

# Quantum Molecular Dynamics Simulations

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QXMD software tutorial:

Anikeya Aditya, Nabankur Dasgupta,  
Suryakanti Debata, Taufeq Razakh



Supported by National Science Foundation,  
Award OAC-2118061



*CyberMAGICS Workshop*  
June 5, 2025

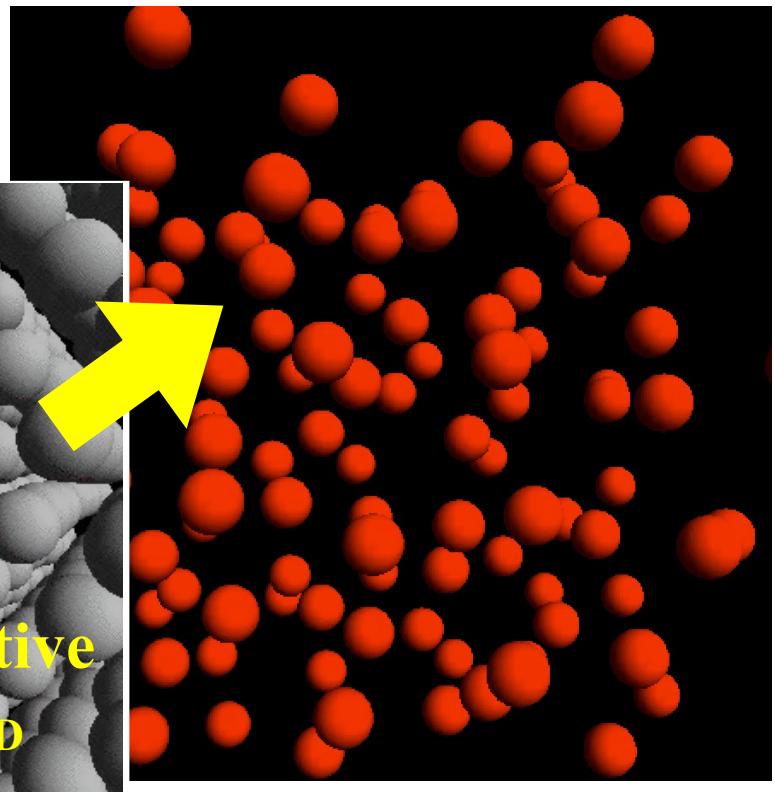
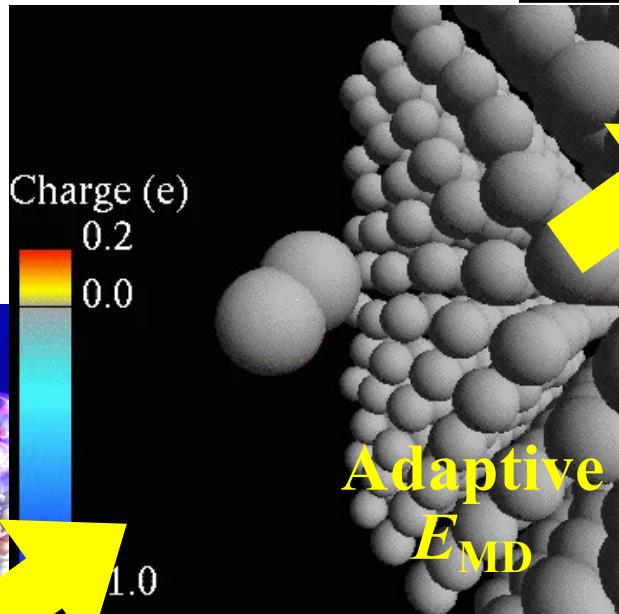
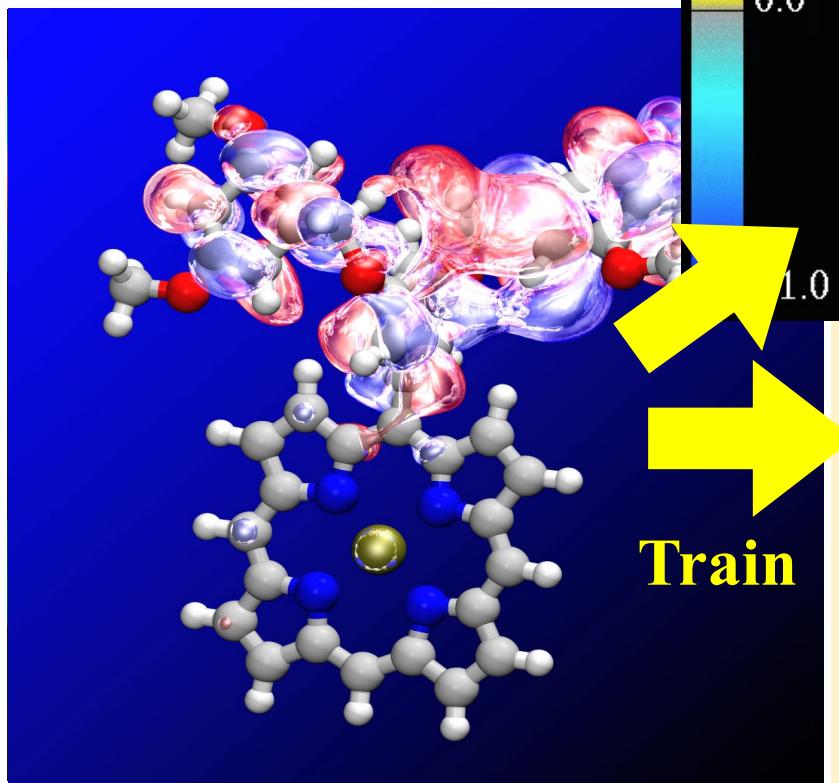


# Molecular Dynamics

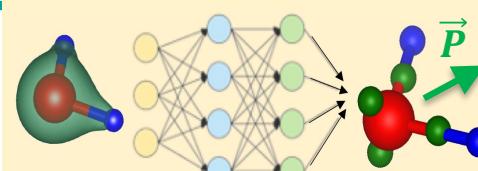
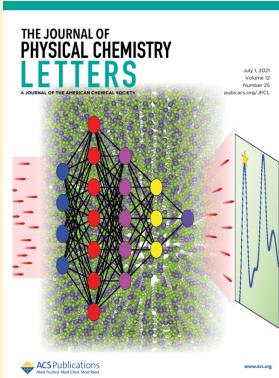
## Molecular Dynamics (*MD*)

### Reactive MD (*RMD*)

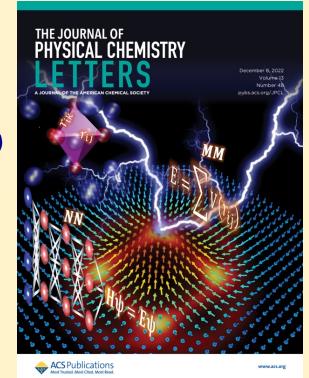
### Nonadiabatic quantum MD (*NAQMD*)



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



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



# 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)



*Electrons*

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

*Complexity reduction*

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       $N$  1-electron problems  
intractable                      tractable

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

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 data-locality 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)

# 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



# Adiabatic Quantum Molecular Dynamics

- 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

- In adiabatic quantum molecular dynamics based on Born-Oppenheimer approximation, 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

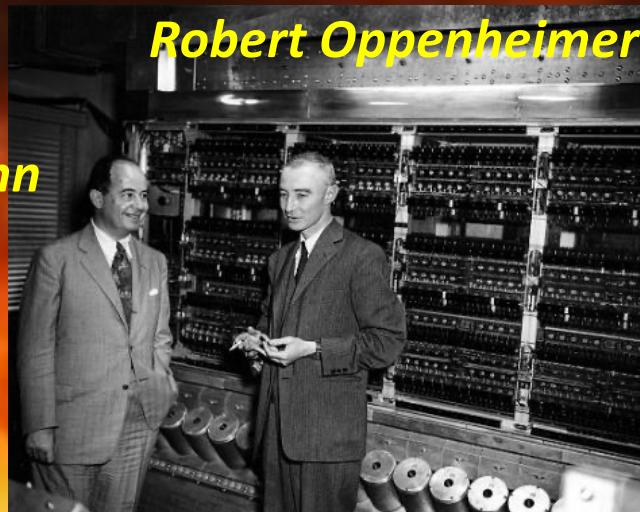
Nº 20

## ANNALEN DER PHYSIK VIERTE FOLGE. BAND 84

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

# In the Beginning ...

*John von Neumann*



**Manhattan project**

*Los Alamos, NM, 1945*

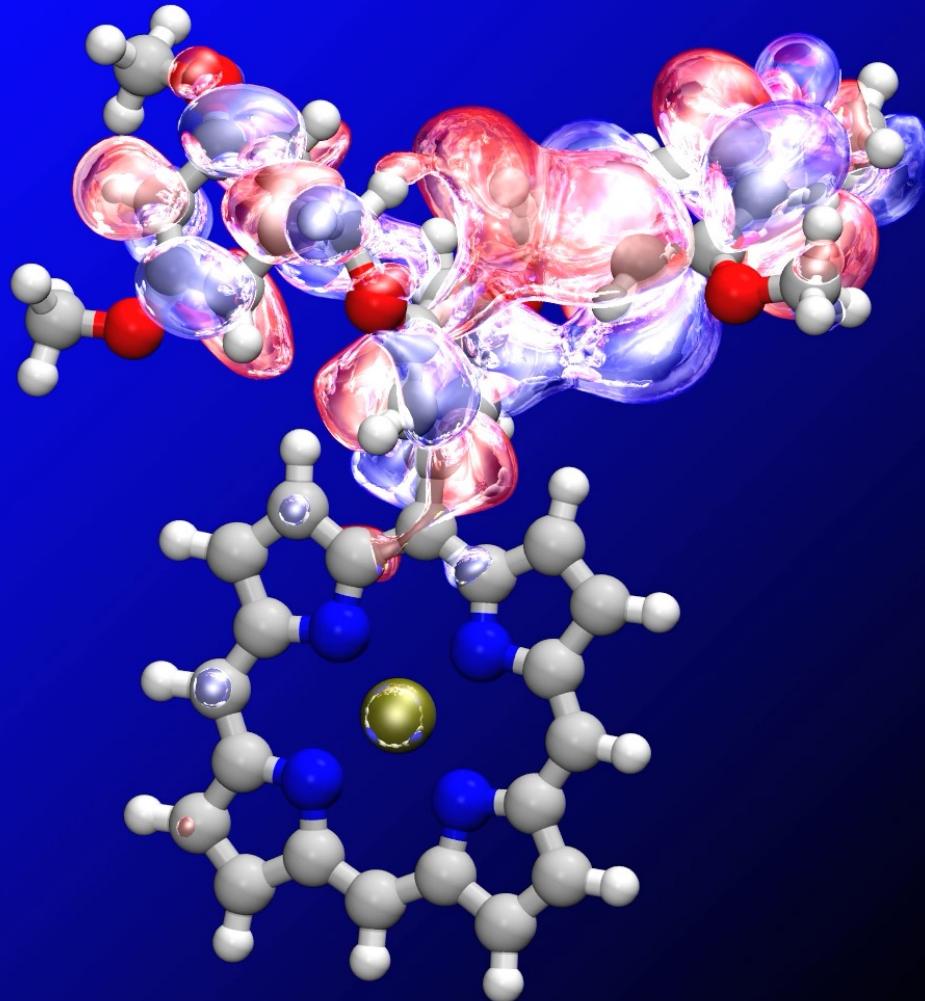
*Journey of Electrons and Atoms: Personal Story of Quantum Mechanics & Supercomputing (Nov. 16 & 17, '24, Culver City, CA)*



<https://magazine.viterbi.usc.edu/spring-2025/engineering/just-dance/>

# Nonadiabatic Quantum Molecular Dynamics

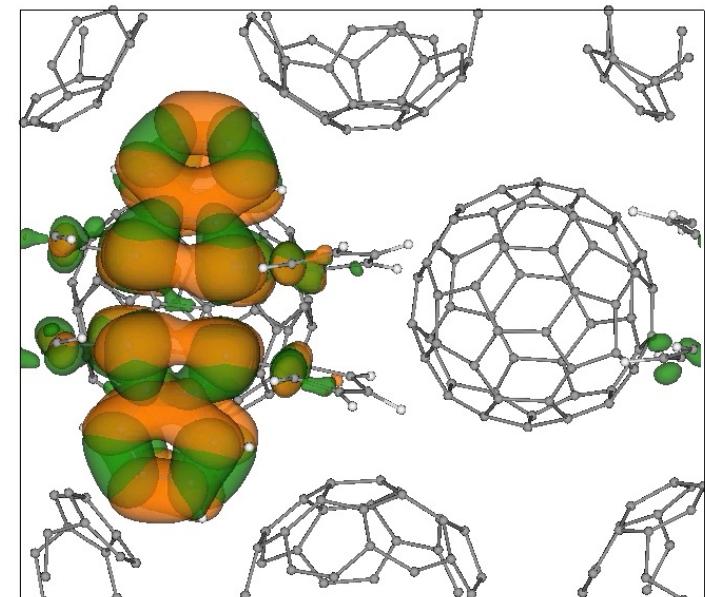
## Beyond Born-Oppenheimer



*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

# Attosecond Light-Matter Interaction

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

Attosecond =  $10^{-18}$  seconds

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"

- Attosecond physics could revolutionize information technology by enabling ultrafast computing & sensing devices

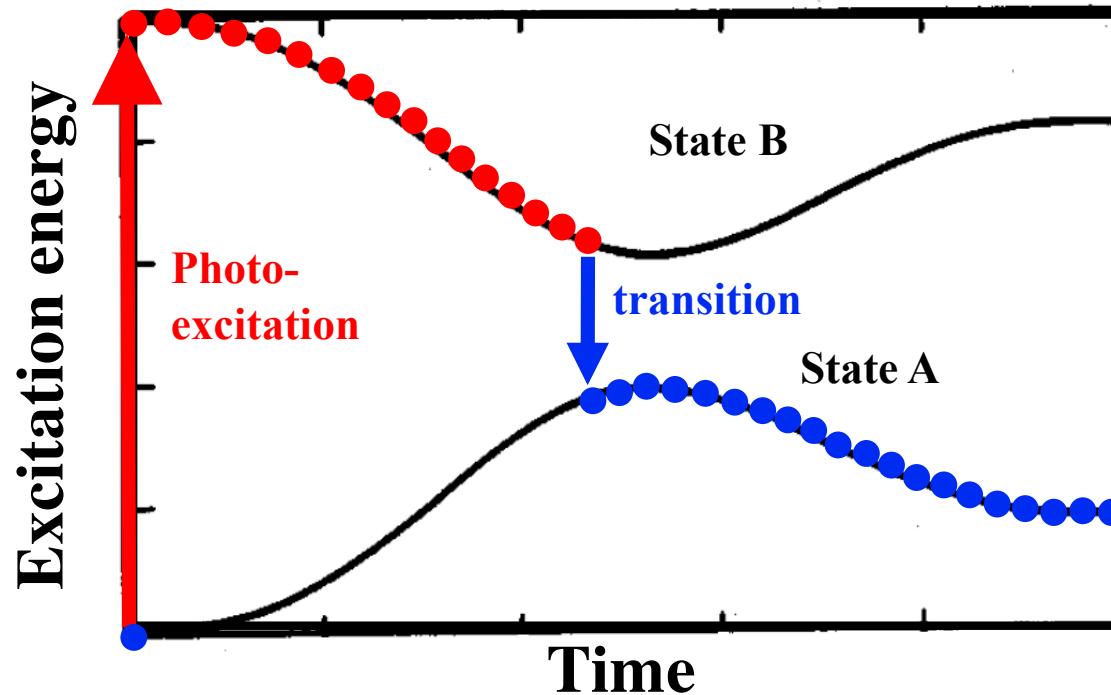
*cf. Petahertz electronics?* [Heide et al., Nat. Rev. Phys. 6, 648 ('24)]

$10^6 \times$  faster than current GHz CMOS technology

# 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

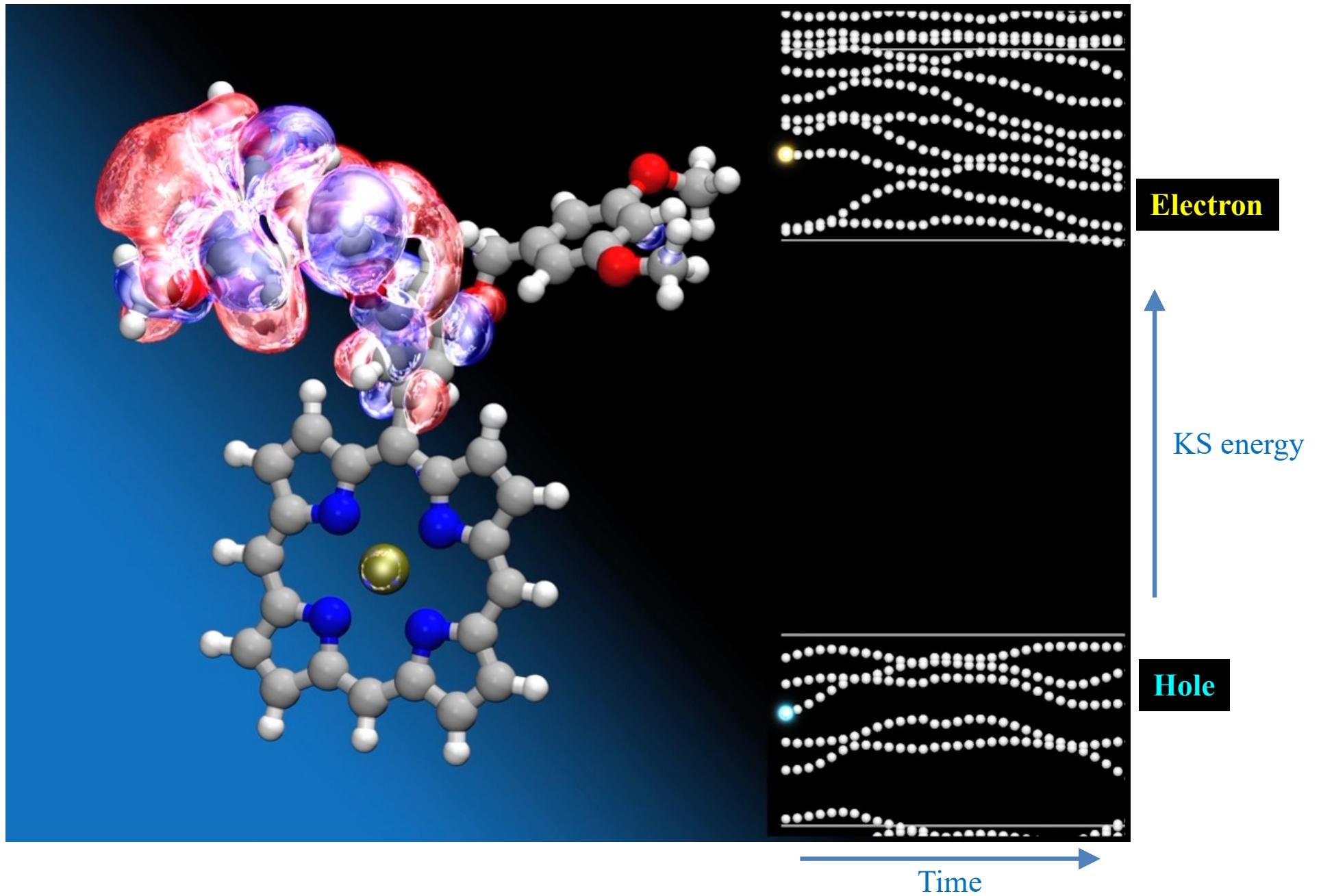
K-th excitation frequency

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

J-th adiabatic excited state

Electronic transition assisted by nuclei motion

# Surface-Hopping in Action



# **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)
- Other features:
  - > Various functionals: spin-polarized, GGA+U, DFT+D, nonlocal correlation
  - > Nudged elastic band (NEB) method for energy-barrier calculation
  - > Berry-phase computation of polarization

**Open-source software publication:** [Shimojo \*et al.\*, SoftwareX \*\*10\*\*, 100307 \('19\)](#)

Also use [VASP](#) & [Quantum Espresso](#)

# 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

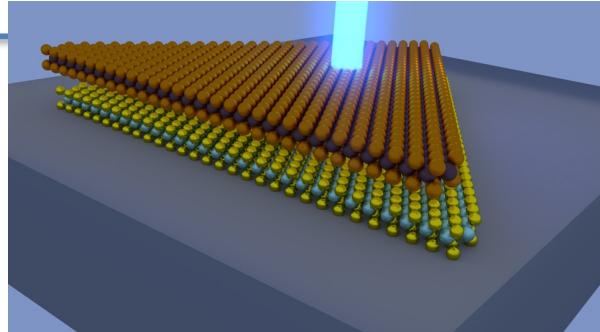
PI and Co-PIs: Aiichiro Nakano—PI, Rajiv K. Kalia, Ken-ichi Nomura, Priya Vasishta



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

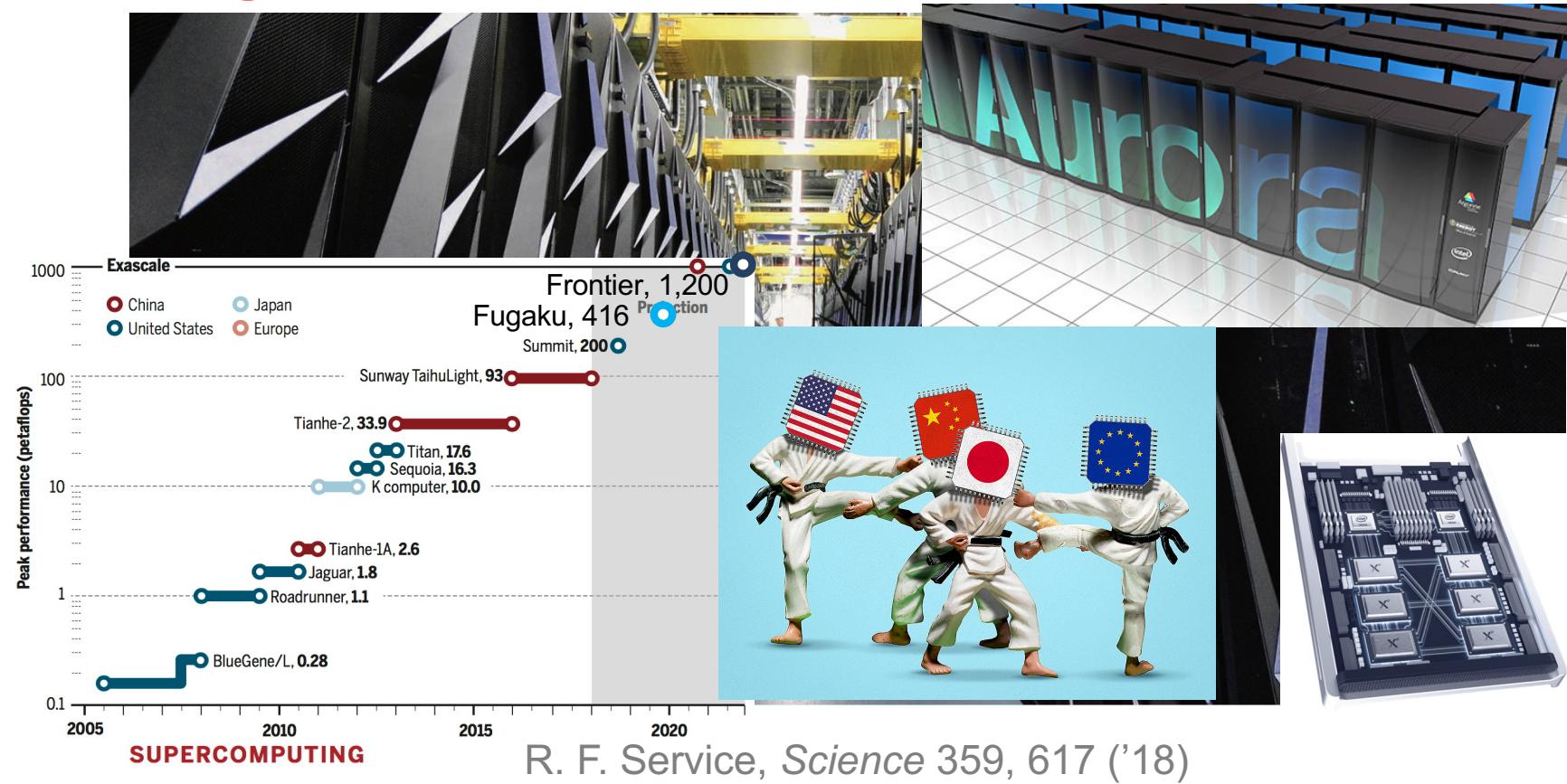


1.01 Exaflop/s  
Intel Aurora

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

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

# CACS@Aurora in the Global Exascale Race



## *Design for U.S. exascale computer takes shape*

Competition with China accelerates plans for next great leap in supercomputing power

Exa(peta)flop/s =  $10^{18}$  ( $10^{15}$ ) floating-point operations per second

By Robert F. Service

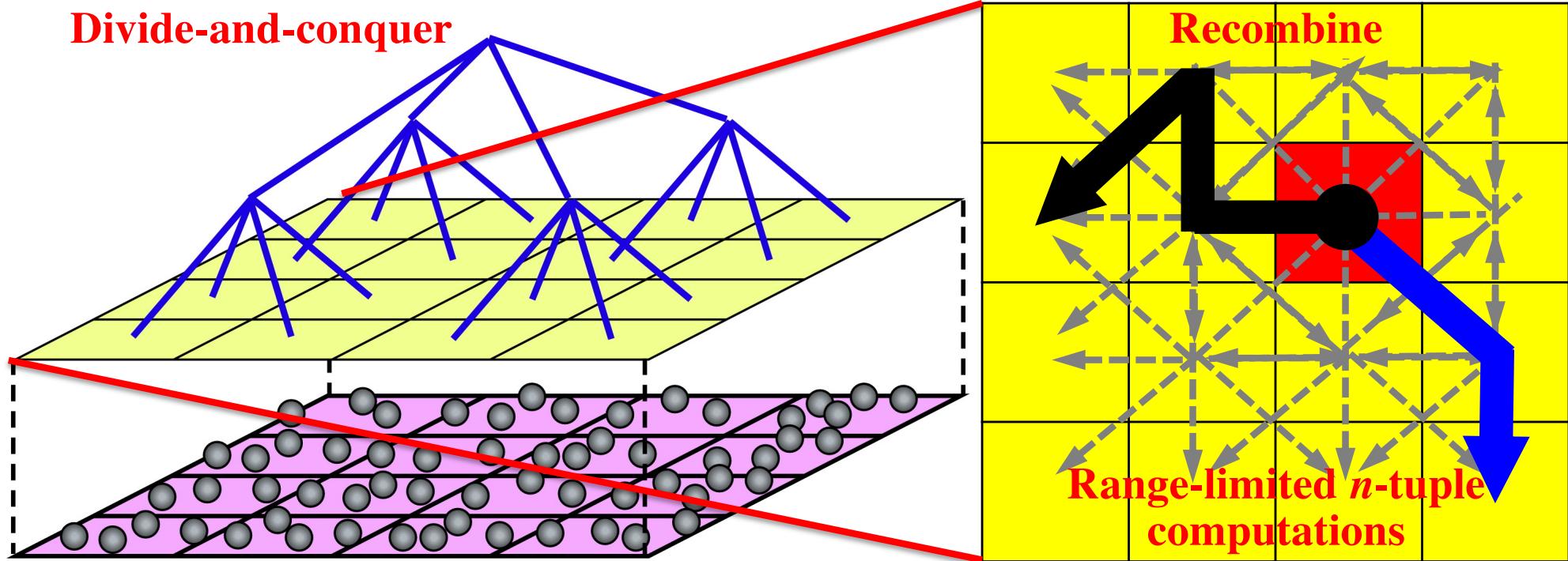
In 1957, the launch of the Sputnik satellite vaulted the Soviet Union to the lead in the space race and galvanized the United States. U.S. supercomputer researchers are today facing their own

Lemont, Illinois. That's 2 years earlier than planned. "It's a pretty exciting time," says Aiichiro Nakano, a physicist at the University of Southern California in Los Angeles who uses supercomputers to model materials made by layering stacks of atomic sheets like graphene.

pace reflects a change of strategy by DOE officials last fall. Initially, the agency set up a "two lanes" approach to overcoming the challenges of an exascale machine, in particular a potentially ravenous appetite for electricity that could require the output of a small nuclear plant.

<https://www.tomshardware.com/news/two-chinese-exascale-supercomputers>

# Divide-Conquer-Recombine (DCR) Engines



M. Kunaseth et al., ACM/IEEE SC13

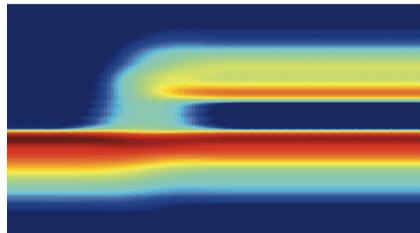
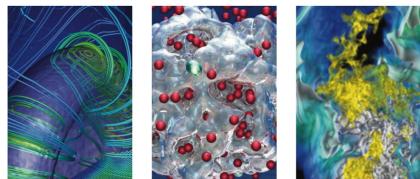
- **Lean divide-&-conquer density functional theory (LDC-DFT) algorithm minimizes the prefactor of  $O(N)$  computational cost**

F. Shimojo et al., *J. Chem. Phys.* **140**, 18A529 ('14); S. Tiwari et al., *HPCAsia20 Best Paper*

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

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

# 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

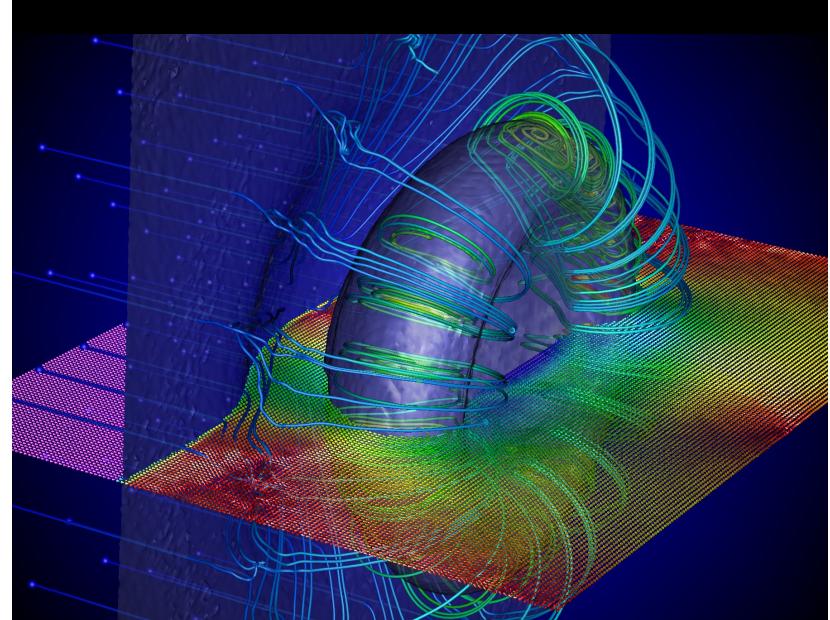
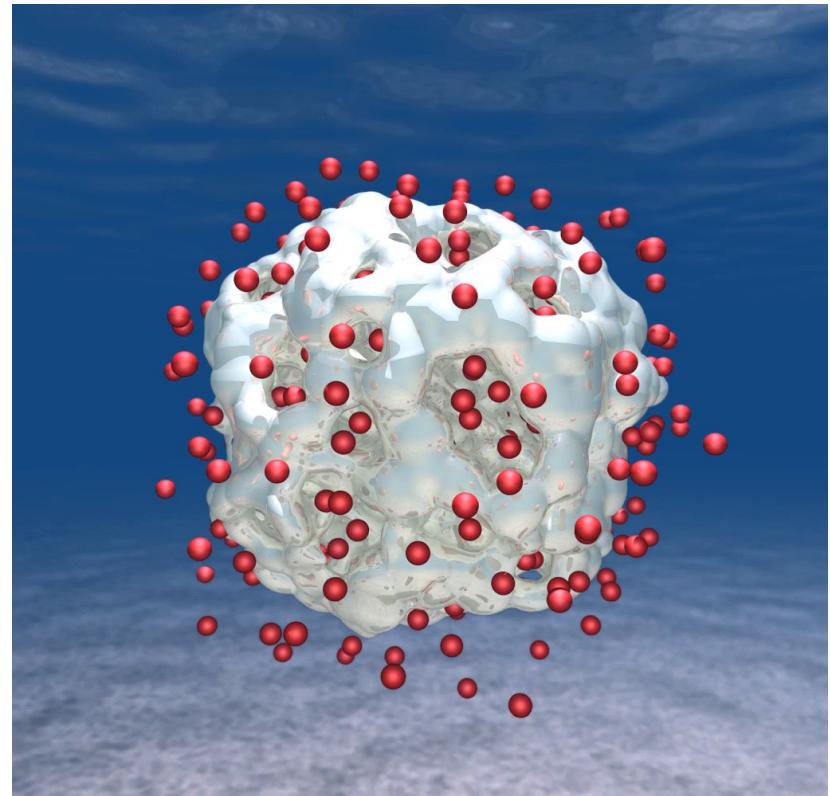
Shimamura *et al.*,  
*Nano Lett.*  
14, 4090 ('14)

*On-demand hydrogen  
production from water*

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

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

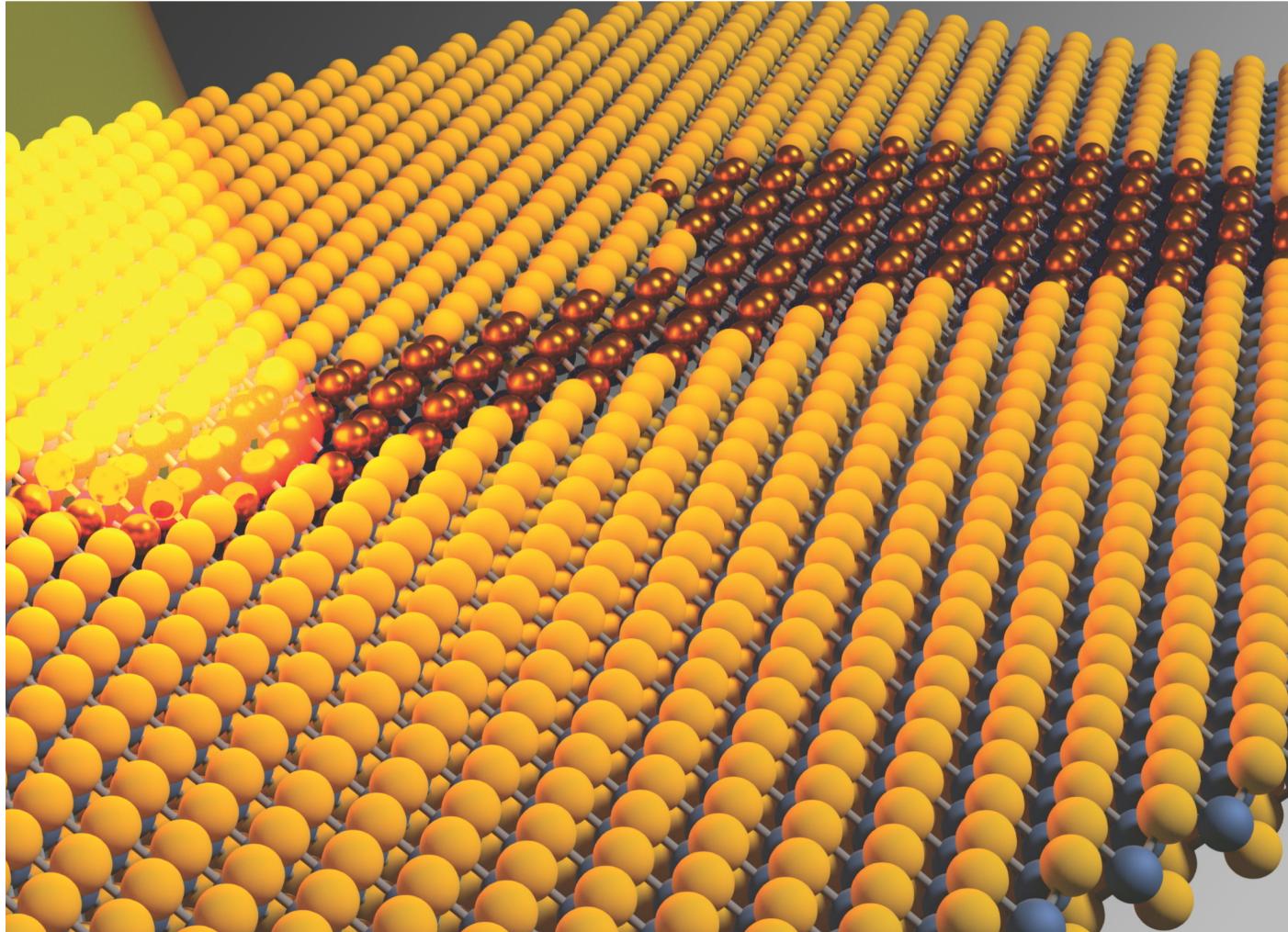
*Fluid dynamics  
atom-by-atom*



# Ultrafast Control of Materials

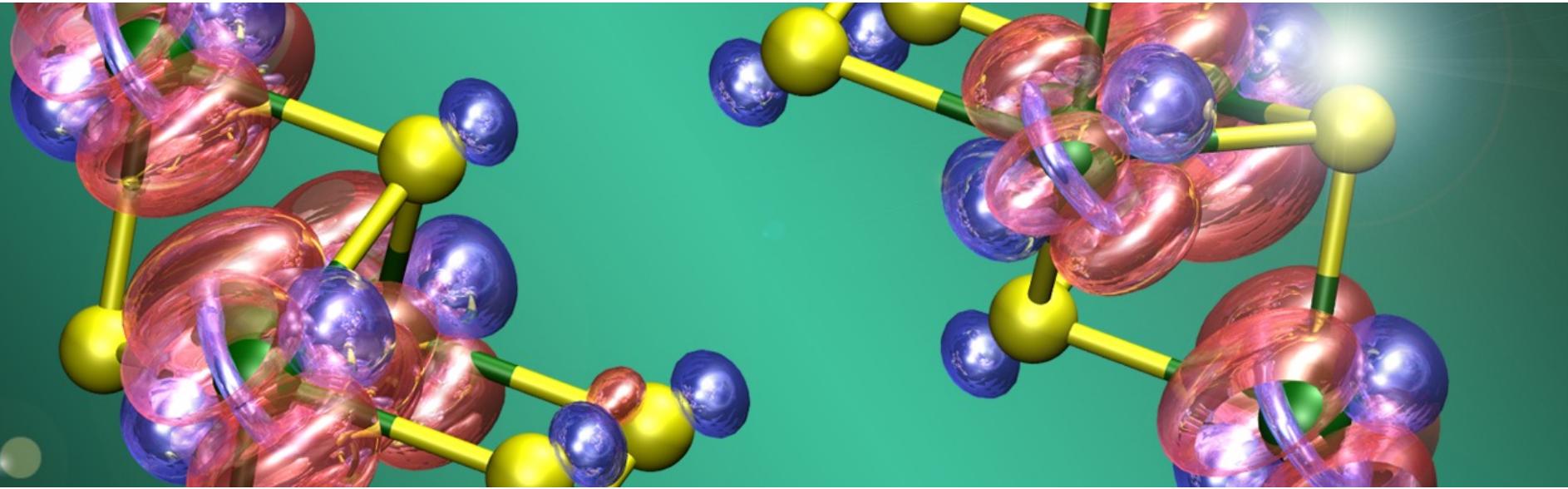
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**Goal:** Use ultrafast laser pulses to transform material structures & properties  
(e.g., semiconductor-to-metal) on demand

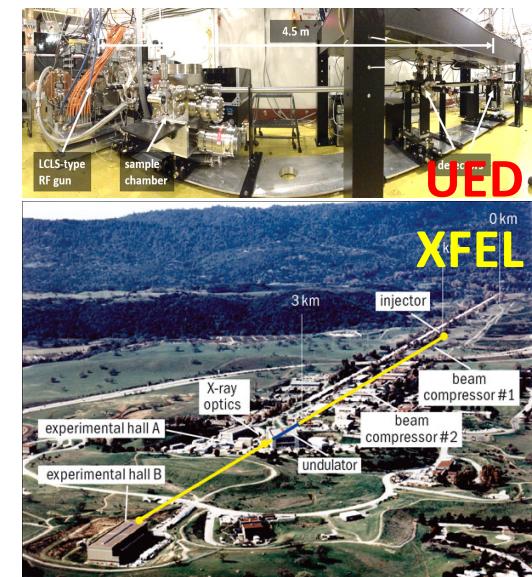


A. Krishnamoorthy *et al.*, *Nanoscale* **10**, 2742 ('18); journal cover

# 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.

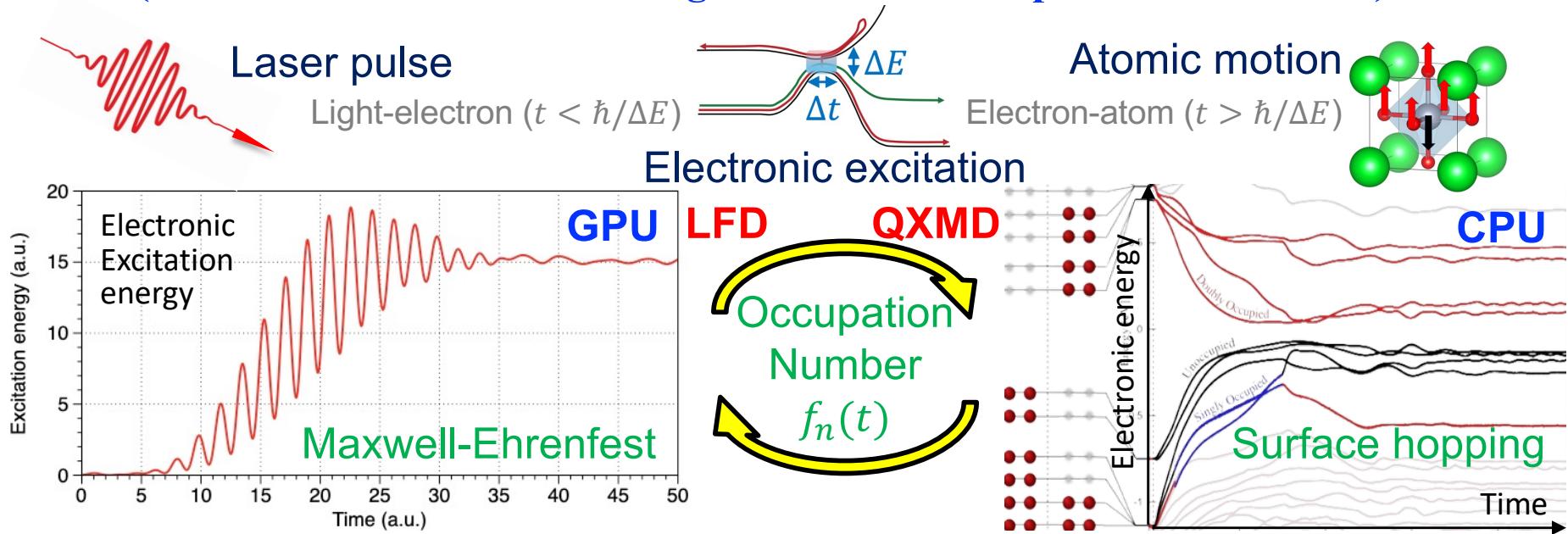


**Ultrafast electron diffraction:** M.F. Lin *et al.*, *Nature Commun.* 8, 1745 ('17)  
**X-ray free-electron laser:** I. Tung *et al.*, *Nature Photon.* 13, 425 ('19)

# Nonadiabatic Quantum MD: DC-MESH

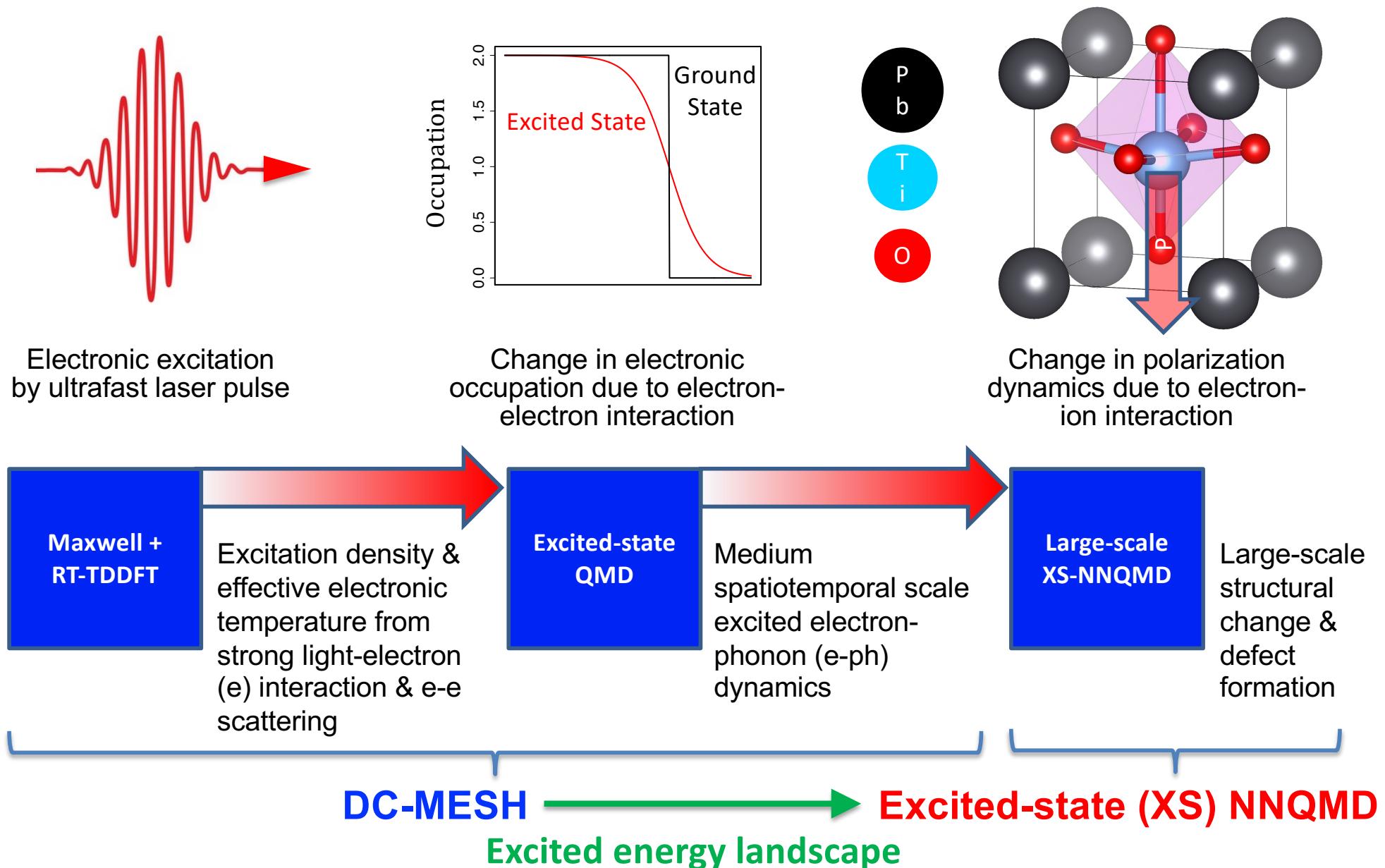
- DC-MESH (divide-&-conquer Maxwell + Ehrenfest + surface-hopping):  $O(N)$  algorithm to simulate photo-induced quantum materials dynamics
- LFD (local field dynamics): Maxwell equations for light & real-time time-dependent density functional theory equations for electrons to describe light-matter interaction
- 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) yet locally-dense (intradomain nonlocal exchange-correlation computation *via* BLAS) solver

Lam et al., *Nature Commun.* 15, 3479 ('24)



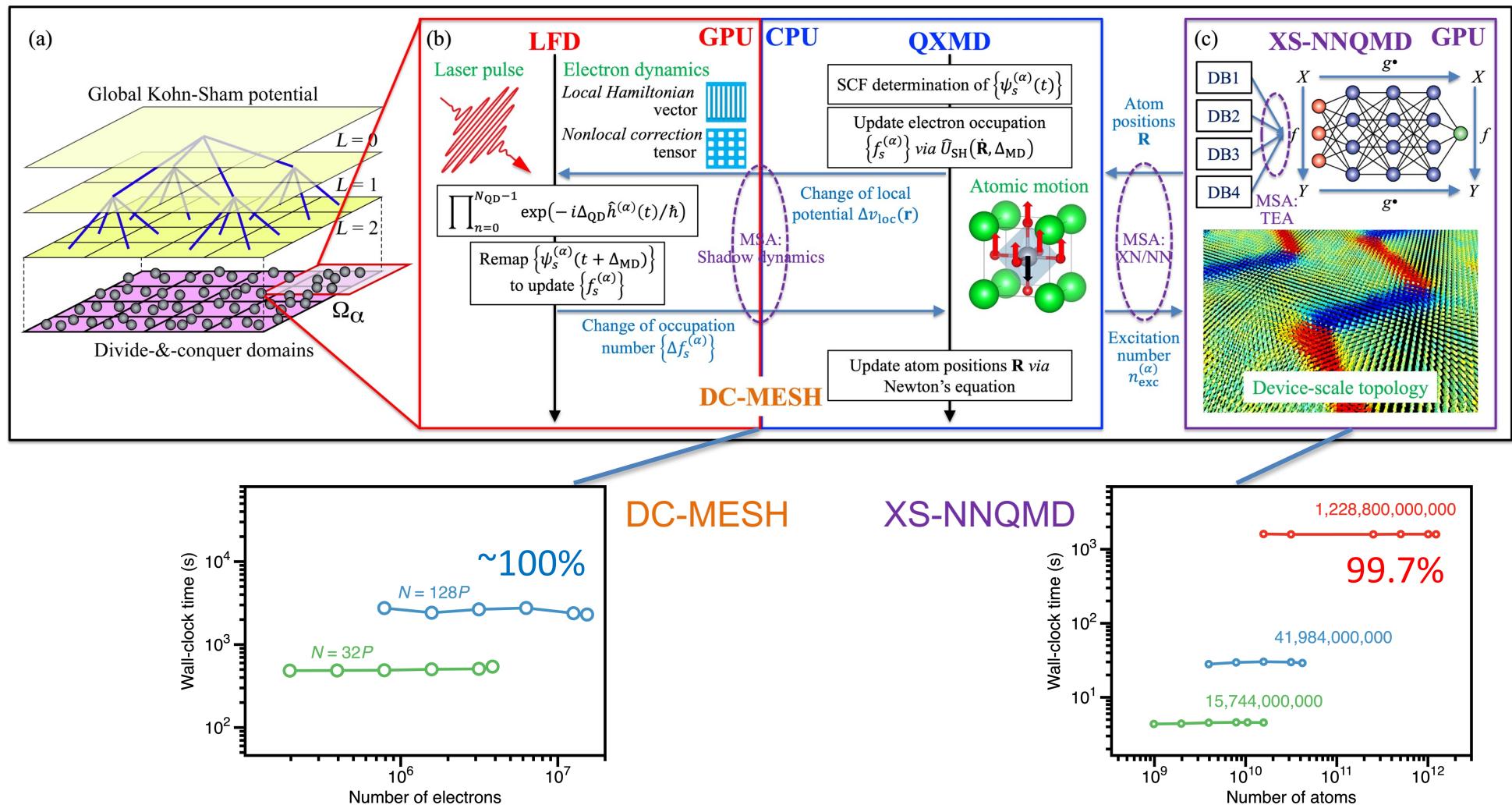
Linker et al., *Science Adv.* 8, eabk2625 ('22); Razakh et al., *IEEE-PDSEC* ('24)

# Multiscale DC-MESH – XS-NNQMD

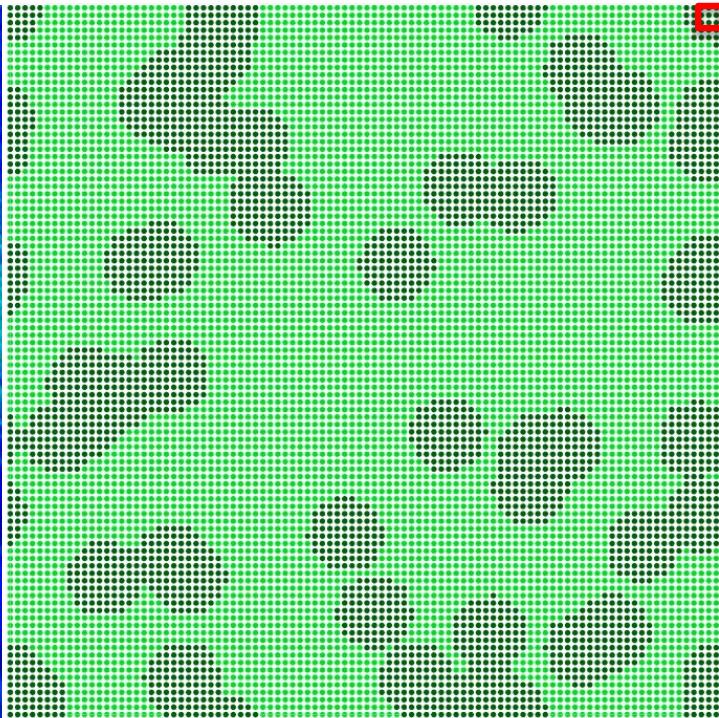
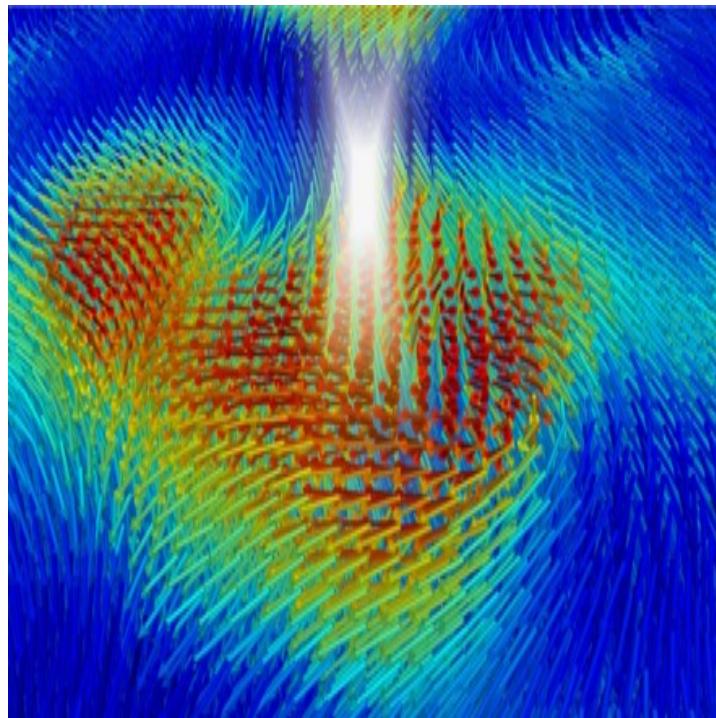


# Breaking Exaflop/s Barrier

- On 60,000 GPUs of Aurora: **1.87 Exaflop/s**;  **$152\times$**  &  **$3,780\times$**  improvements of time-to-solution compared with state-of-the-art for 15.4M-electron DC-MESH & 1.2T-atom XS-NNQMD; near-perfect parallel efficiency



# Application: Ferroelectric Opto-Toptronics



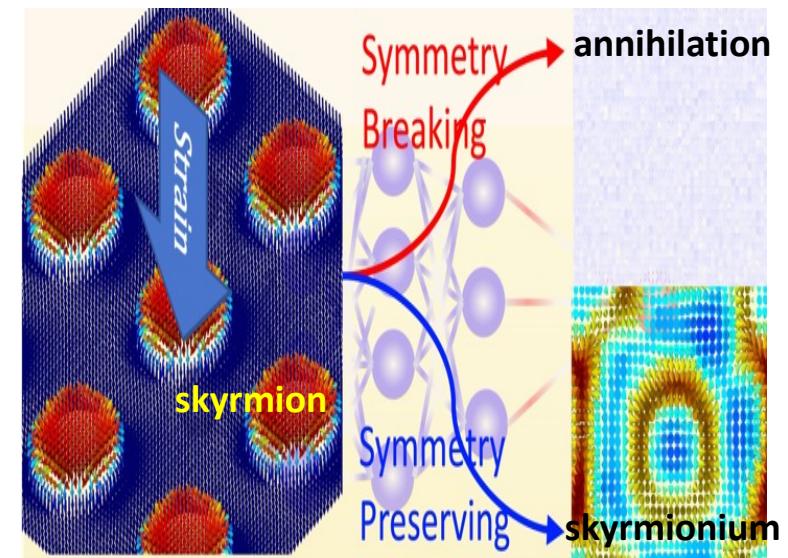
System size  
simulated  
with  
NAQMD

Large-scale  
structure  
simulated  
with  
NNQMD

- 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

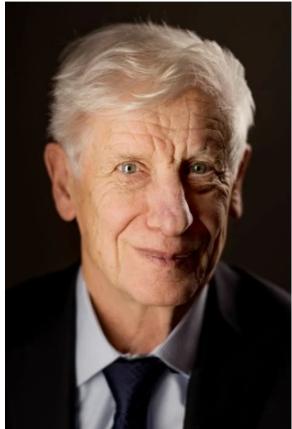
\*Composite of skyrmions with opposite topological charges

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

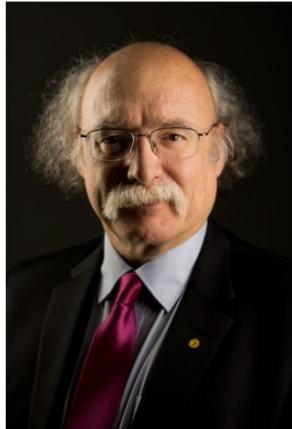


# Topological Quantum Matter

Nobel Prize in Physics 2016



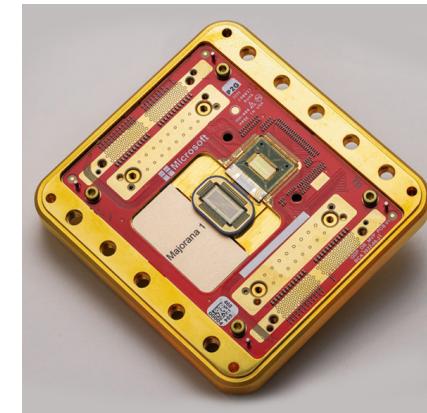
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David J. Thouless  
Prize share: 1/2



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F. Duncan M. Haldane  
Prize share: 1/4



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J. Michael Kosterlitz  
Prize share: 1/4



[News](#) • February 19, 2025 • 7 min read

## Microsoft unveils Majorana 1, the world's first quantum processor powered by topological qubits

*Nature* 638, 651 ('25)

- Topological quantum matter holds key to sustainable society with ubiquitous & power-hungry AI, by enabling ultralow-power transistors & robust quantum computers

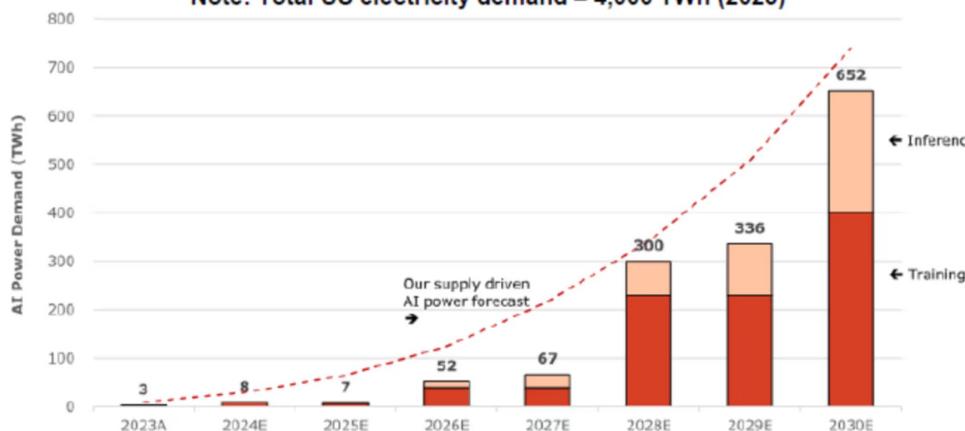
*cf. AttoJoule-switching logic?* [Datta et al., *Science* 378, 733 ('22)]

# Who Will Power Your Future AI?

## Summary of GenAI demand forecast

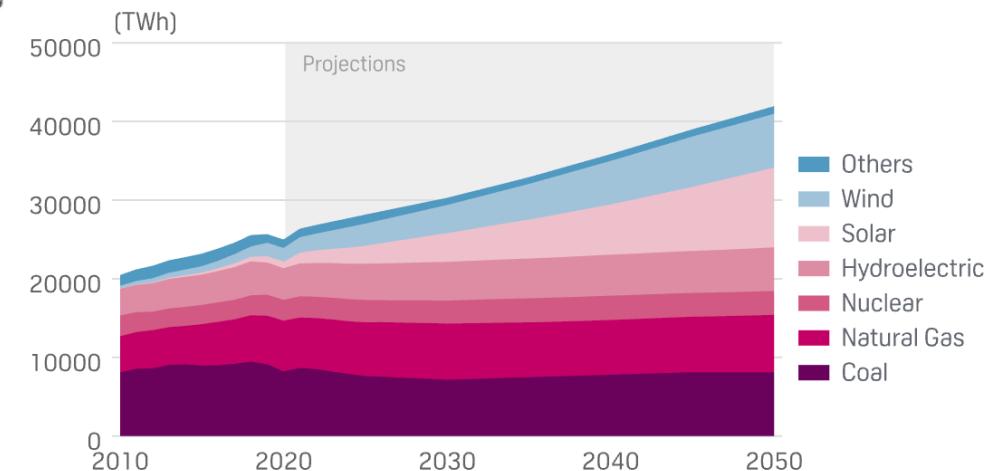
Source: Wells Fargo

Note: Total US electricity demand – 4,000 TWh (2023)



- Exponential growth of AI energy demand

## GLOBAL NET ELECTRICITY GENERATION BY SOURCE



Source: EIA's International Energy Outlook 2021

- Linear growth of global energy supply (don't forget CO<sub>2</sub> emission)

2025 Energy Transition Summit  
USC Ershaghi Center for Energy Transition

<https://ecet.usc.edu/energy-transition-summit-2025>



**USC E-CET**  
Ershaghi Center for Energy Transition

# Where to Go from Here

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Detailed lecture notes are available at a USC course home page

## EXTREME-SCALE QUANTUM SIMULATIONS

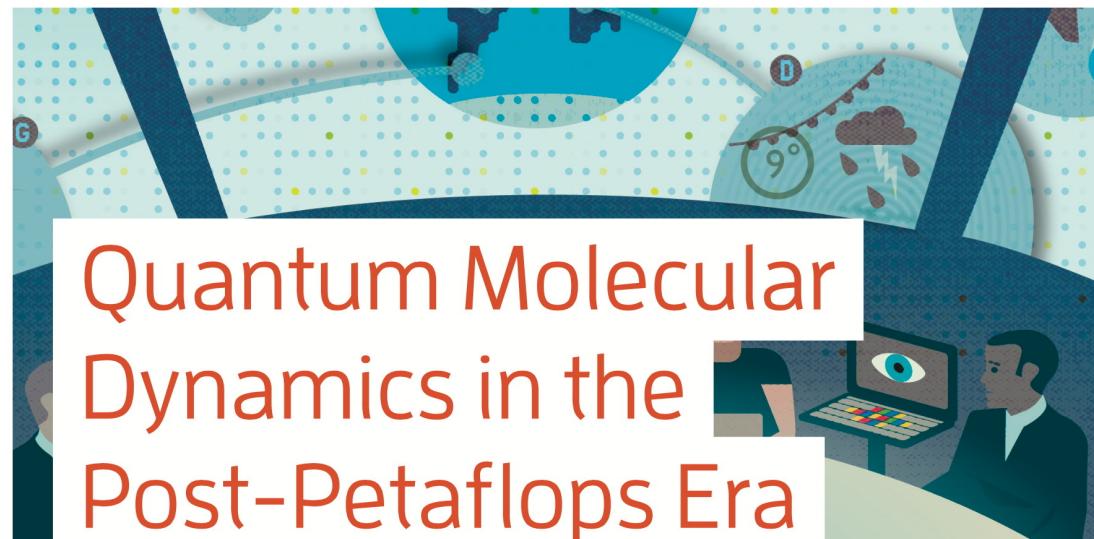
This course surveys & projects algorithmic & computing technologies that will make quantum-dynamics simulations metascalable, *i.e.*, "design once, continue to scale on future computer architectures".

<https://aiichironakano.github.io/phys760-lecture.html>

See also N. Romero *et al.*, *IEEE Computer* **48(11)**, 33 ('15)

<https://aiichironakano.github.io/phys516/Romero-QMD-IEEEComputer15.pdf>

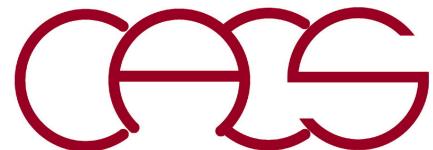
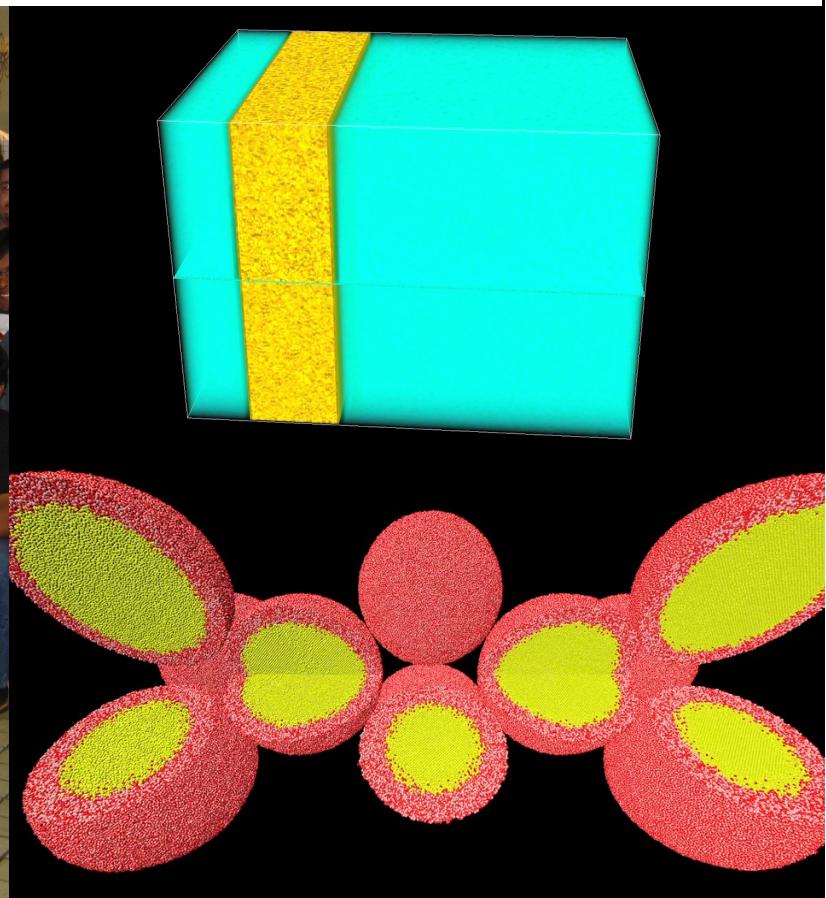
COVER FEATURE GRAND CHALLENGES IN SCIENTIFIC COMPUTING



Standard textbook: R. Martin, [\*Electronic Structure\*](#) (Cambridge Univ. Press, '20)

# Conclusion

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



Supported by the National Science Foundation,  
CyberTraining Award OAC-2118061

