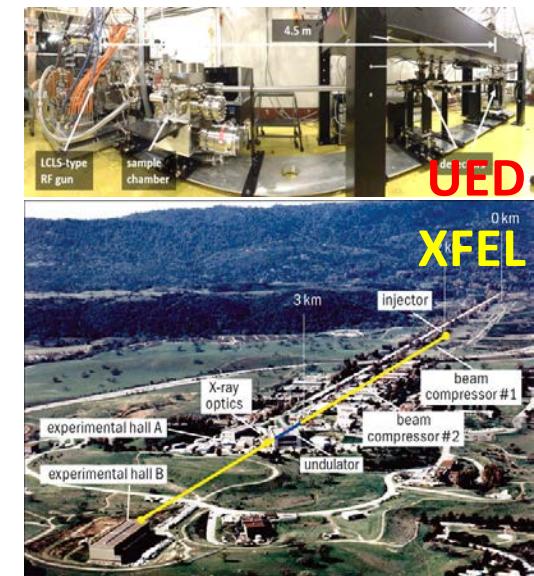


A. Krishnamoorthy *et al.*, *Nanoscale* **10**, 2742 ('18); journal cover

# Simulation-Experiment Synergy



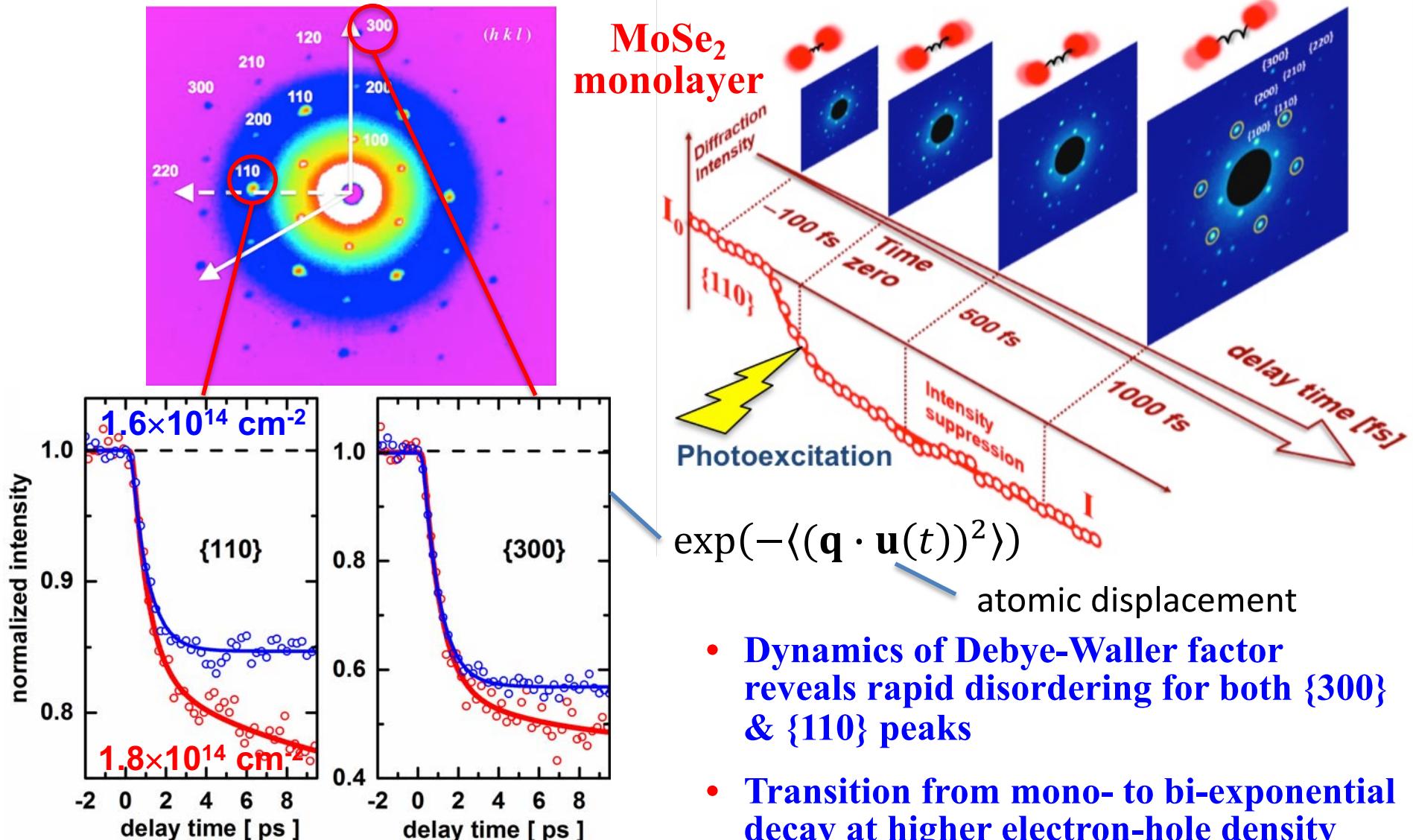
- In ultrafast ‘electron & X-ray cameras,’ laser light hitting a material is almost completely converted into nuclear motions — key to switching material properties on & off at will for future electronics applications.
- High-end nonadiabatic quantum molecular dynamics simulations reproduce the ultrafast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

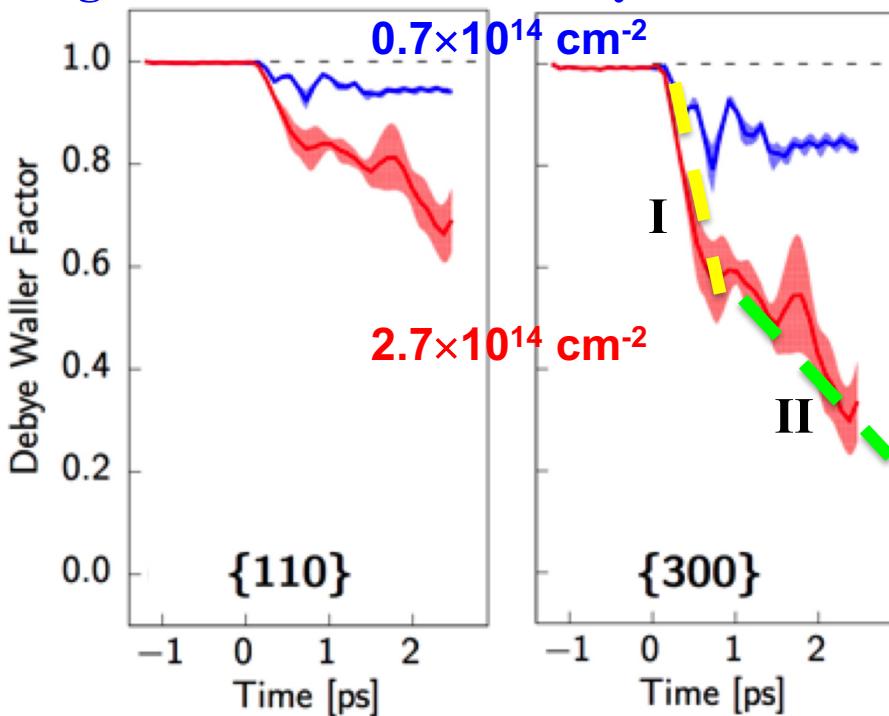
# Ultrafast Coupled Electron-Lattice Dynamics

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



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

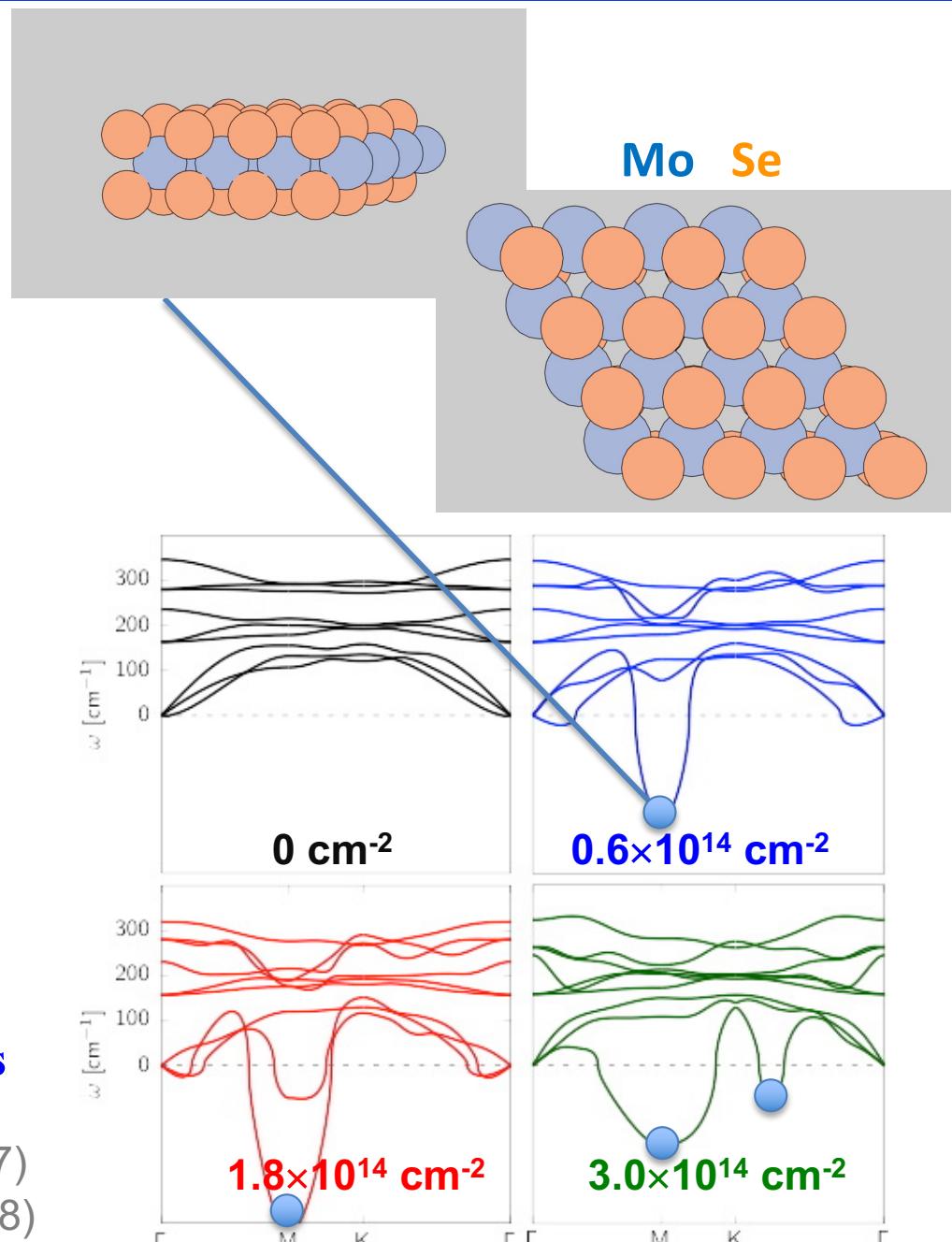
- NAQMD simulations reproduce (1) rapid photo-induced lattice dynamics & (2) mono- to bi-exponential transition at higher electron-hole density



- 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

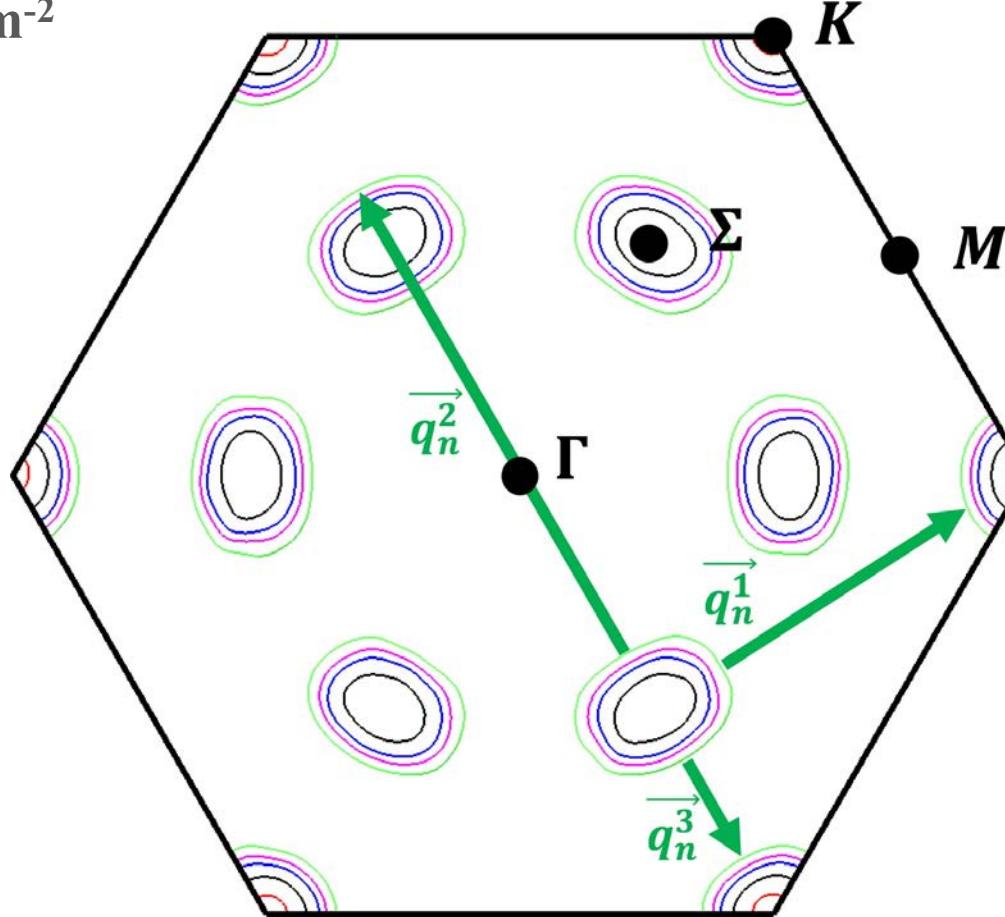
Lin et al., *Nature Commun.* **8**, 1745 ('17)

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  $2 \times 10^{14} \text{ cm}^{-2}$

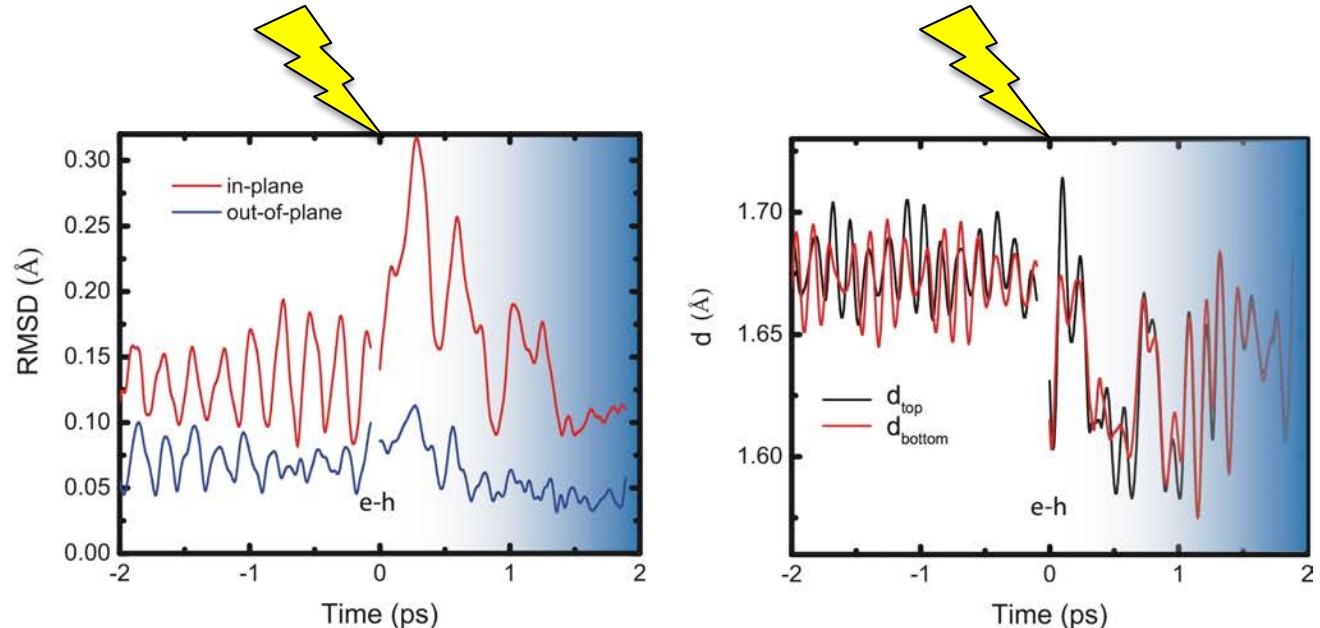
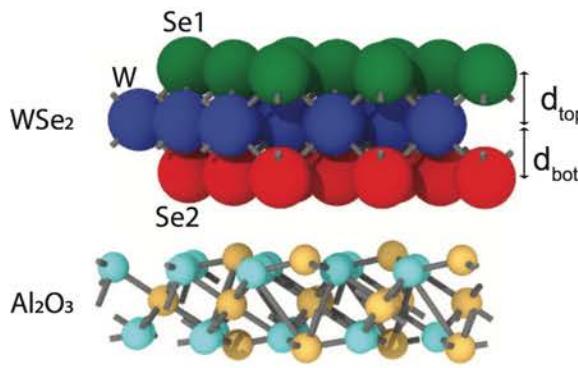


$$n(\text{e-h}) = 0.22, 1, 2, 3, 4 \times 10^{14} \text{ cm}^{-2}$$

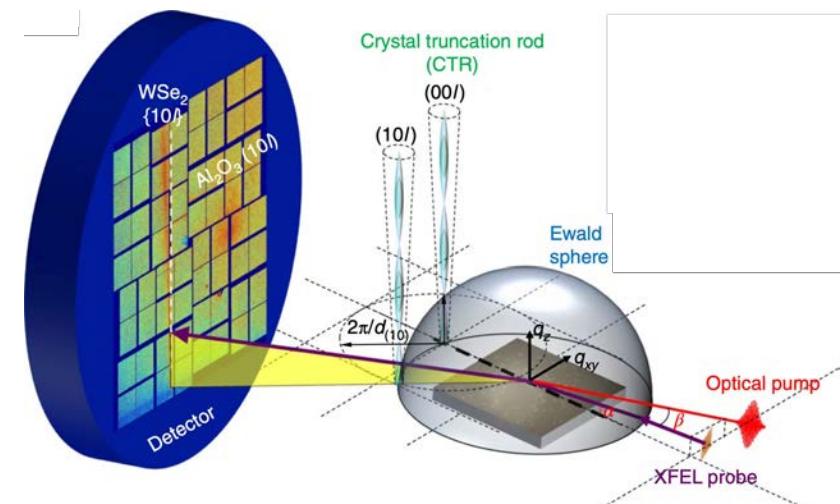
- While the Fermi surface is localized at K-points at minimal excitation (red), it occupies  $\Sigma$ -pockets at larger  $n(\text{e-h})$  (black & blue), enabling electron scattering by emitting  $\vec{q}_n^1$  ( $M$ ),  $\vec{q}_n^2$  ( $\Sigma$ ) and  $\vec{q}_n^3$  ( $K$ ) phonons

# 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)

# Attosecond Light-Matter Interaction

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## The Nobel Prize in Physics 2023



© Nobel Prize Outreach. Photo:

Clément Morin

**Pierre Agostini**

Prize share: 1/3



© Nobel Prize Outreach. Photo:

Clément Morin

**Ferenc Krausz**

Prize share: 1/3



© Nobel Prize Outreach. Photo:

Clément Morin

**Anne L'Huillier**

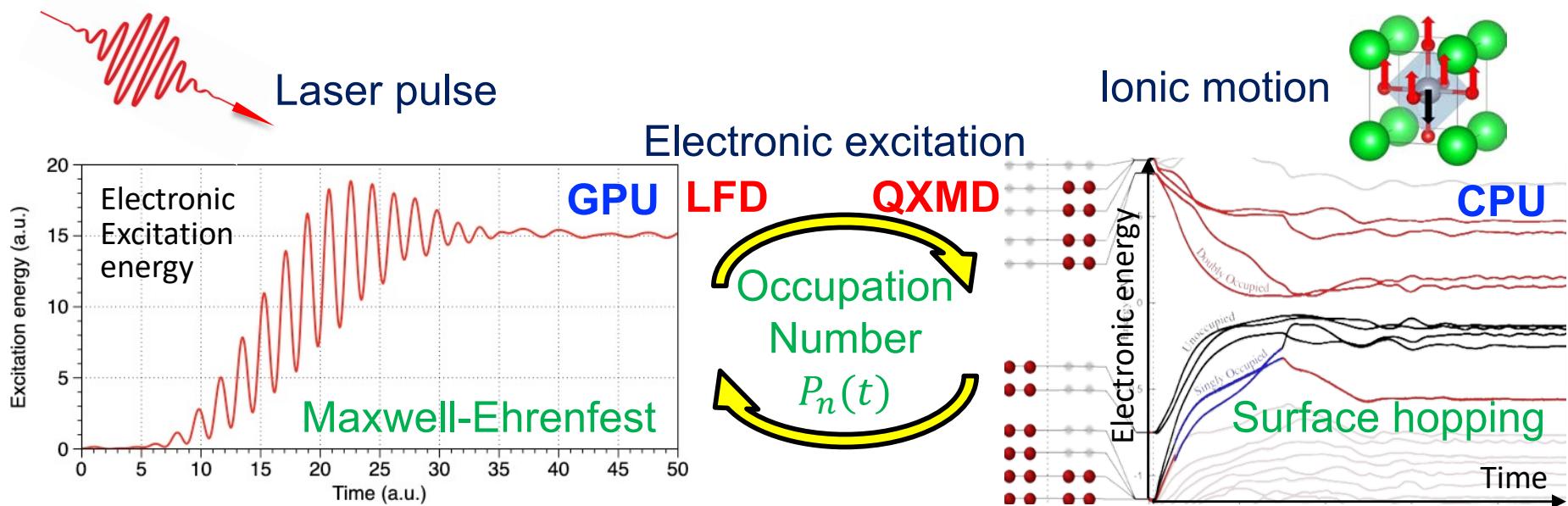
Prize share: 1/3

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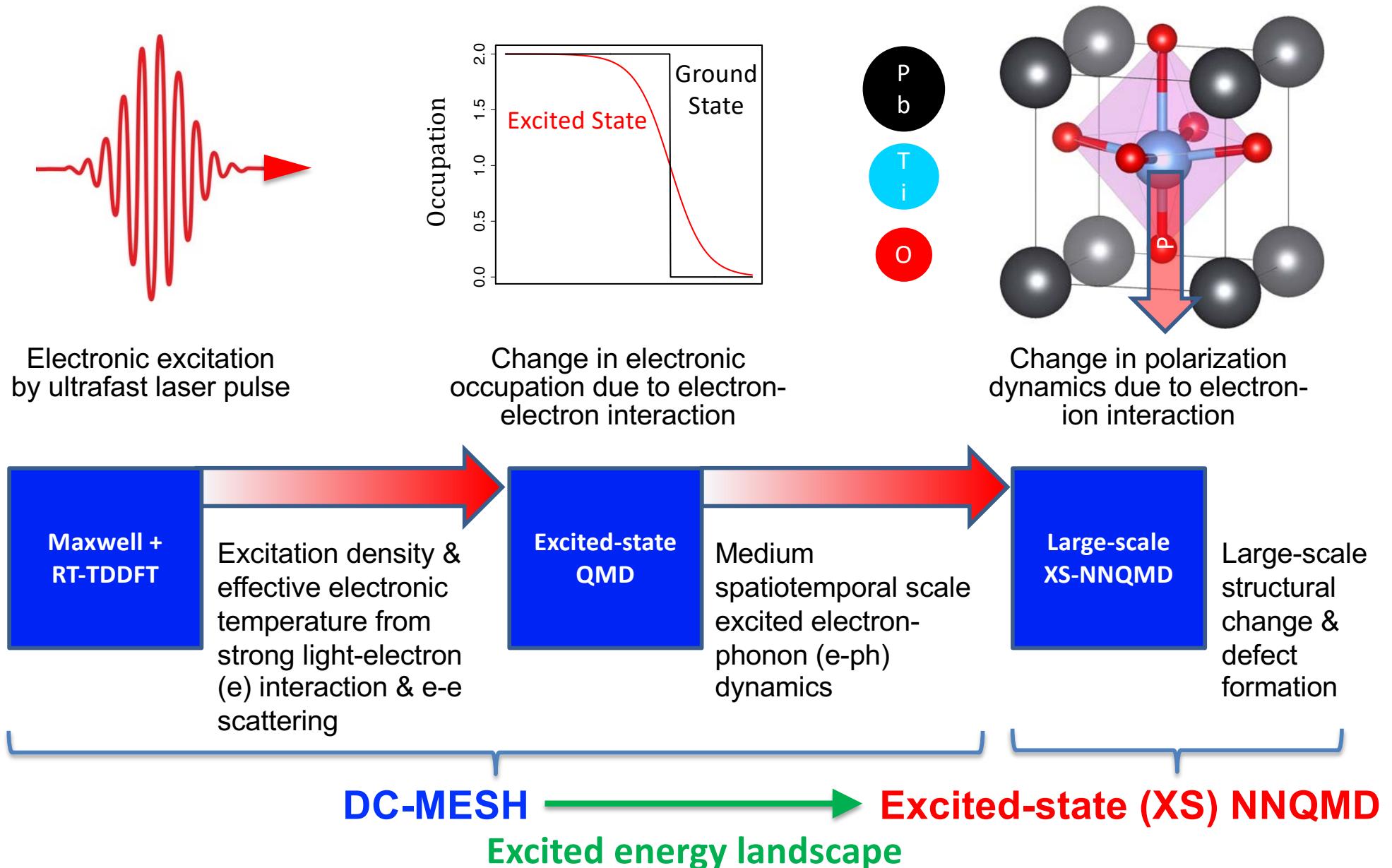
The Nobel Prize in Physics 2023 was awarded to Pierre Agostini, Ferenc Krausz and Anne L'Huillier "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

# 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



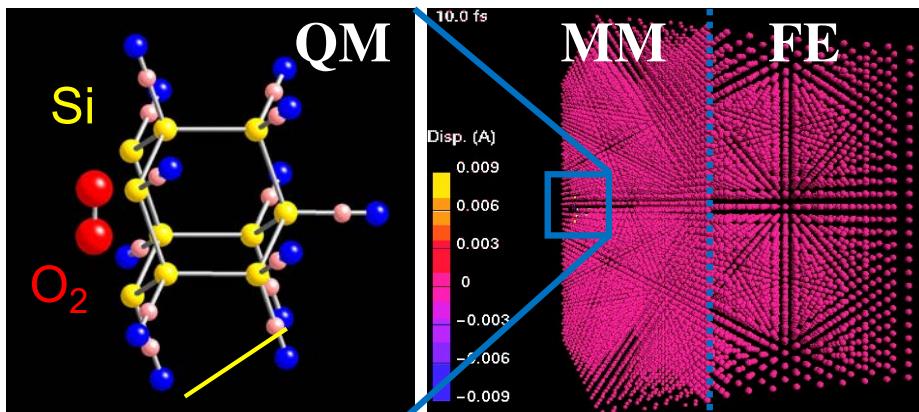
# Multiscaling from DC-MESH to XS-NNQMD



# 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)

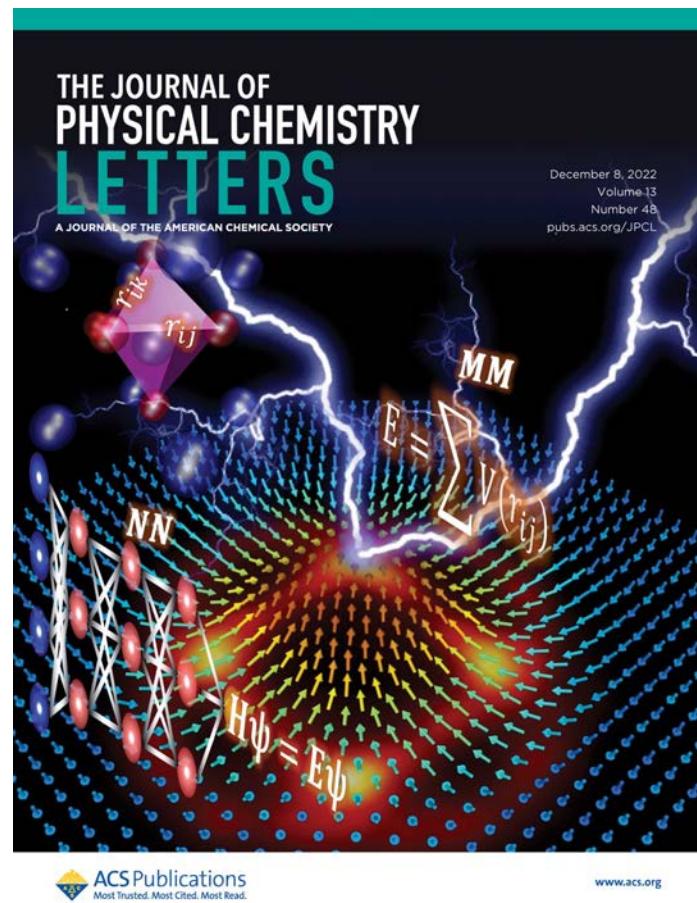
Warshel, *Angew. Chem.* **53**, 10020 ('14)



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

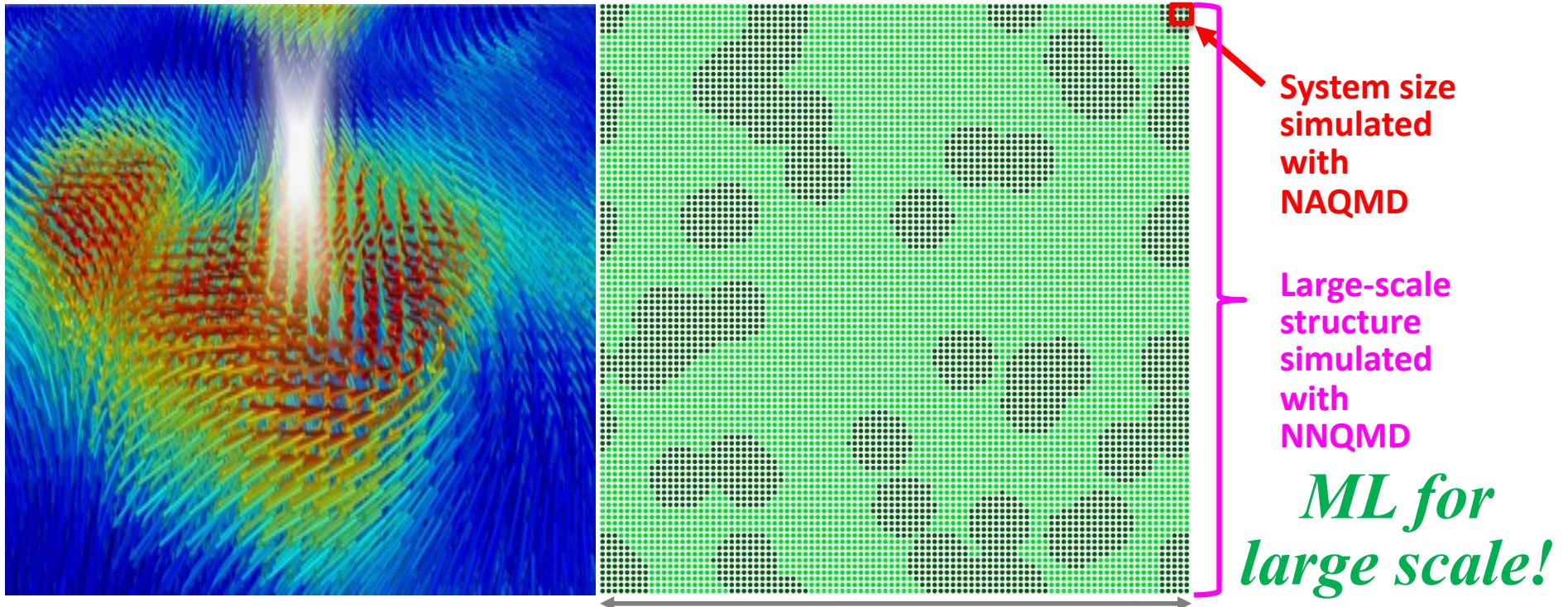
Ogata *et al*, *Comput. Phys. Commun.* **138**, 143 ('01)

- **NN/MM:** NNQMD for ferroelectric ( $\text{PbTiO}_3$ : PTO) embedded in MM for paraelectric ( $\text{SrTiO}_3$ : STO) to apply appropriate strain boundary condition



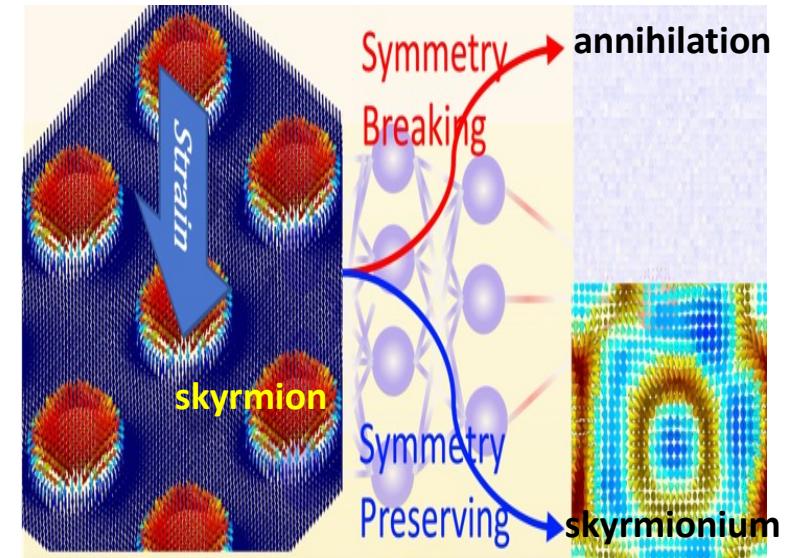
Linker *et al.*, *J. Phys. Chem. Lett.* **13**, 11335 ('22); *Nano Lett.* **23**, 7456 ('23)

# Application: Ferroelectric Opto-Toptronics



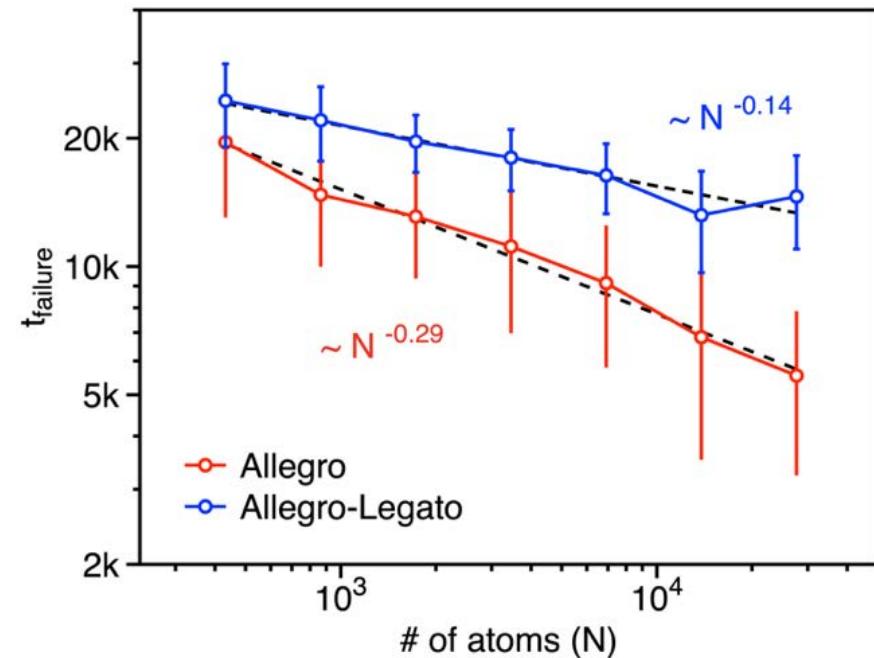
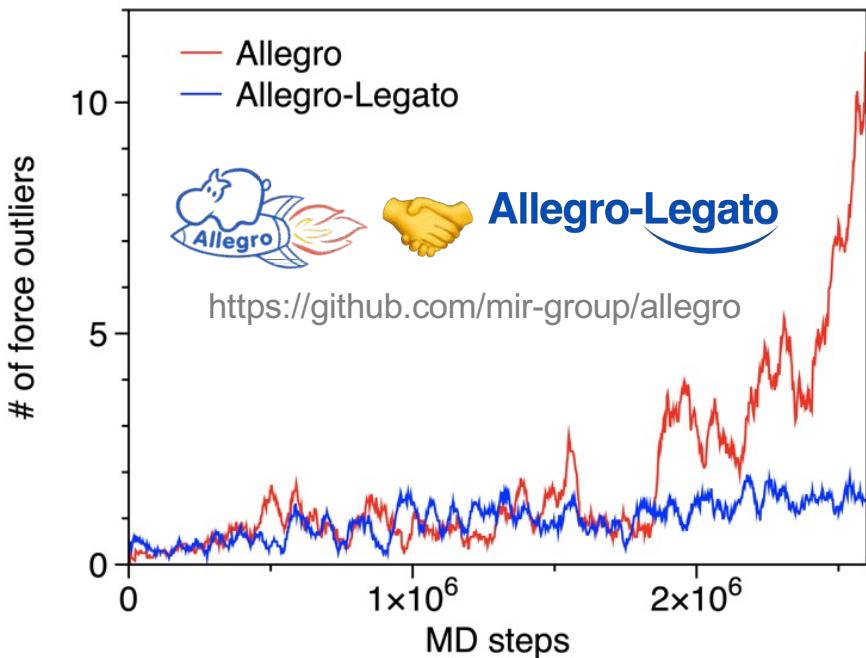
- Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications
- Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)
- Symmetry-controlled skyrmion-to-skyrmionium\* switching

\*Composite of skyrmions with opposite topological charges  
Linker *et al.*, *Science Adv.* **8**, eabk2625 ('22);  
*JPCl* **13**, 11335 ('22); *Nano Lett.* **23**, 7456 ('23)



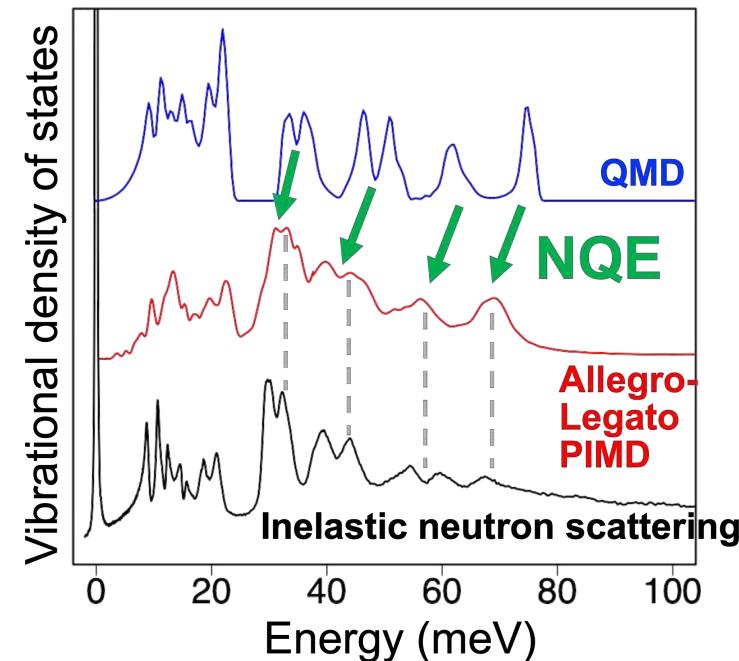
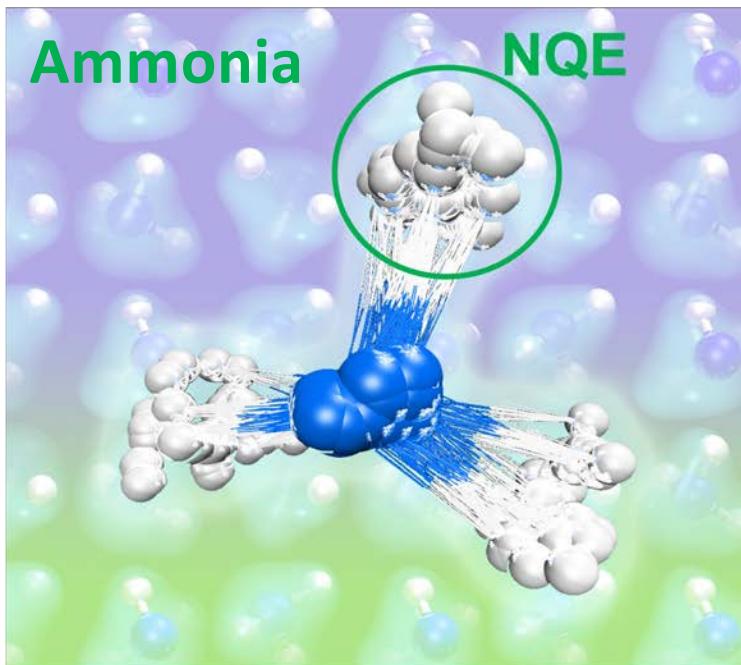
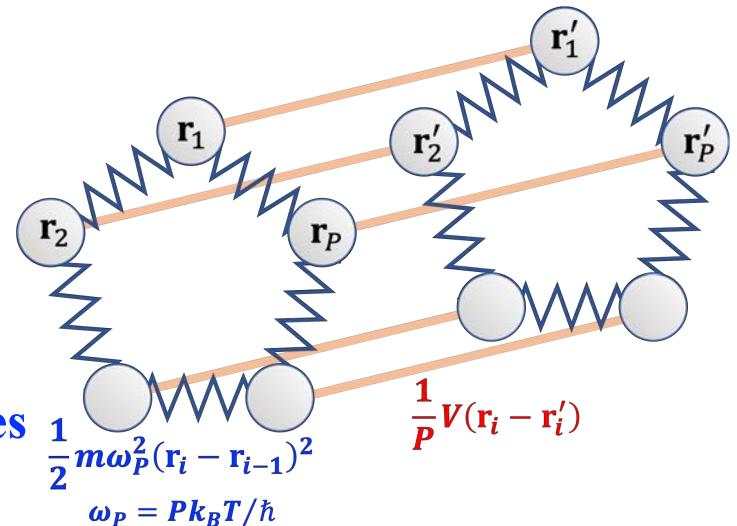
# Fast & Robust NNQMD: Allegro-Legato

- **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



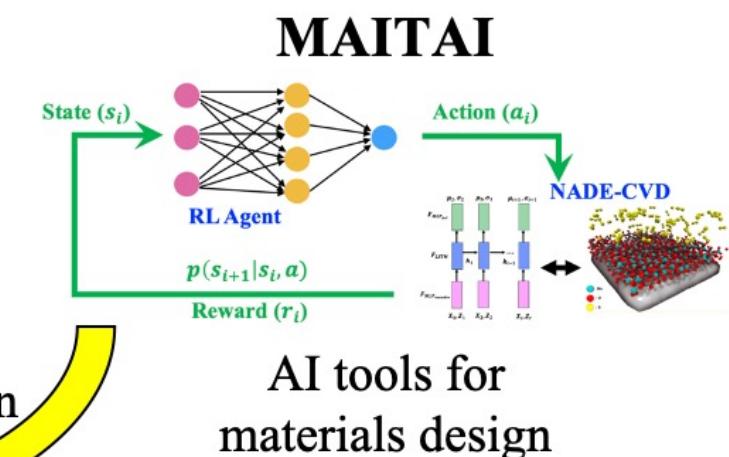
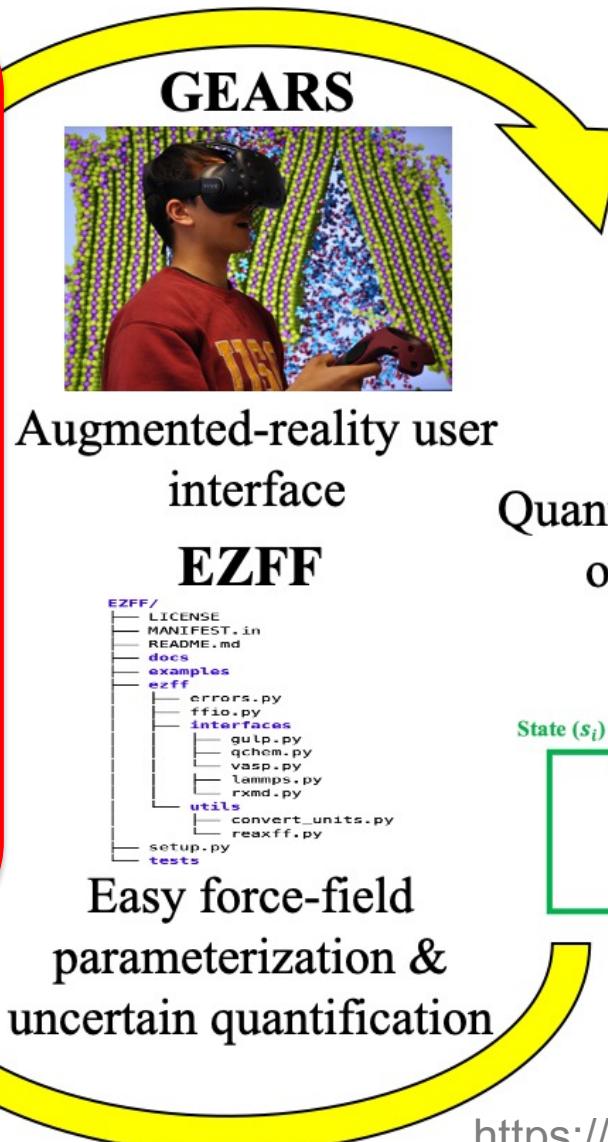
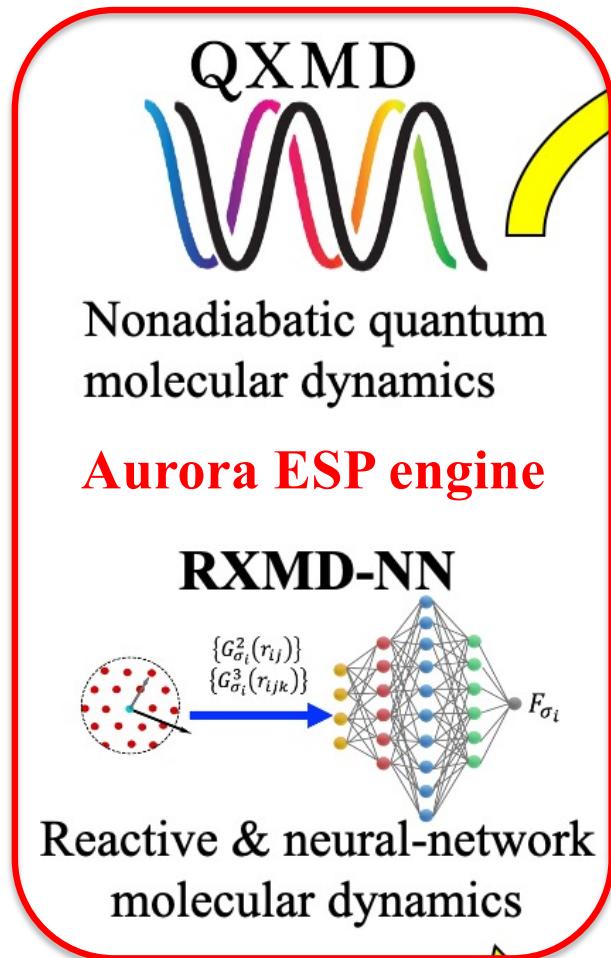
# Nuclear-Quantum NNQMD

- **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

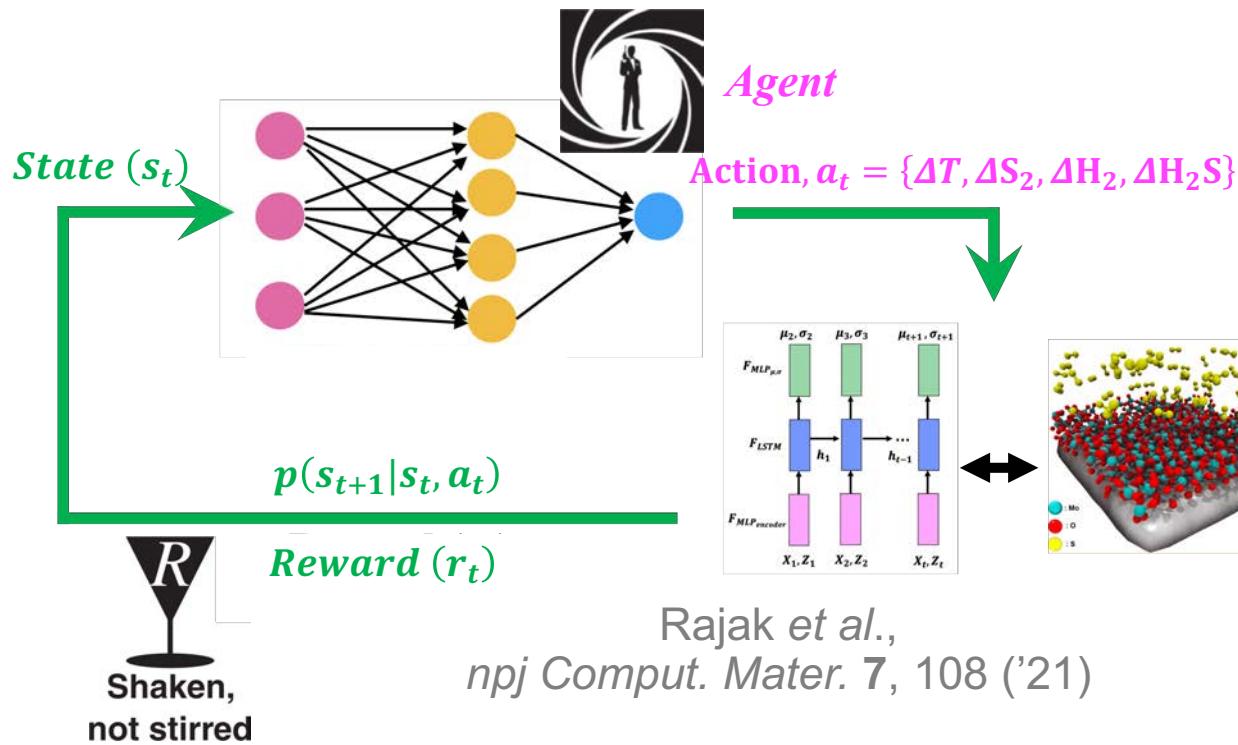


You will obtain hands-on training on QXMD

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

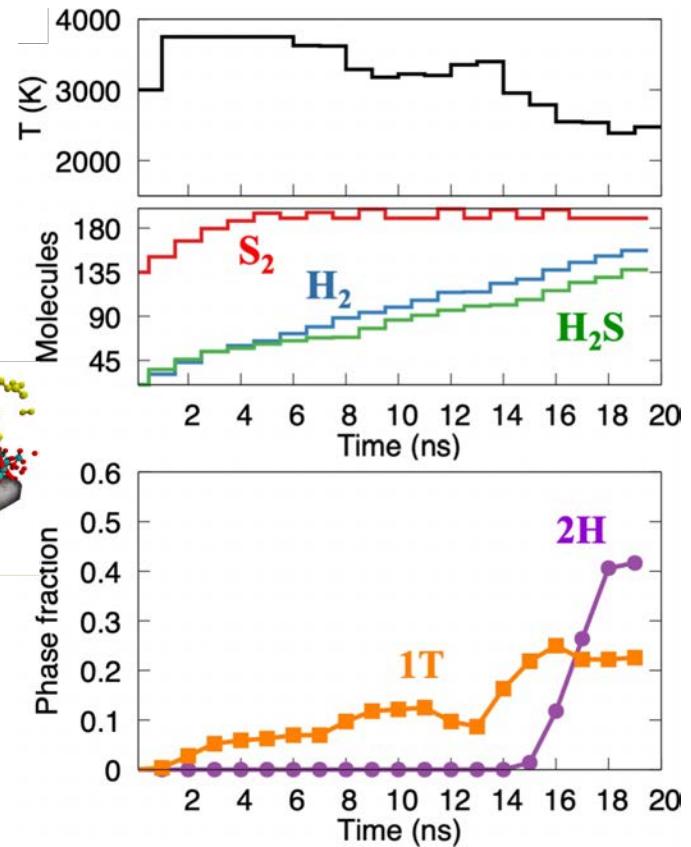
# Reinforcement Learning for Growth

- 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)

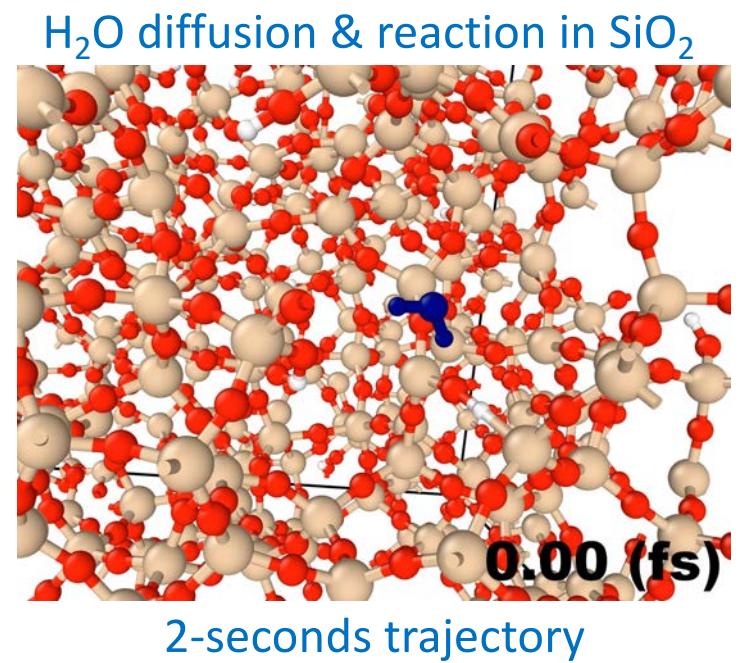
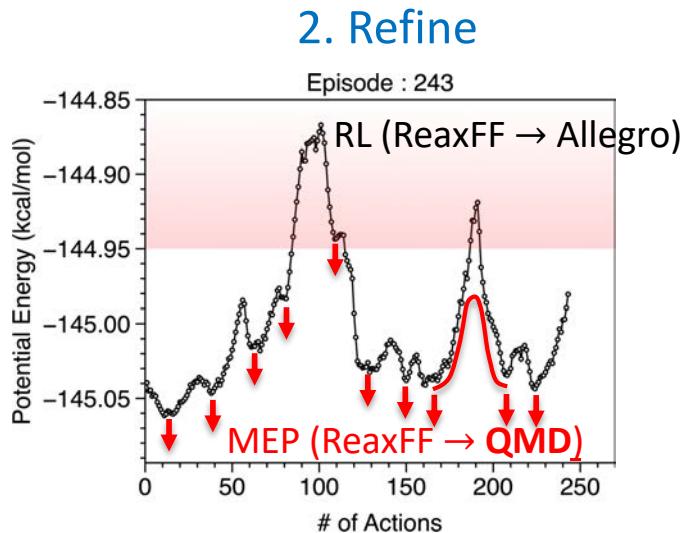
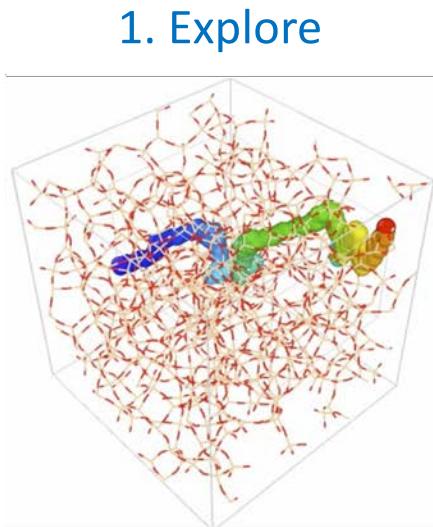
cf. Sgroi et al., *Phys. Rev. Lett.* 126, 020601 ('21)



# 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

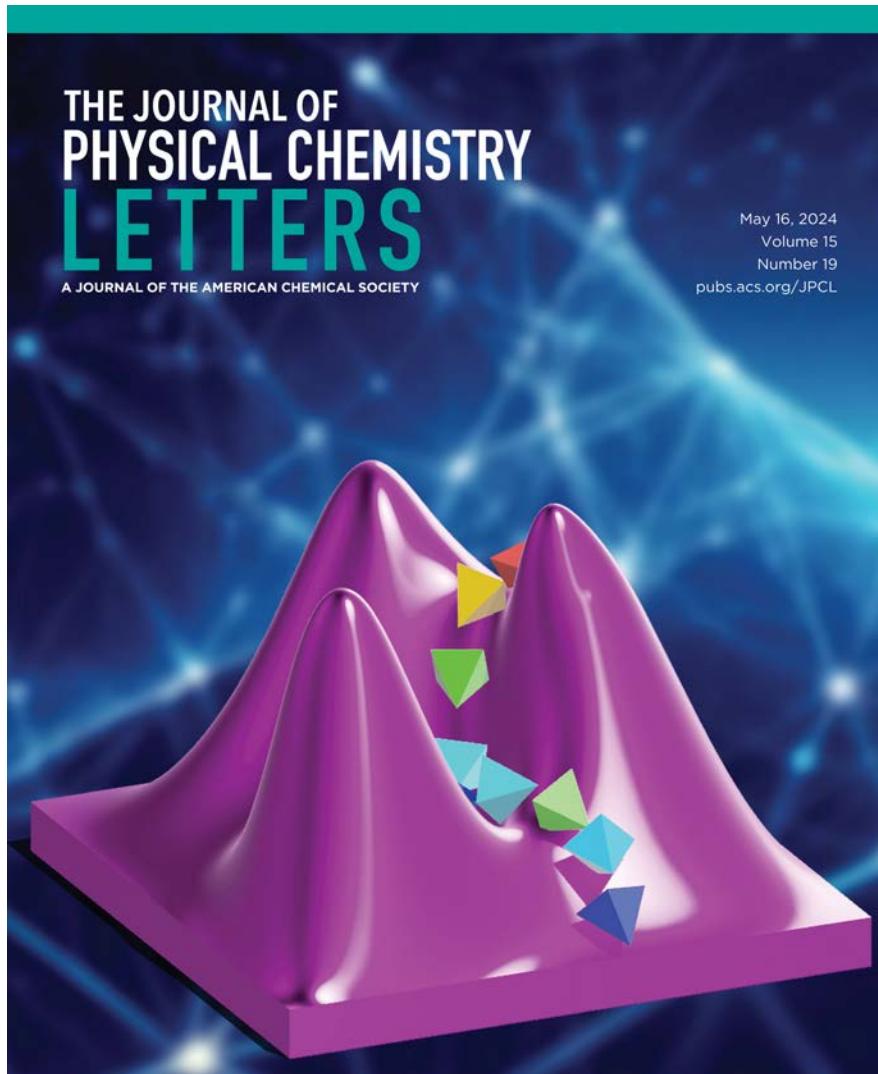
$$t_{\text{migration}} = \sum_{i \in \{\text{activation events}\}} \frac{\hbar}{k_B T} \exp\left(\frac{E_i^{\text{activation}}}{k_B T}\right)$$



*AI for long time!*

# RL to Enable Long-Time Dynamics

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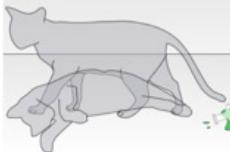


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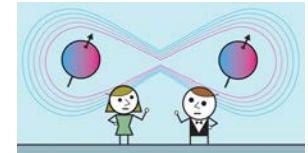
[www.acs.org](http://www.acs.org)

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*

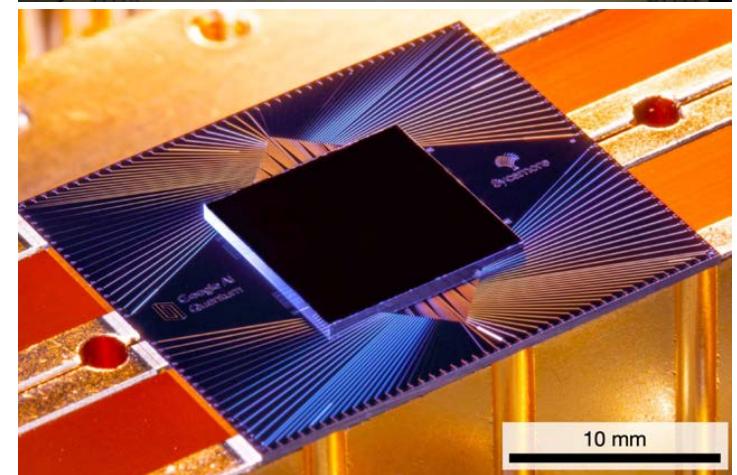
R. P. Feynman, *Int. J. Theo. Phys.* **21**, 467 ('82);  
S. Lloyd, *Science* **273**, 1073 ('96)

- 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

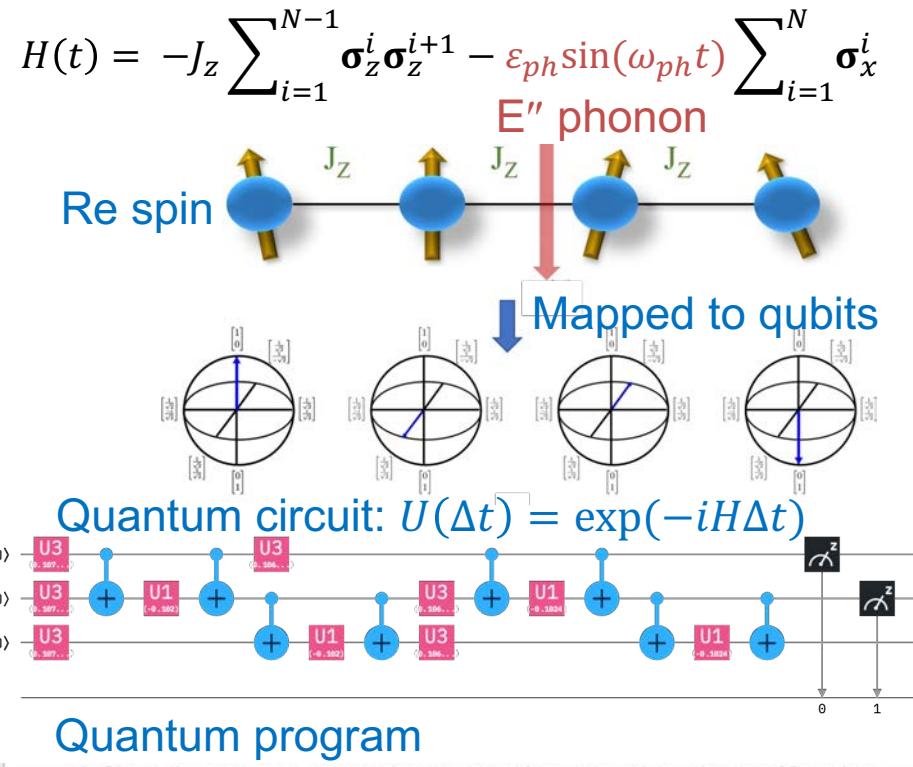
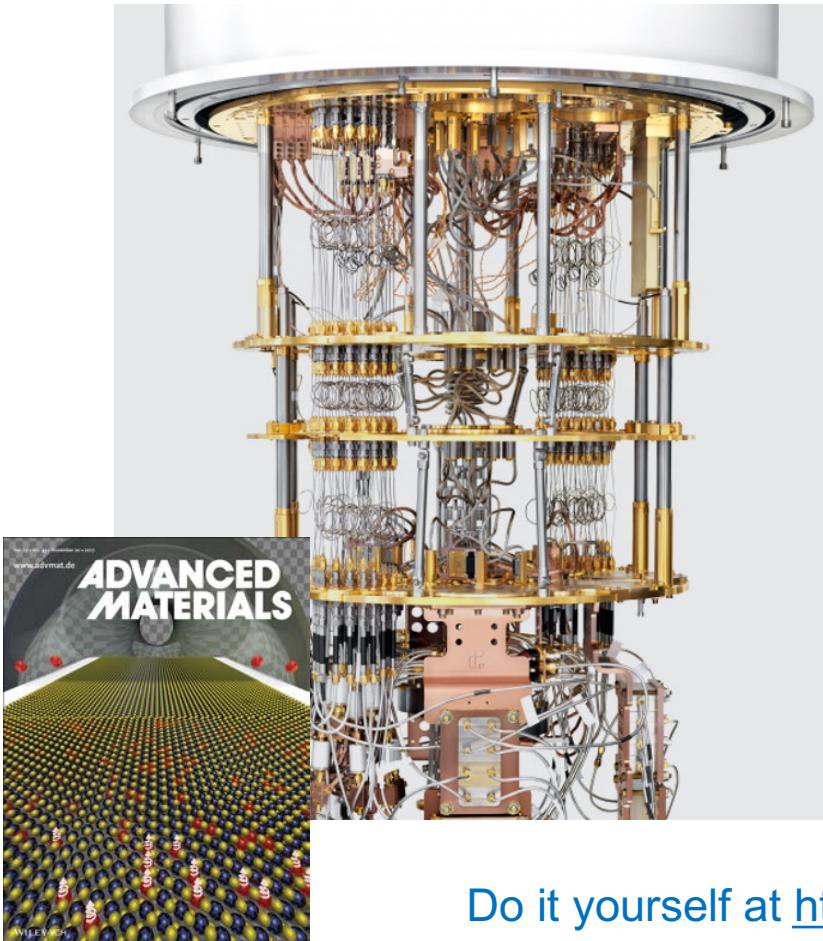
J. Preskill, *Quantum* **2**, 79 ('18)



54-qubit Google Sycamore

# Quantum Computing of Magnetism

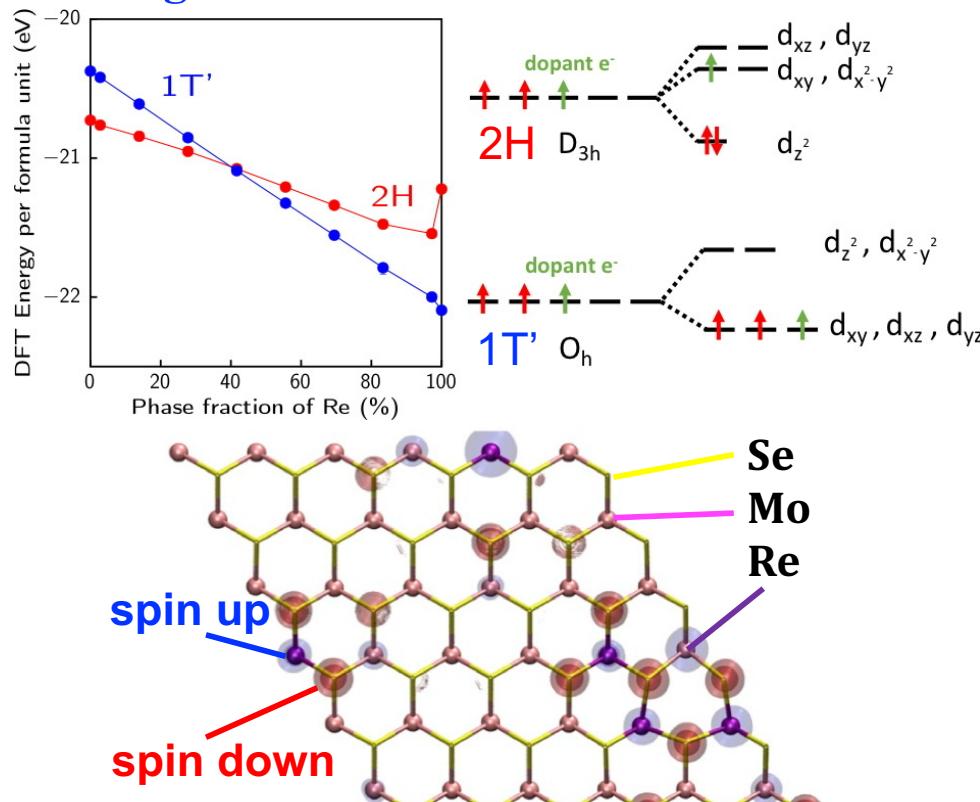
- Simulated quantum many-body dynamics on IBM's Q16 Melbourne & Rigetti's Aspen quantum processors
- Electromagnetic-field control of quantum states in a chain of rhenium-magnets in MoSe<sub>2</sub> monolayer to realize desired material properties on demand, thereby pushing the envelope of “quantum materials science”



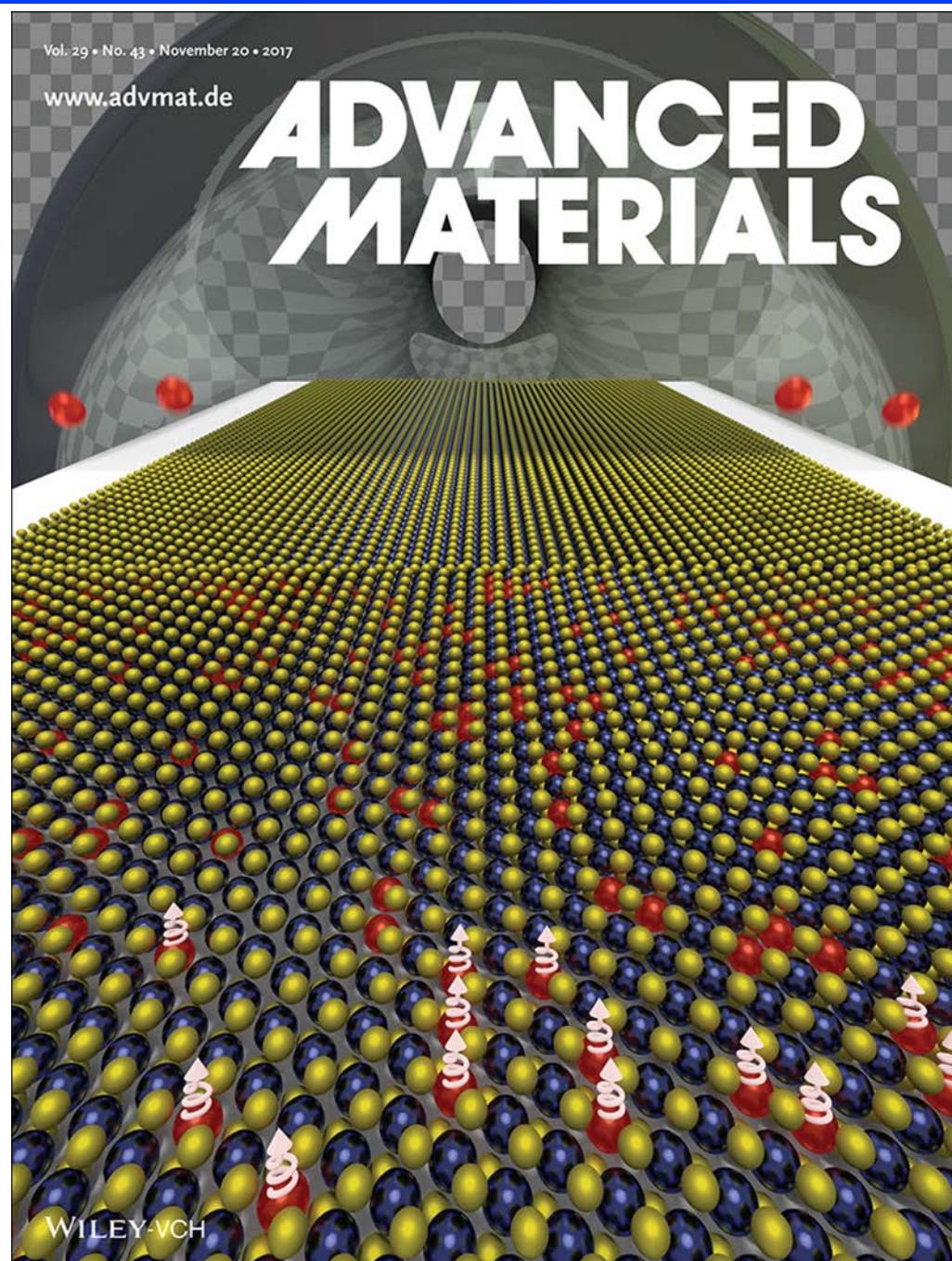
Do it yourself at <https://quantum-computing.ibm.com>

# 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)

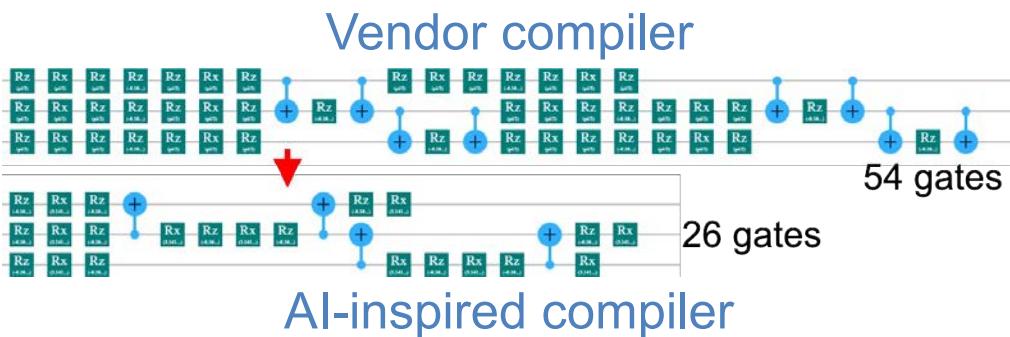


# Quantum Dynamics on Quantum Computers

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

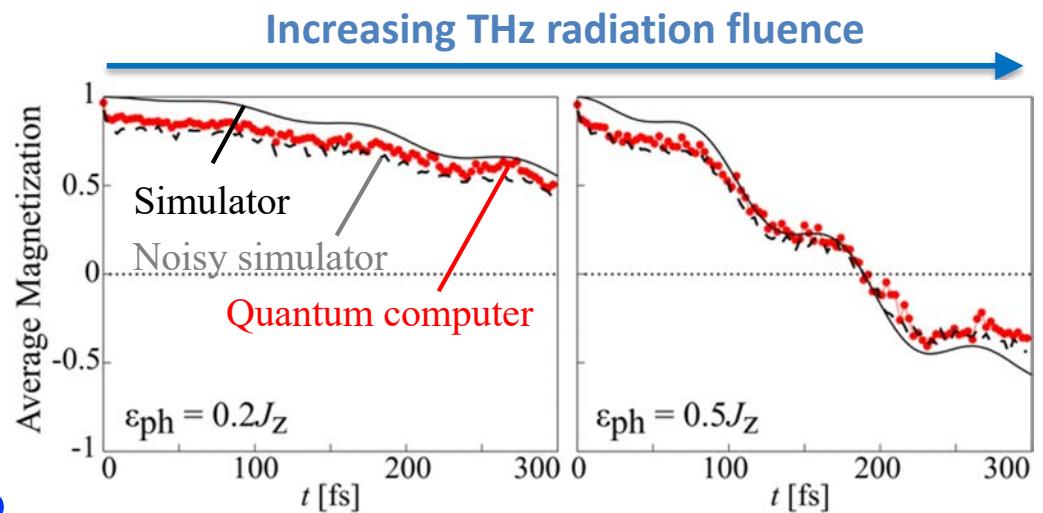
Bassman *et al.*,  
Phys. Rev. 101, 184305 ('20)

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

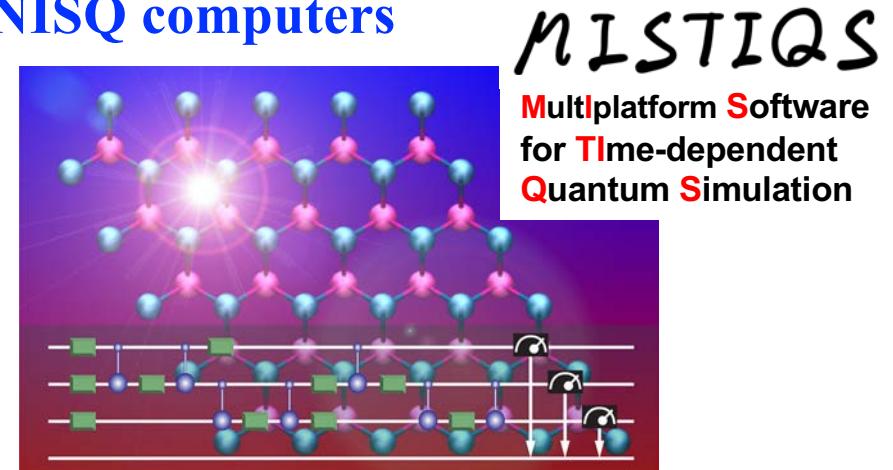


Bassman *et al.*,  
Quantum Sci. Tech. 6, 014007 ('21)

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

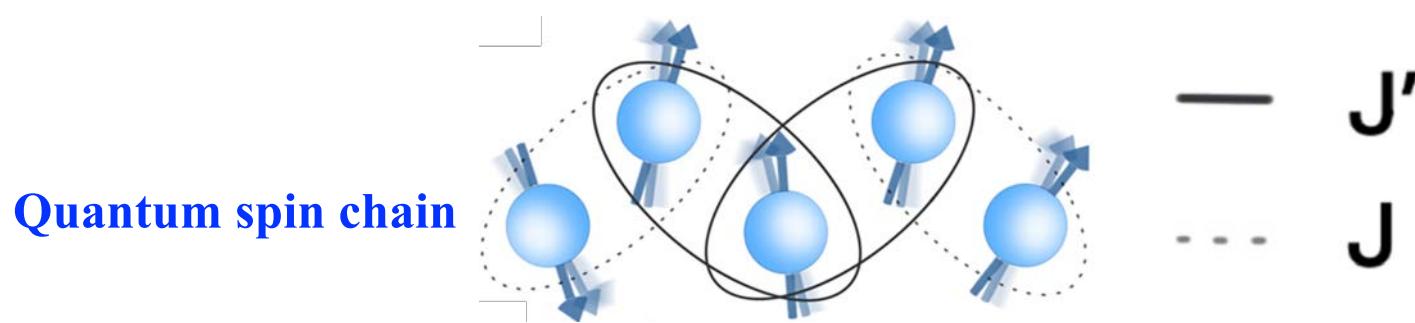


- Full-stack, cross-platform software for quantum dynamics simulations on NISQ computers



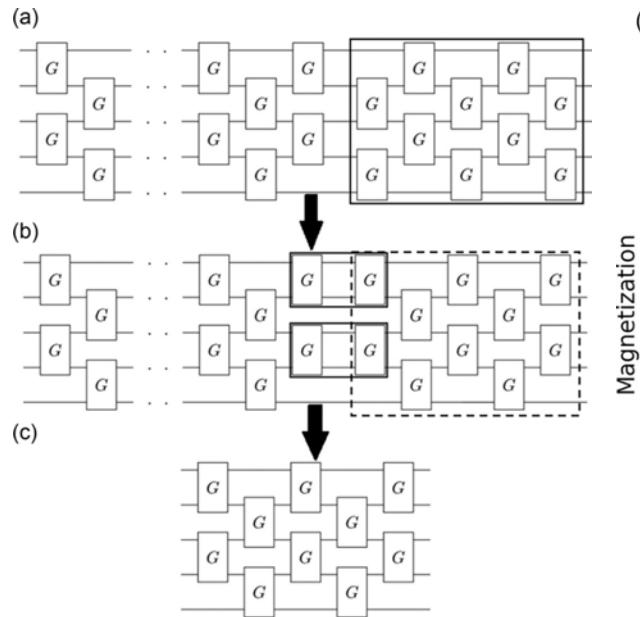
Powers *et al.*, SoftwareX 14, 100696 ('21)  
<https://github.com/USCCACS/MISTIQS>

# Topological Quantum Dynamics

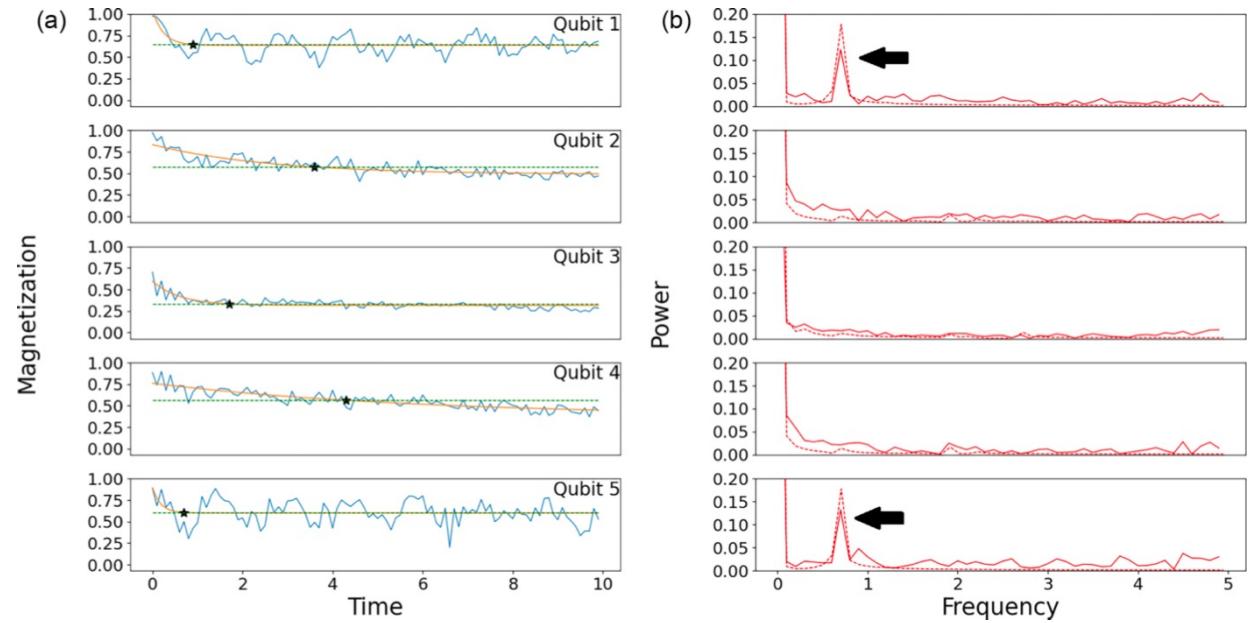


Quantum spin chain

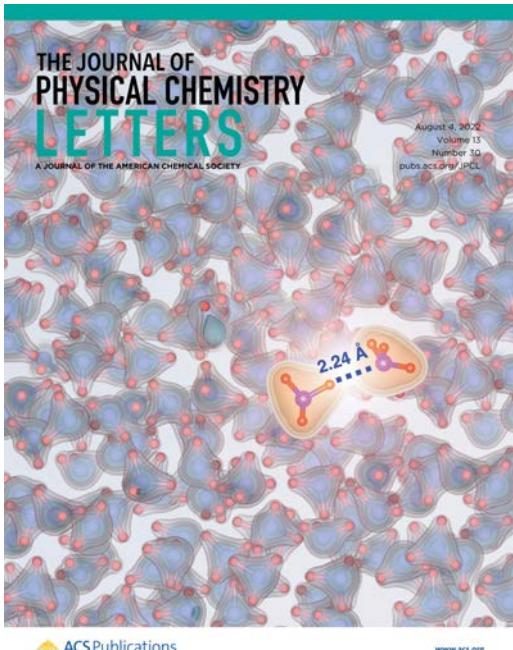
Constant-depth quantum circuit



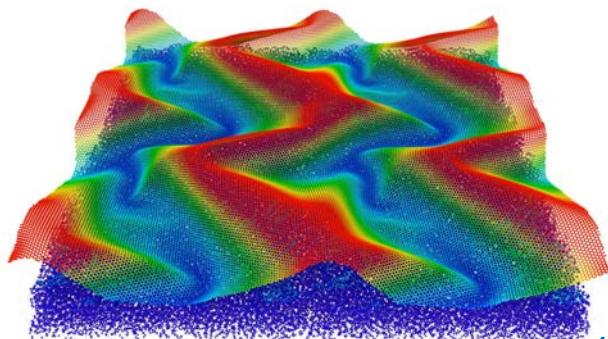
Topological surface mode



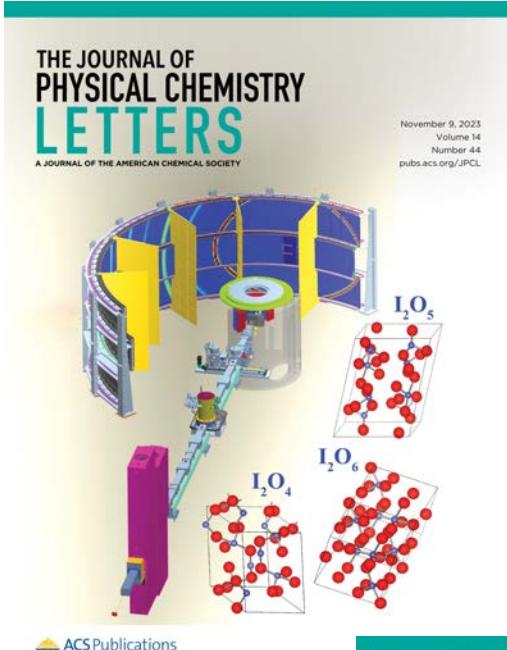
# Other Applications



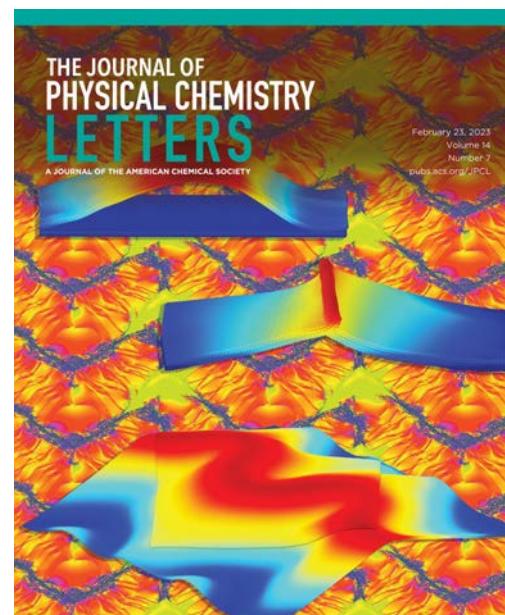
Green ammonia for  
renewable power  
(Aug. 4, '22)



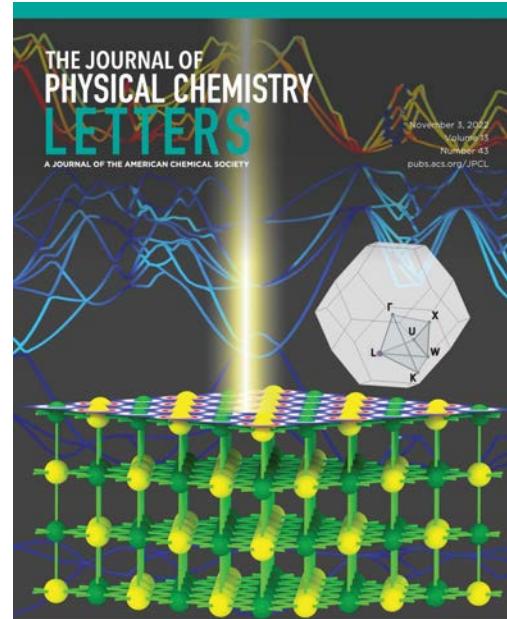
Strain self-  
assembly of  
2D  
metasurface  
(Feb. 23, '23)



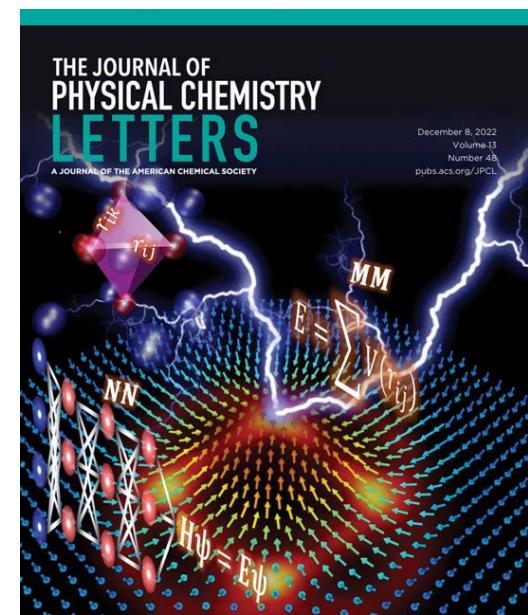
Neutron  
scattering  
(Nov. '23)



ACS Publications  
Most Trusted. Most Cited. Most Read.



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

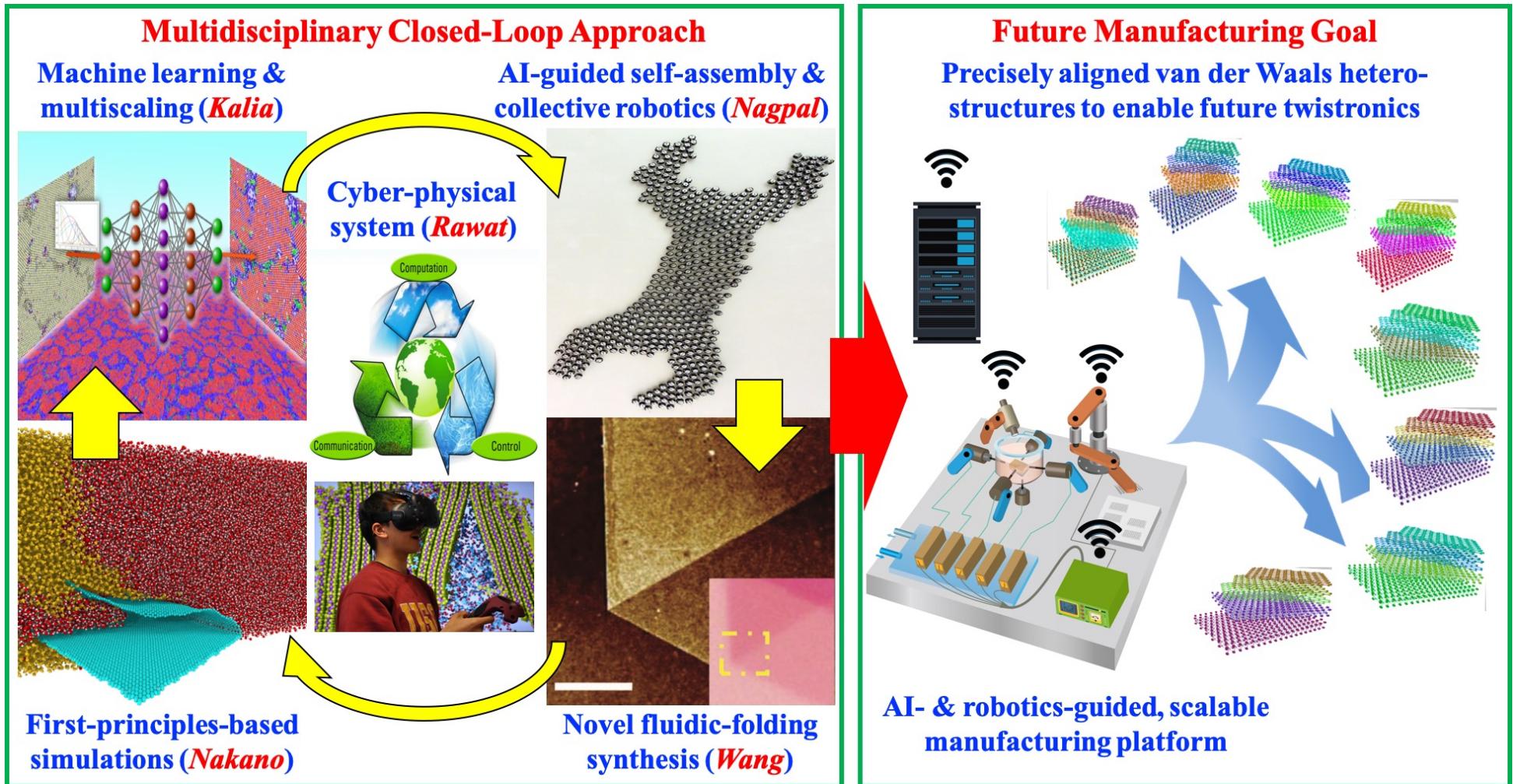


ACS Publications  
Most Trusted. Most Cited. Most Read.

Topotronics  
(Dec. 8, '22)

# 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)

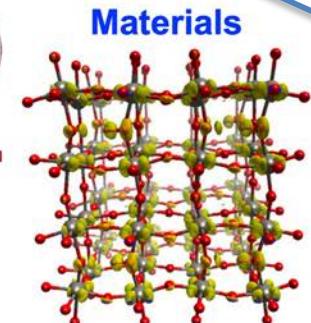
# NSF Future Semiconductors Project

- 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**

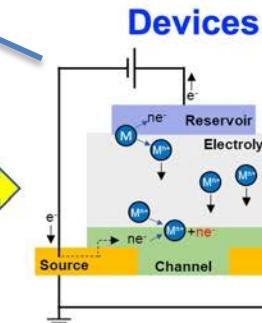
## Conductivity switch in H-doped WO<sub>3</sub>

### Protonic synapse (MIT)

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



Dev, Krishnamoorthy, Kalia, Li

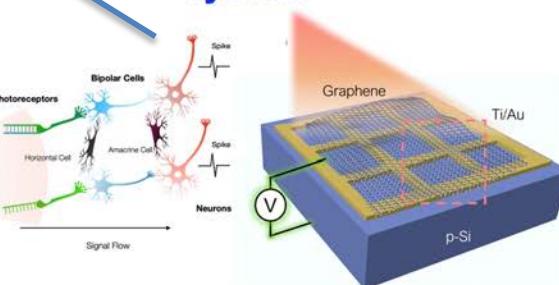


Wang, Yang, Zhang, Yildiz

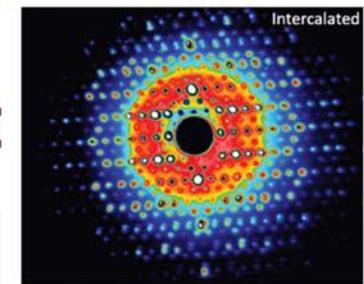
## Photo-induced negative differential resistance (NDR) in graphene/Si

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

### System



del Alamo, Kapadia, Lin, Raina



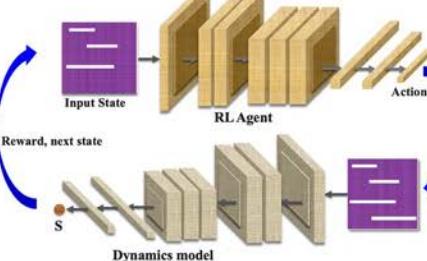
Lindenberg, Wei

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

### UE/XS

### AI/CS

### FuSe Workforce Development



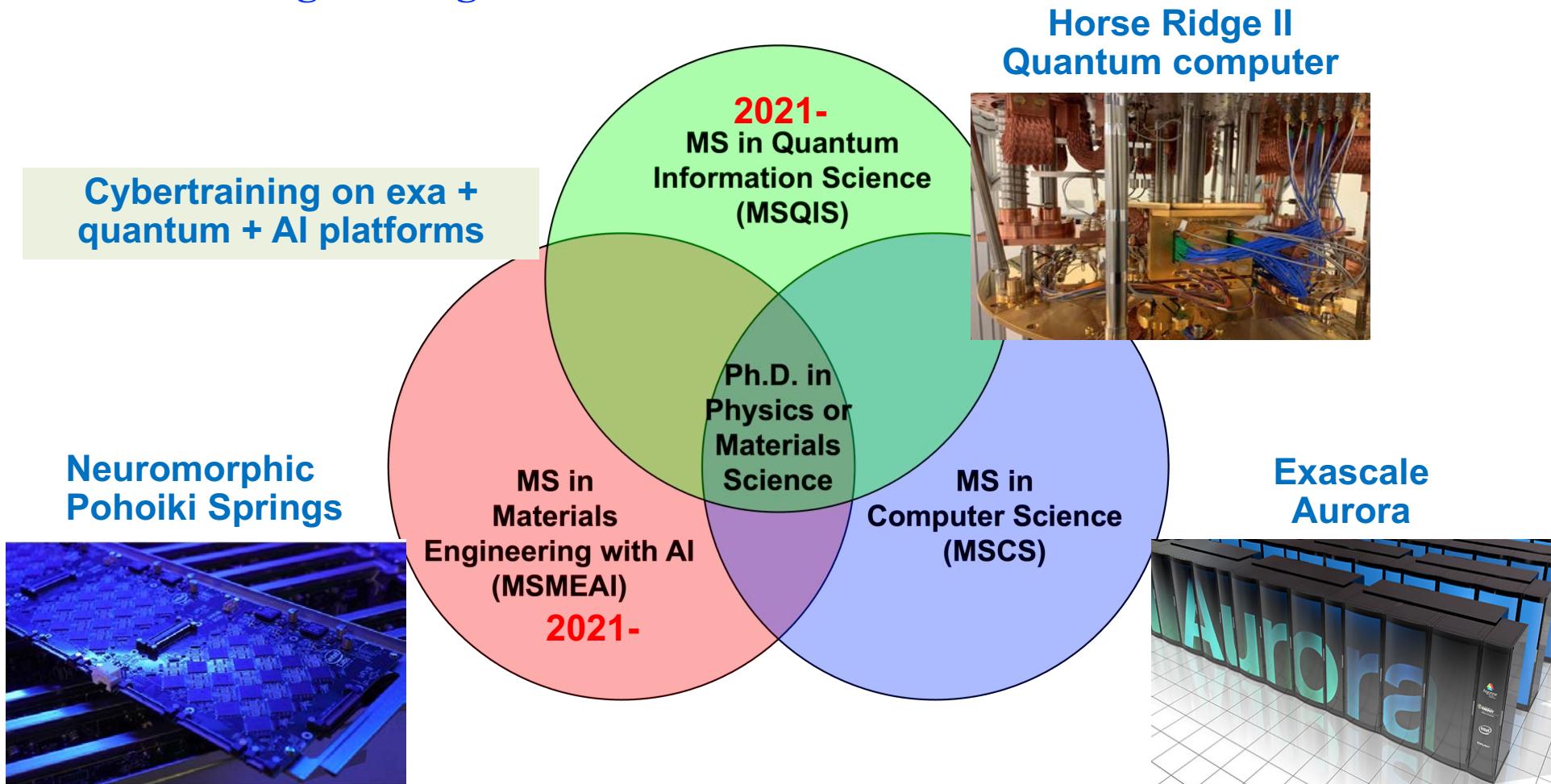
Vashishta, Levi, Nakano, Nomura



Nikias, Katehi, Raghavendra, Rawat

# Training Cyber-Science Workforce

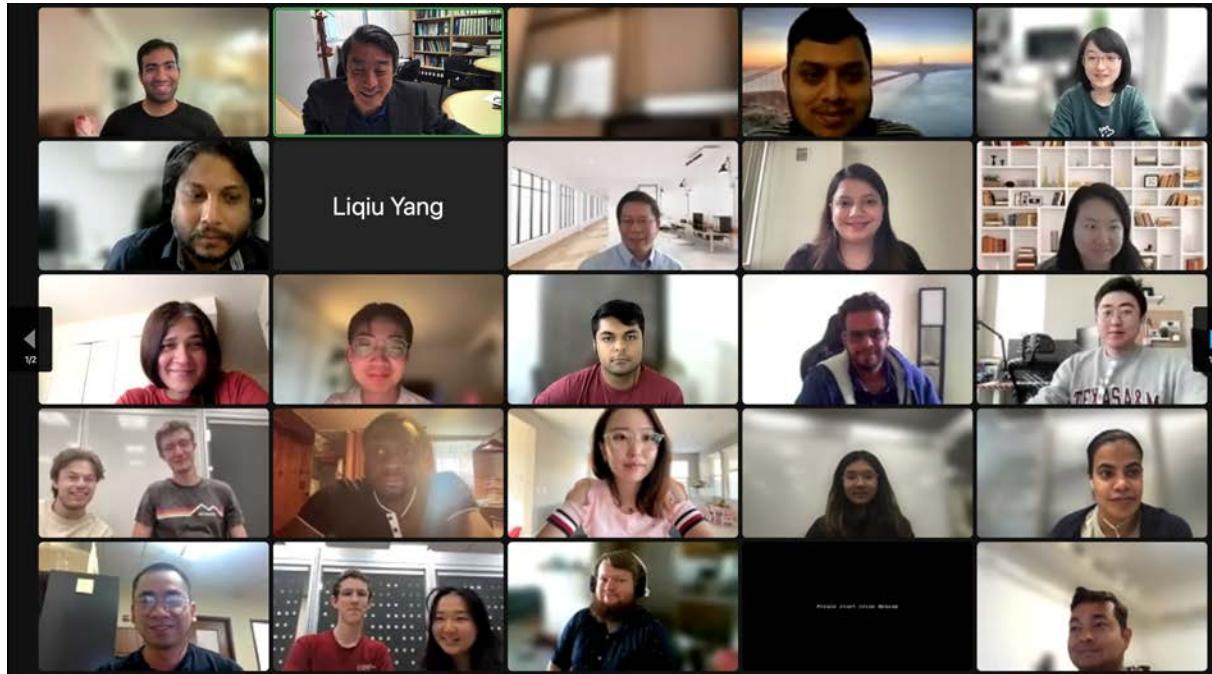
- New generation of computational scientists at the **nexus of exascale computing, quantum computing & AI**
- Unique dual-degree program: Ph.D. in materials science or physics, along with MS in computer science, MS in quantum information science or MS in materials engineering with AI



# 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

