

Hybrid Particle-Continuum Simulation

Aiichiro Nakano

Collaboratory for Advanced Computing & Simulations

Department of Computer Science

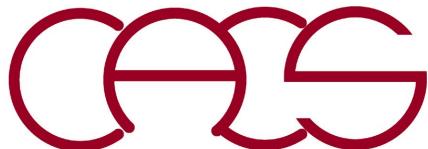
Department of Physics & Astronomy

Department of Quantitative & Computational Biology

University of Southern California

Email: anakano@usc.edu

Adaptively manage accuracy-cost trade-off;
coarse-graining by heuristics
(*i.e.*, switching to different abstract)



Multiscale Modeling

The Nobel Prize in Chemistry 2013



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Martin Karplus



Photo: Keilana via
Wikimedia Commons
Michael Levitt

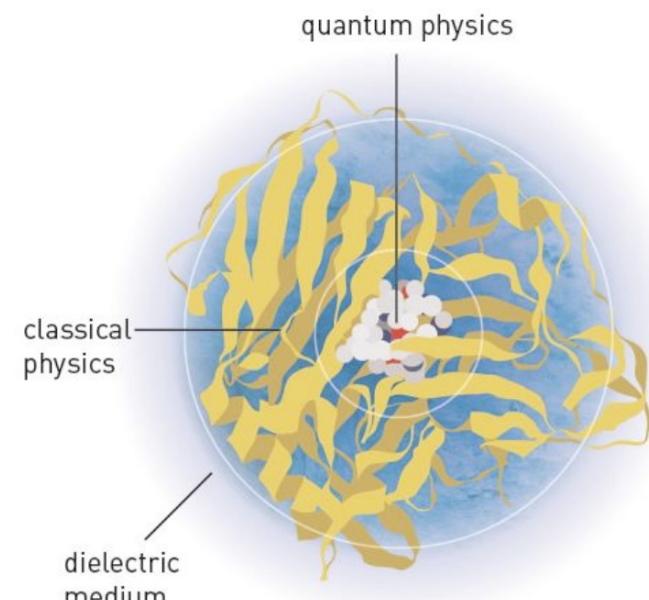


Photo: Wikimedia
Commons
Arieh Warshel



The Nobel Prize in Chemistry 2013 was awarded jointly to Martin Karplus, Michael Levitt and Arieh Warshel *"for the development of multiscale models for complex chemical systems"*.

- A. Warshel & M. Karplus, *J. Am. Chem. Soc.* **94**, 5612 ('72)
A. Warshel & M. Levitt, *J. Mol. Biol.* **103**, 227 ('76)



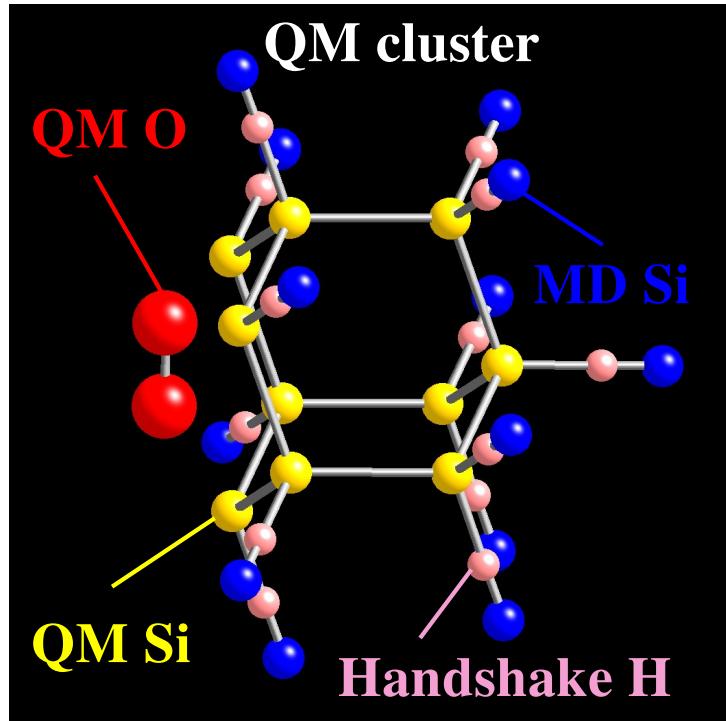
**QM/MM:
quantum-
mechanical/molecular-
mechanical modeling**

Find multiscale modeling in your area!

See Arieh's interview: <https://www.nature.com/articles/s43588-022-00285-2>

Adaptive Multiscale Dynamics

QMD

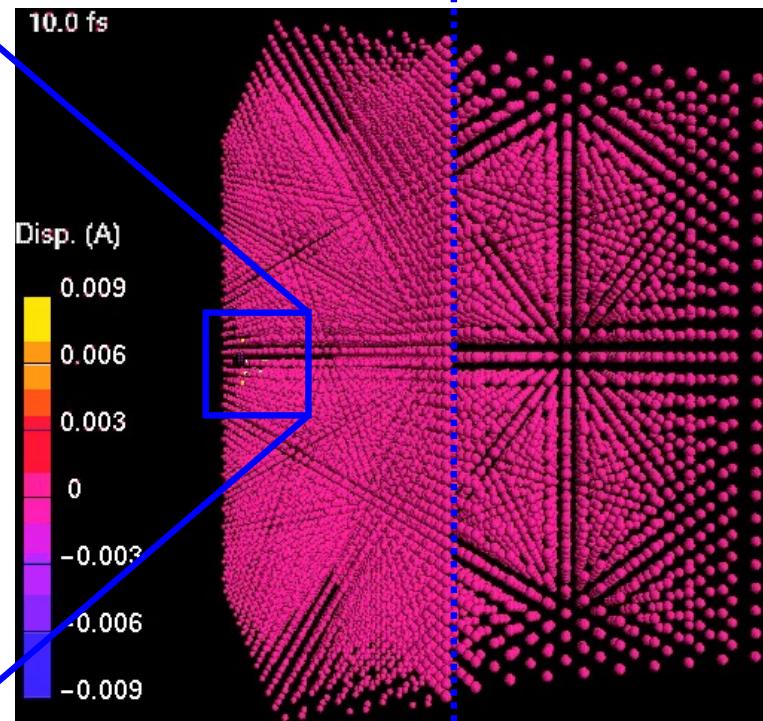


Oxidation of Si

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

L. Lidorikis *et al.*, *Phys. Rev. Lett.* 87, 086104 ('01)

MD



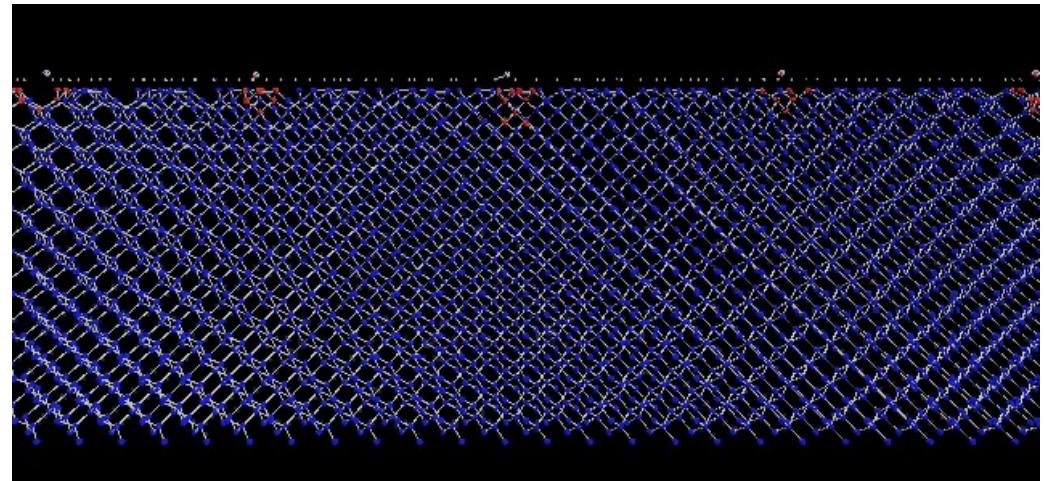
FED



High-energy
beam oxidation
of Si (SIMOX)

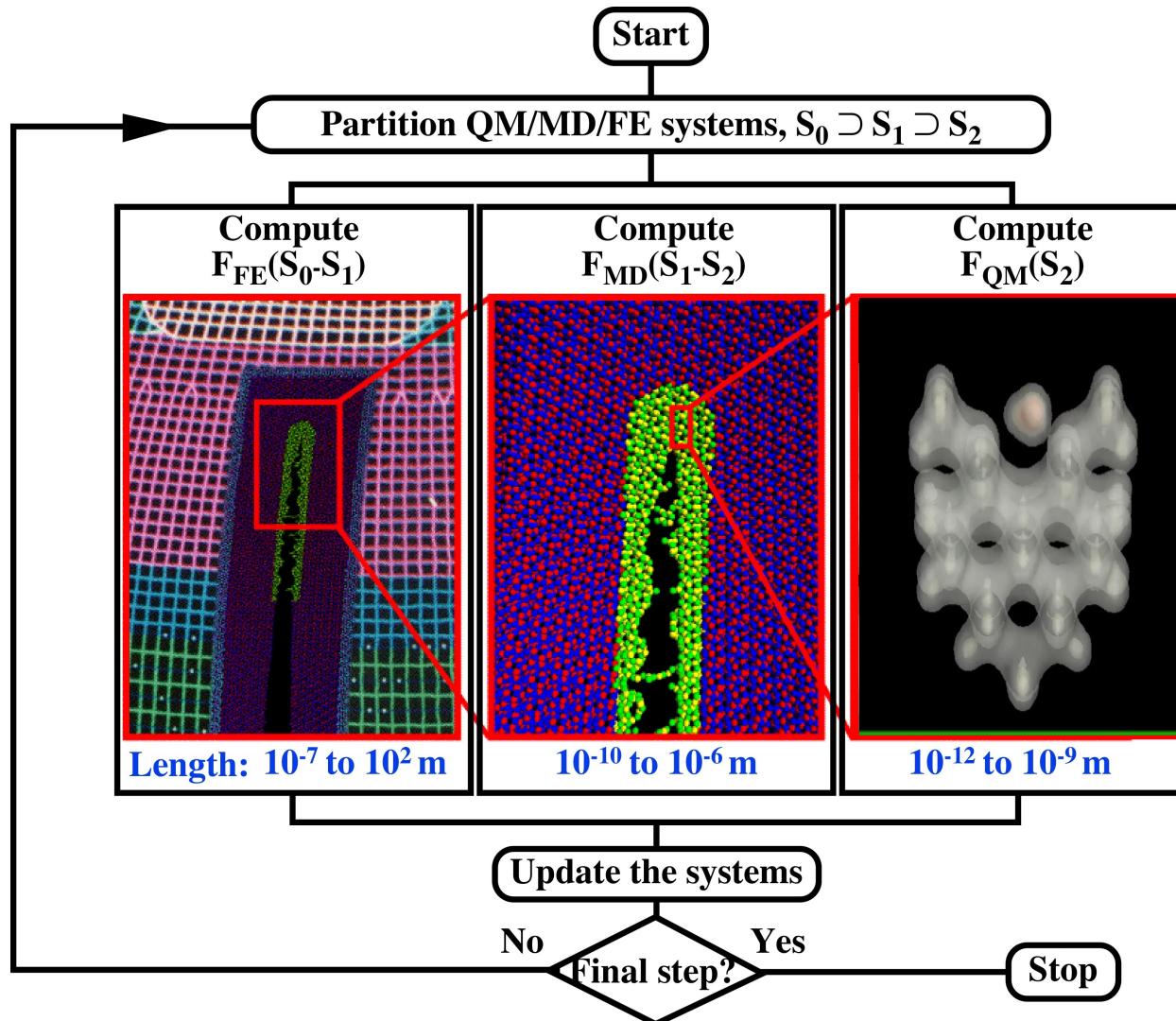
H. Takemiya *et al.*,
IEEE/ACM Supercomputing (SC06)

QMD/MD/FED:
quantum molecular dynamics/
molecular dynamics/
finite-element dynamics simulation



Multiscale FED/MD/QMD Simulation

- Embed high-accuracy computations only when & where needed
- Train coarse simulations by fine simulations



Multiscale simulation to seamlessly couple:

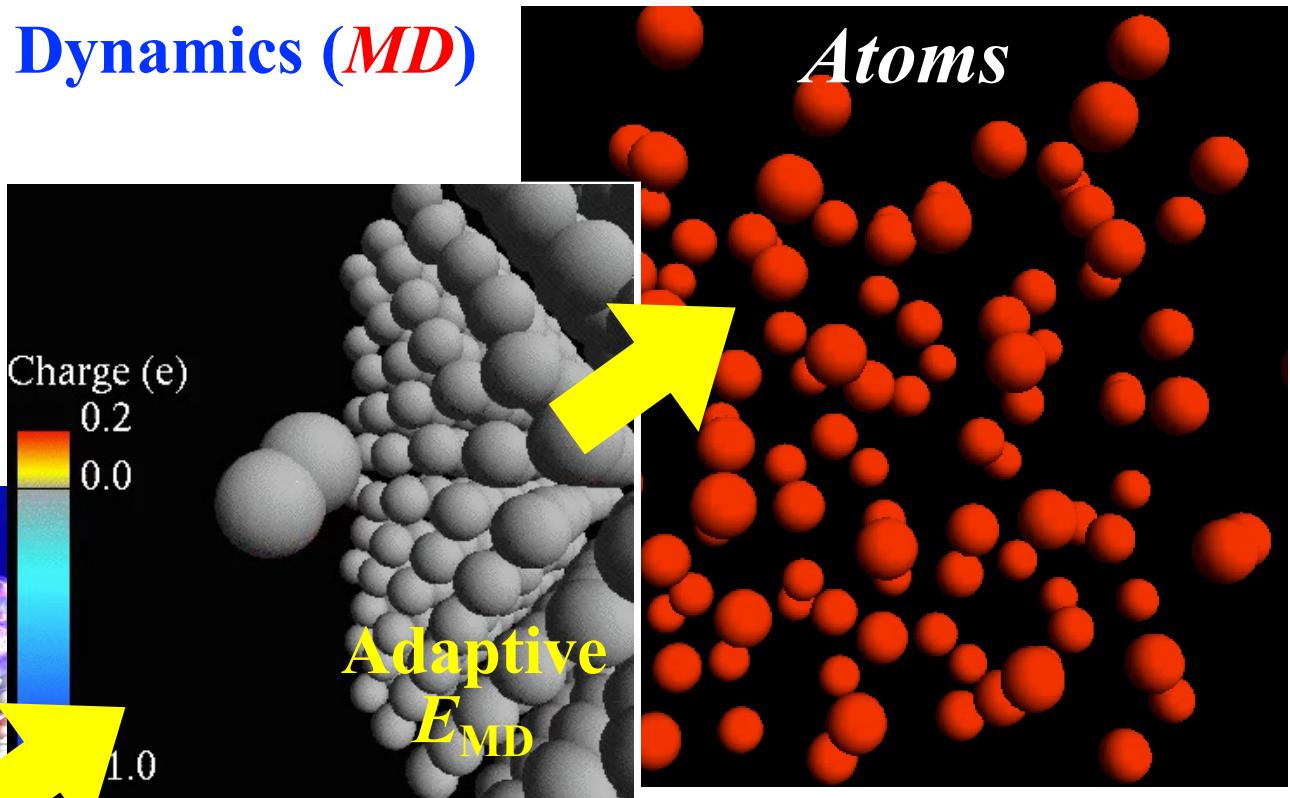
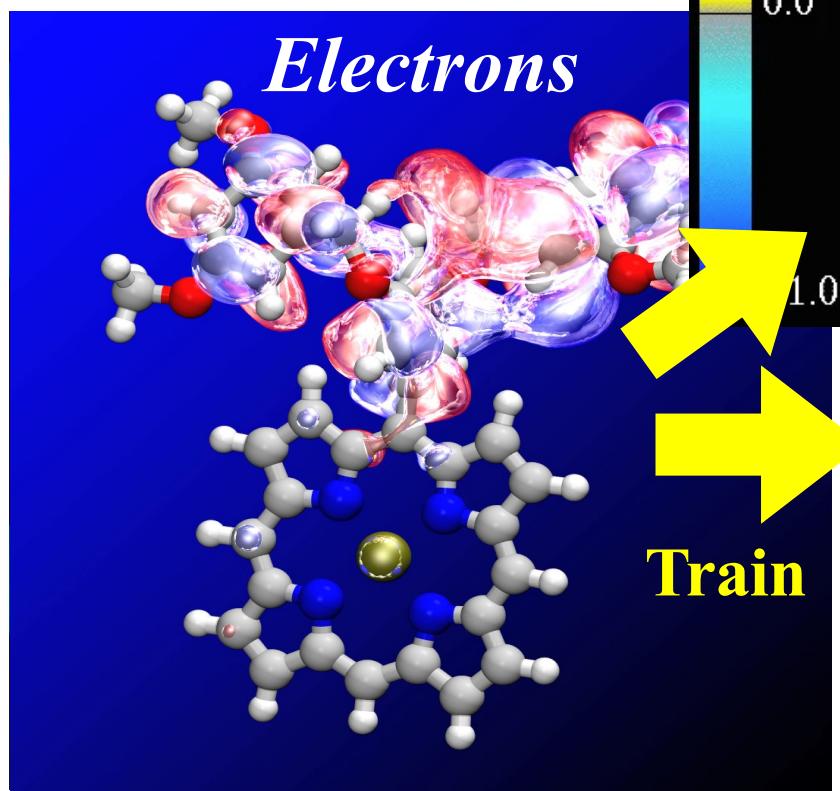
- Finite element (FE) dynamics based on continuum elasticity
- Atomistic molecular dynamics (MD) simulation
- Quantum molecular dynamics (QMD) based on the density functional theory (DFT)

Molecular Dynamics & Machine Learning

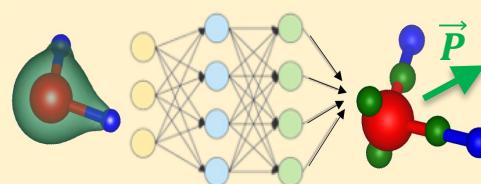
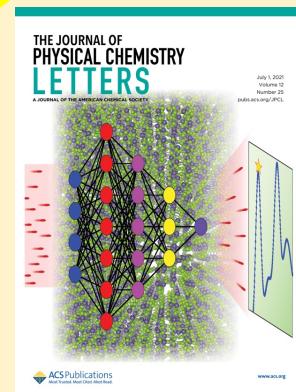
Molecular Dynamics (*MD*)

Reactive MD (*RMD*)

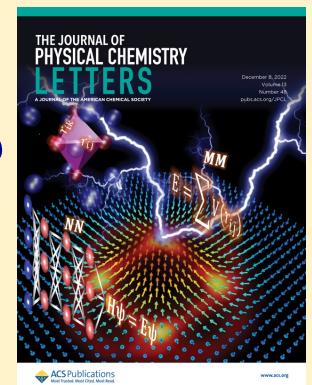
Nonadiabatic quantum MD (*NAQMD*)



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

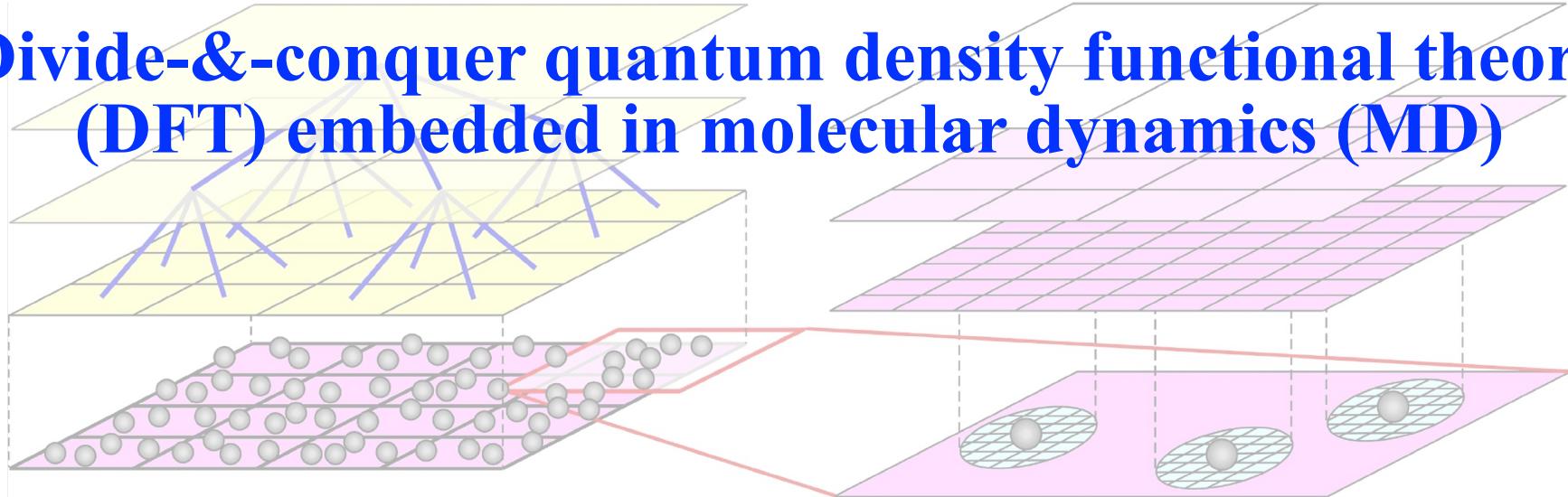


Physical Review Letters
Editor's choice
(May 25, '21)

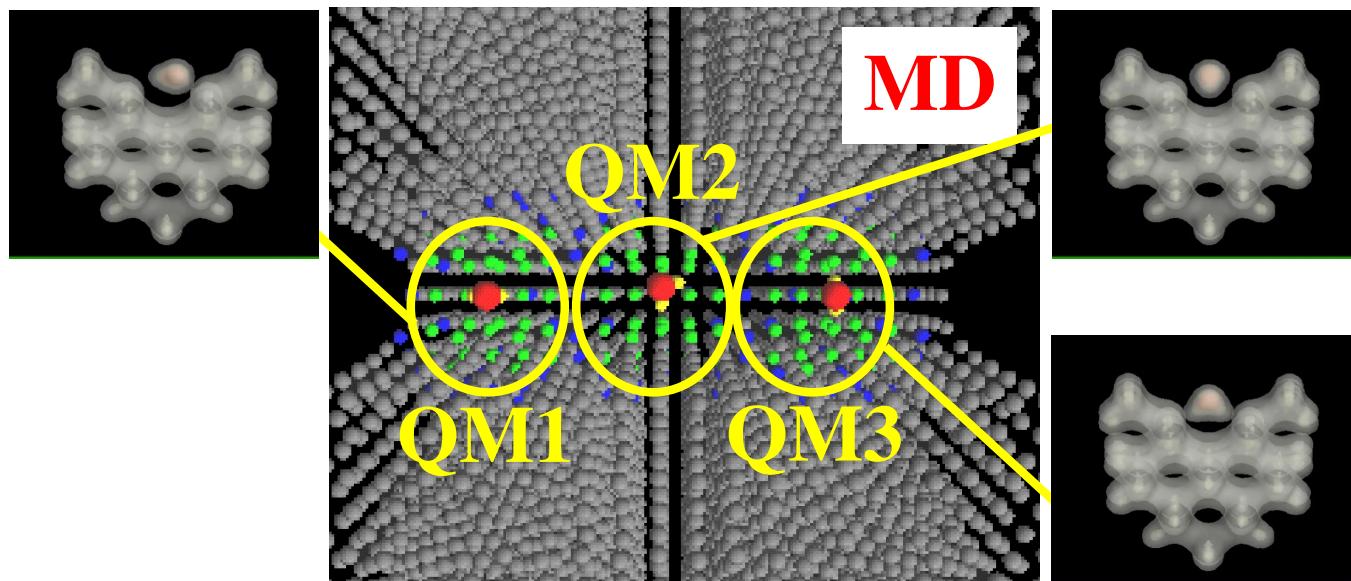


DC Multiscale MD/QM (DFT)

Divide-&-conquer quantum density functional theory (DFT) embedded in molecular dynamics (MD)



$$E_{\text{MD/QM}} = E_{\text{MD}}^{\text{system}} + \sum_{\text{cluster}} [E_{\text{QM}}^{\text{cluster}}(\{\mathbf{r}_{\text{QM}}\}, \{\mathbf{r}_{\text{HS}}\}) - E_{\text{MD}}^{\text{cluster}}(\{\mathbf{r}_{\text{QM}}\}, \{\mathbf{r}_{\text{HS}}\})]$$

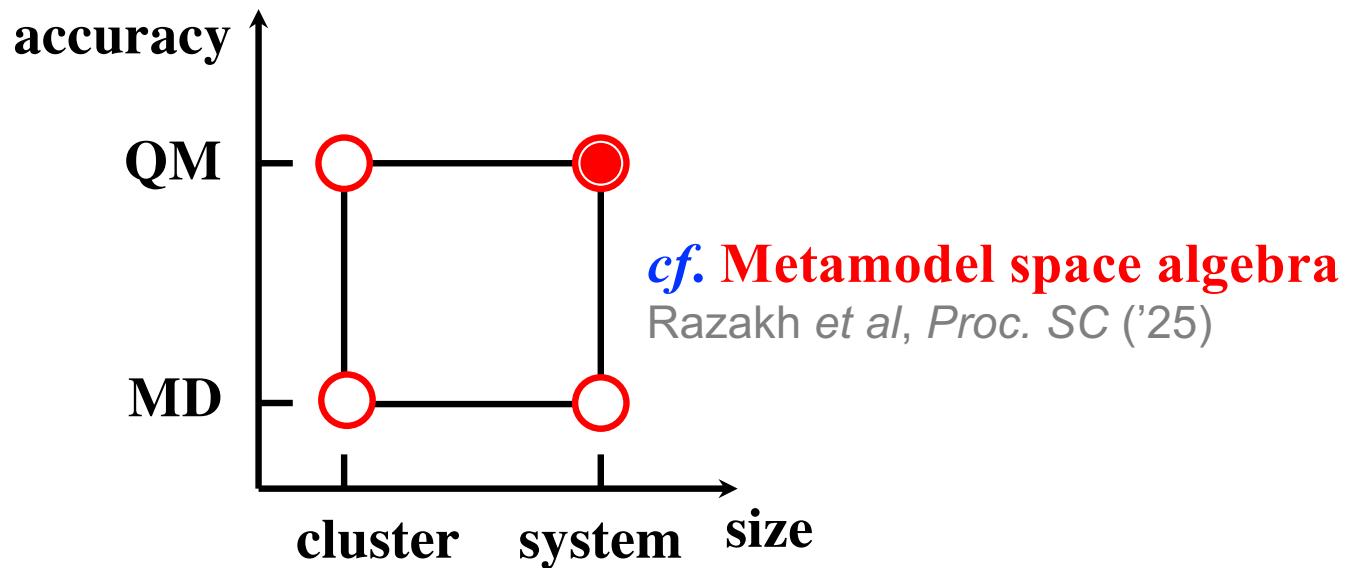


Additive Hybridization

Additive hybridization

Morokuma et al., *J. Mol. Struct.* 461-462, 1 ('99)

- Extrapolation in 2D meta-model space (accuracy vs. size)
- Resulting in linear combination of MD & QM energies
- Modular
 - Reuse of existing MD & QM (density functional theory) codes
 - Minimal inter-model dependence/communication

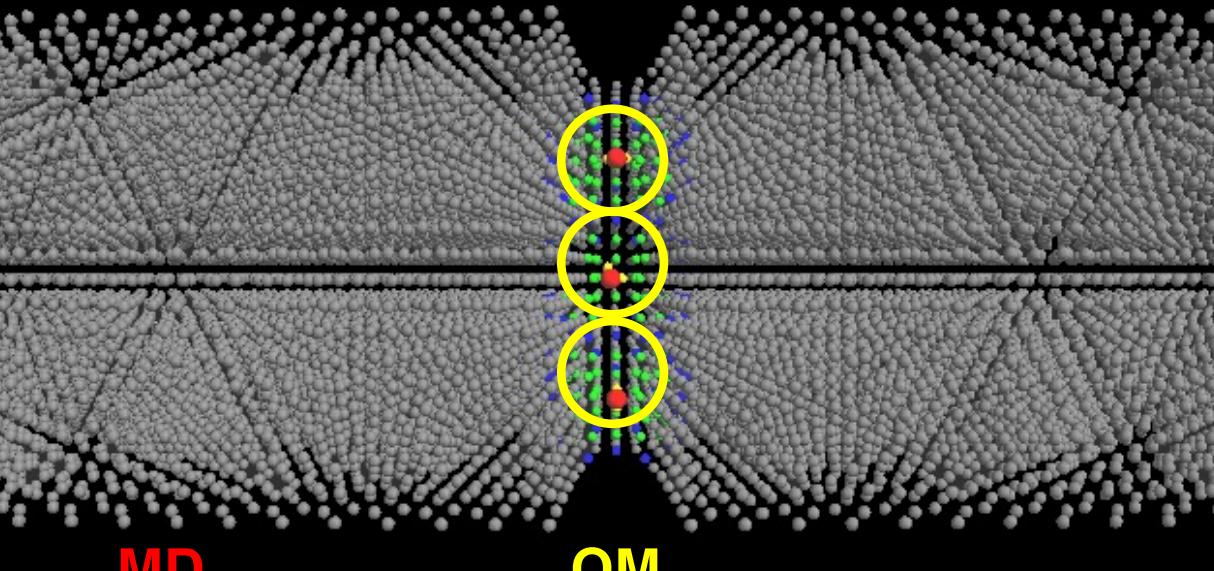


$$E_{\text{QM}}(s) - E_{\text{QM}}(c) \cong E_{\text{MD}}(s) - E_{\text{MD}}(c)$$

$$\therefore E_{\text{QM}}(s) = E_{\text{MD}}(s) + [E_{\text{QM}}(c) - E_{\text{MD}}(c)] = E_{\text{MD}}(s) + \delta E_{\text{QM}/\text{MD}}(c)$$

Environmental Effect on Fracture

Reaction of H₂O molecules at a Si crack tip

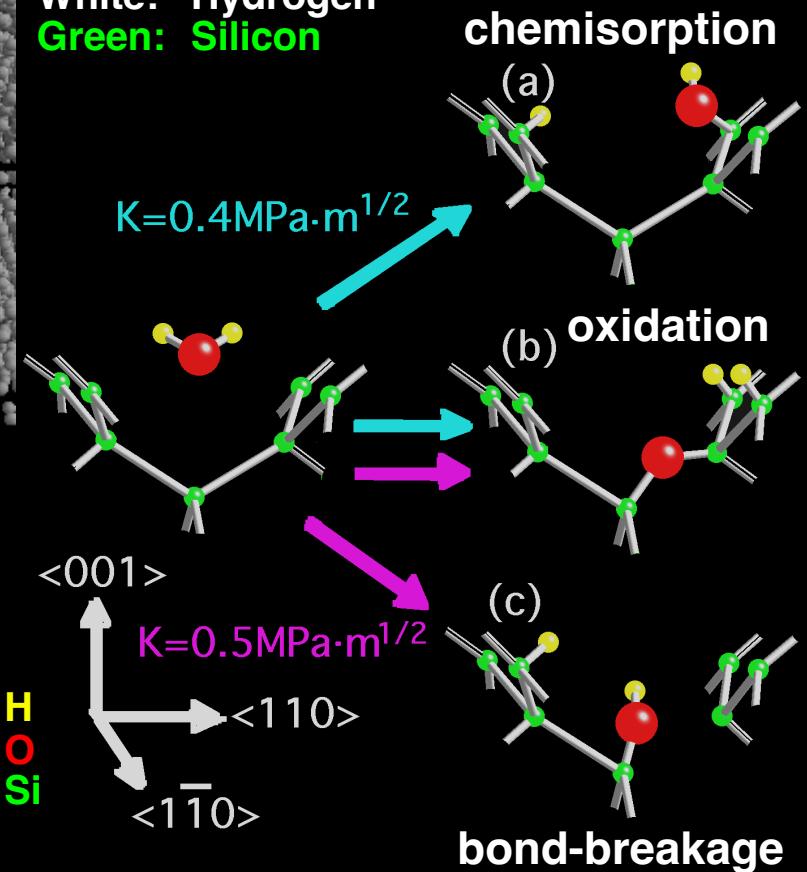


MD

QM

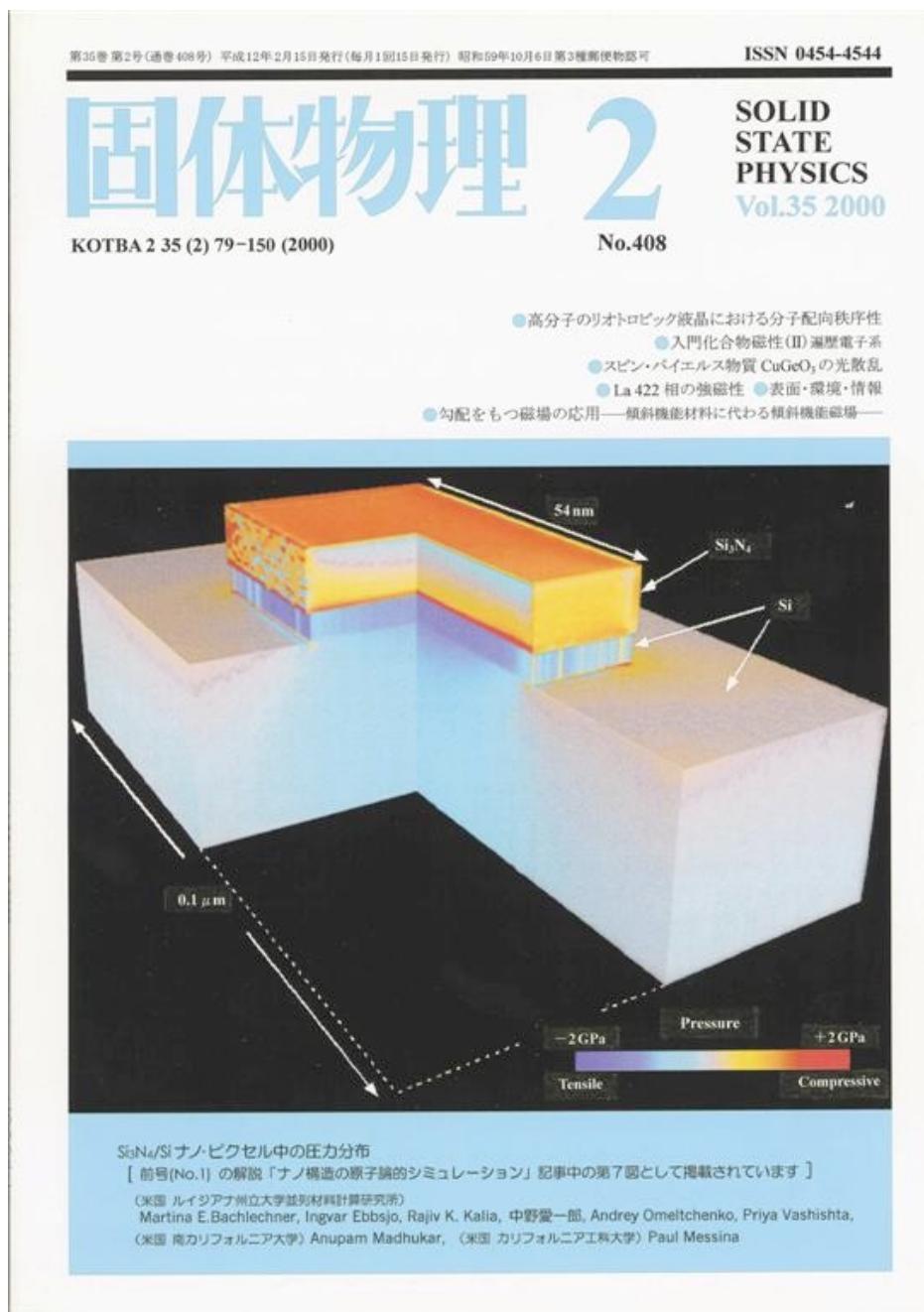
Collaborators: S. Ogata (NIT),
F. Shimojo (Kumamoto)

Blue: Oxygen
White: Hydrogen
Green: Silicon



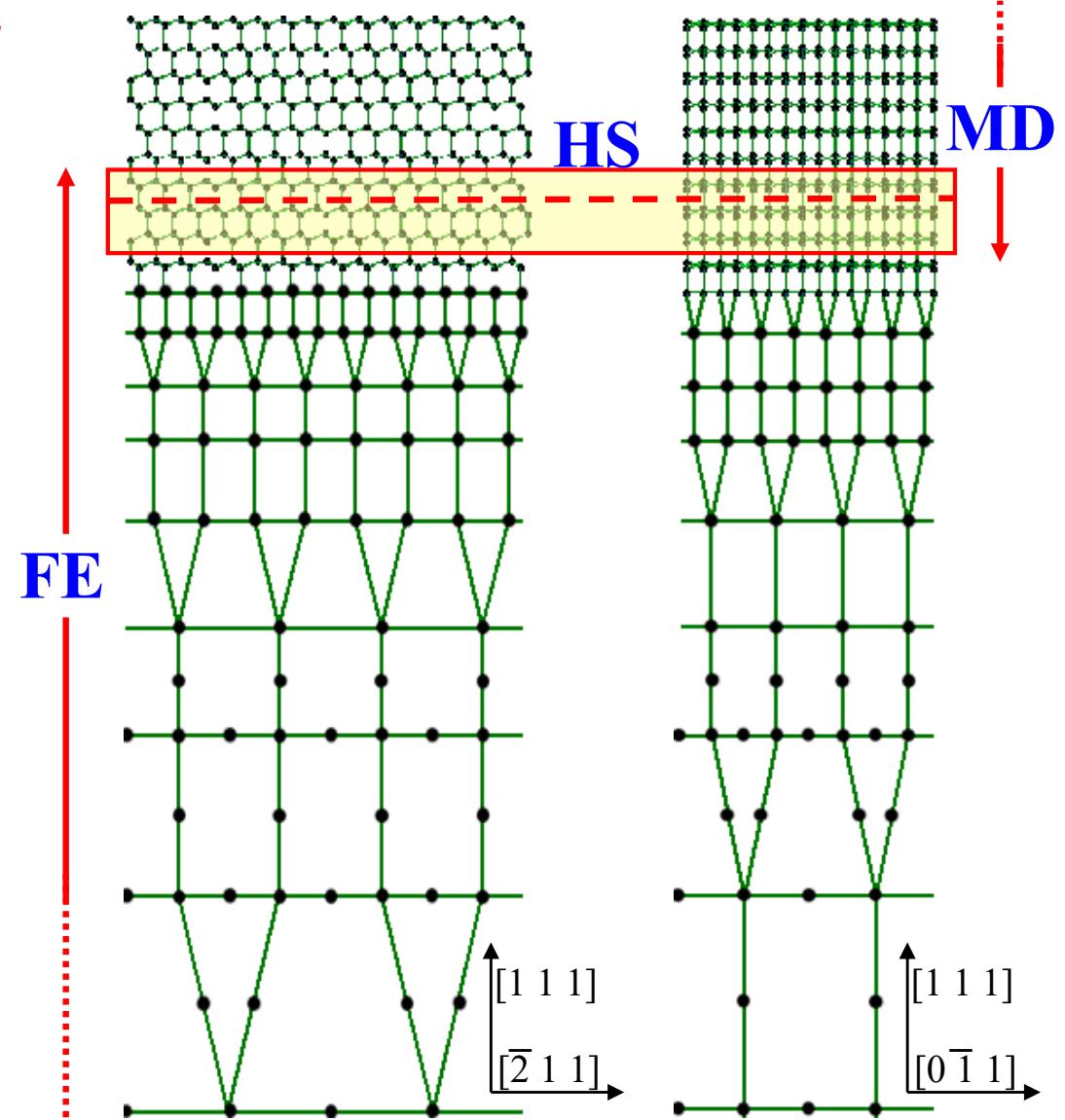
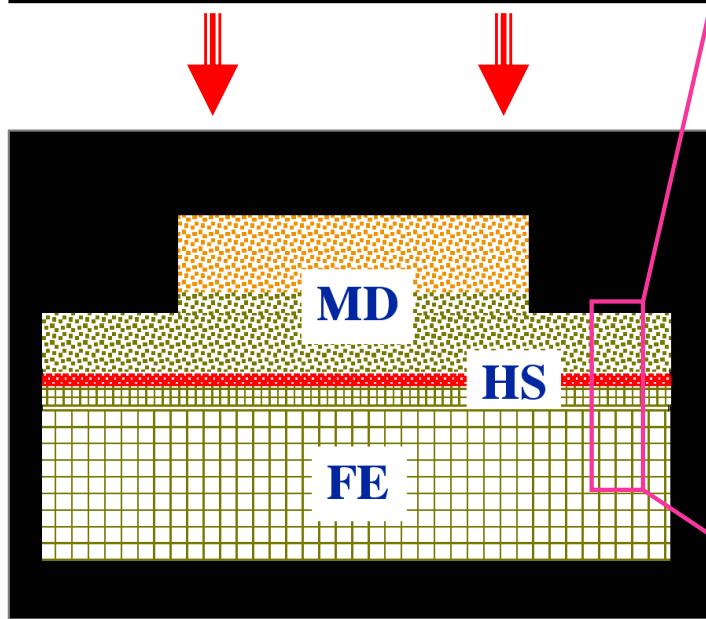
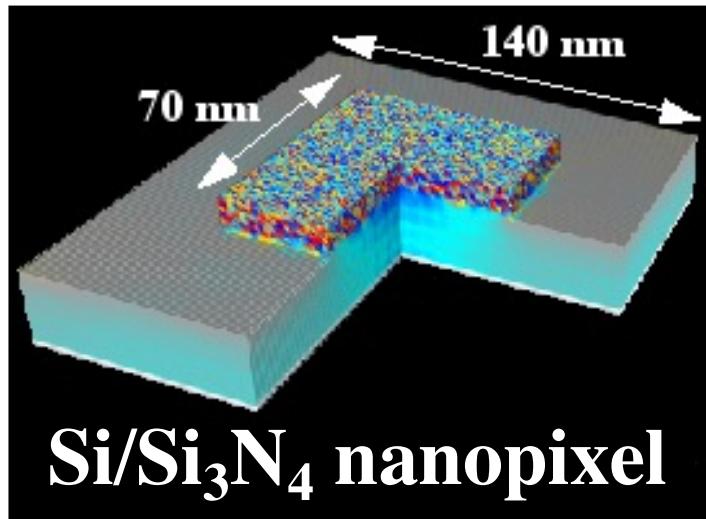
Significant dependence of the
reaction on stress intensity factor

Atomistic Simulations of Nanodevices



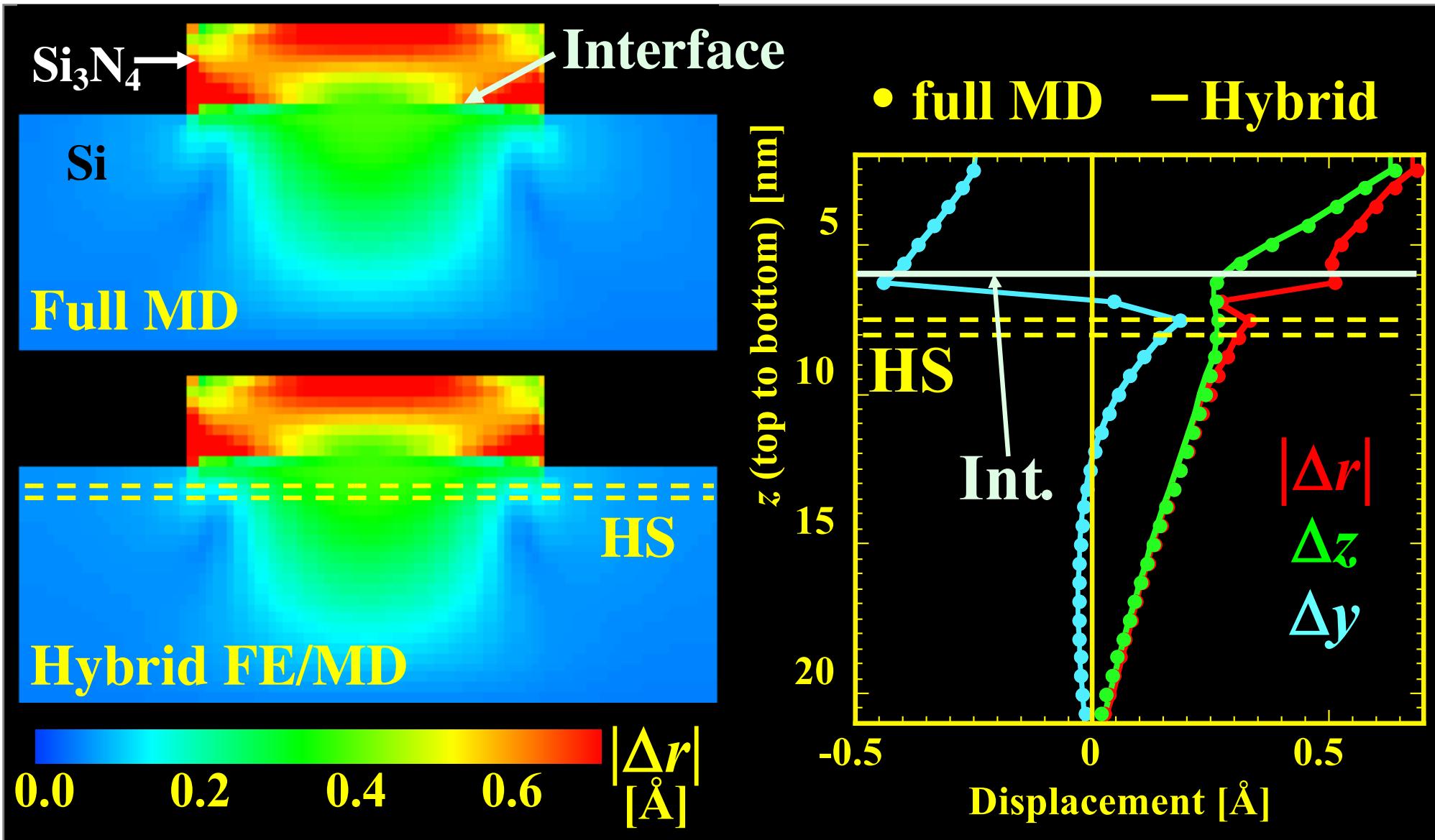
Hybrid FE/MD Algorithm

- FE nodes & MD atoms coincide in the handshake region
- Additive hybridization

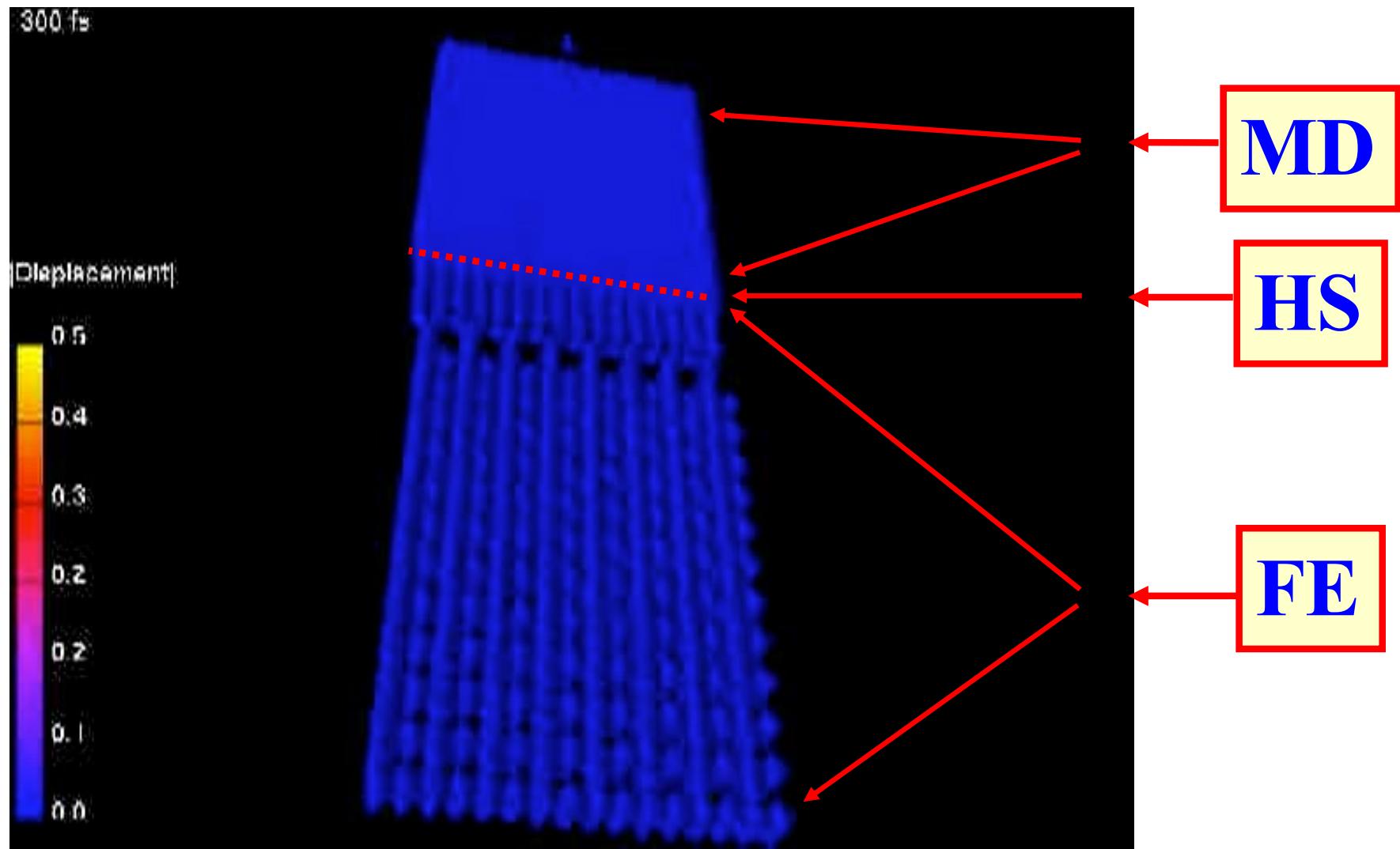


Si(111)/Si₃N₄(0001) Nanopixel

Displacement from equilibrium positions



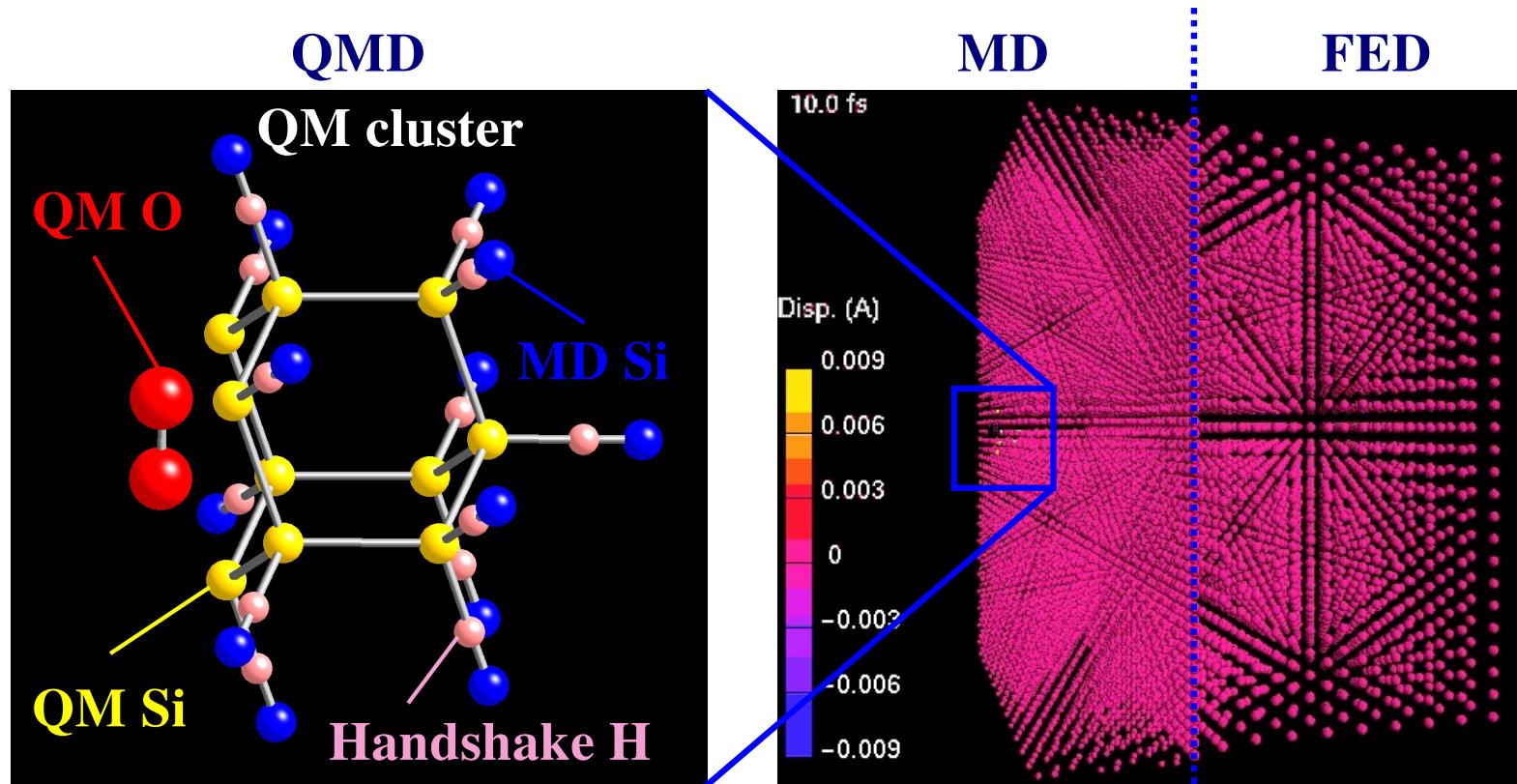
Dynamics Test & Demonstration: Projectile Impact on Silicon



Waves propagate seamlessly into the FE region

Application of Multiscale Simulations

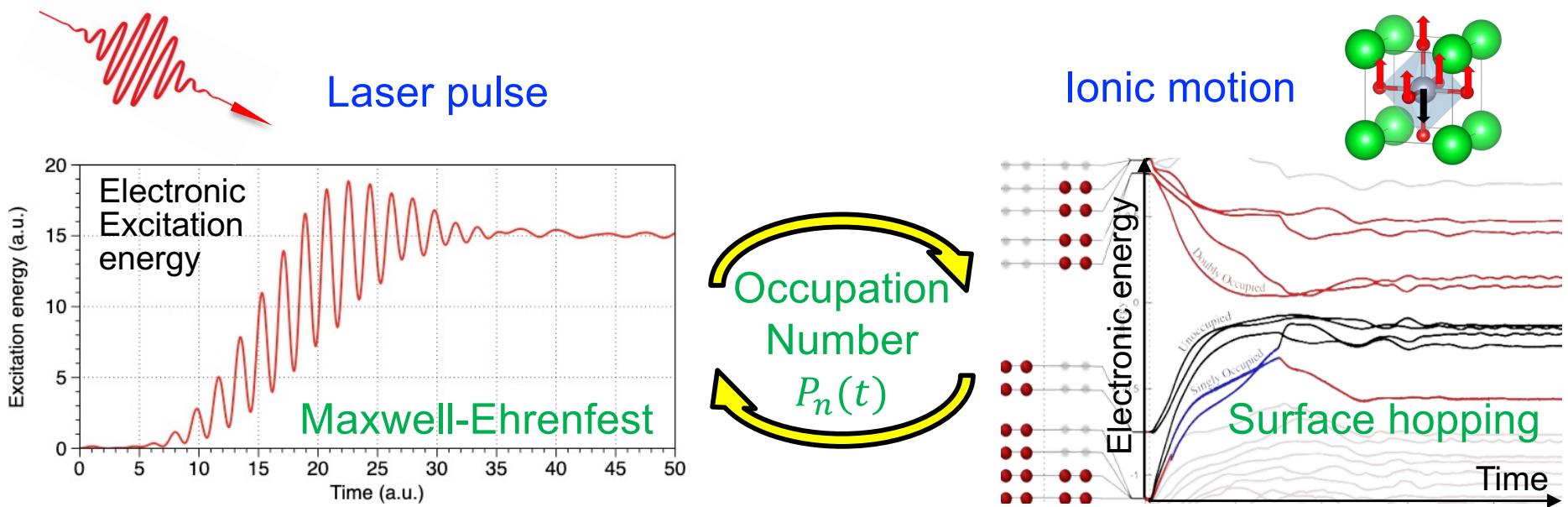
Oxidation dynamics on Si surface



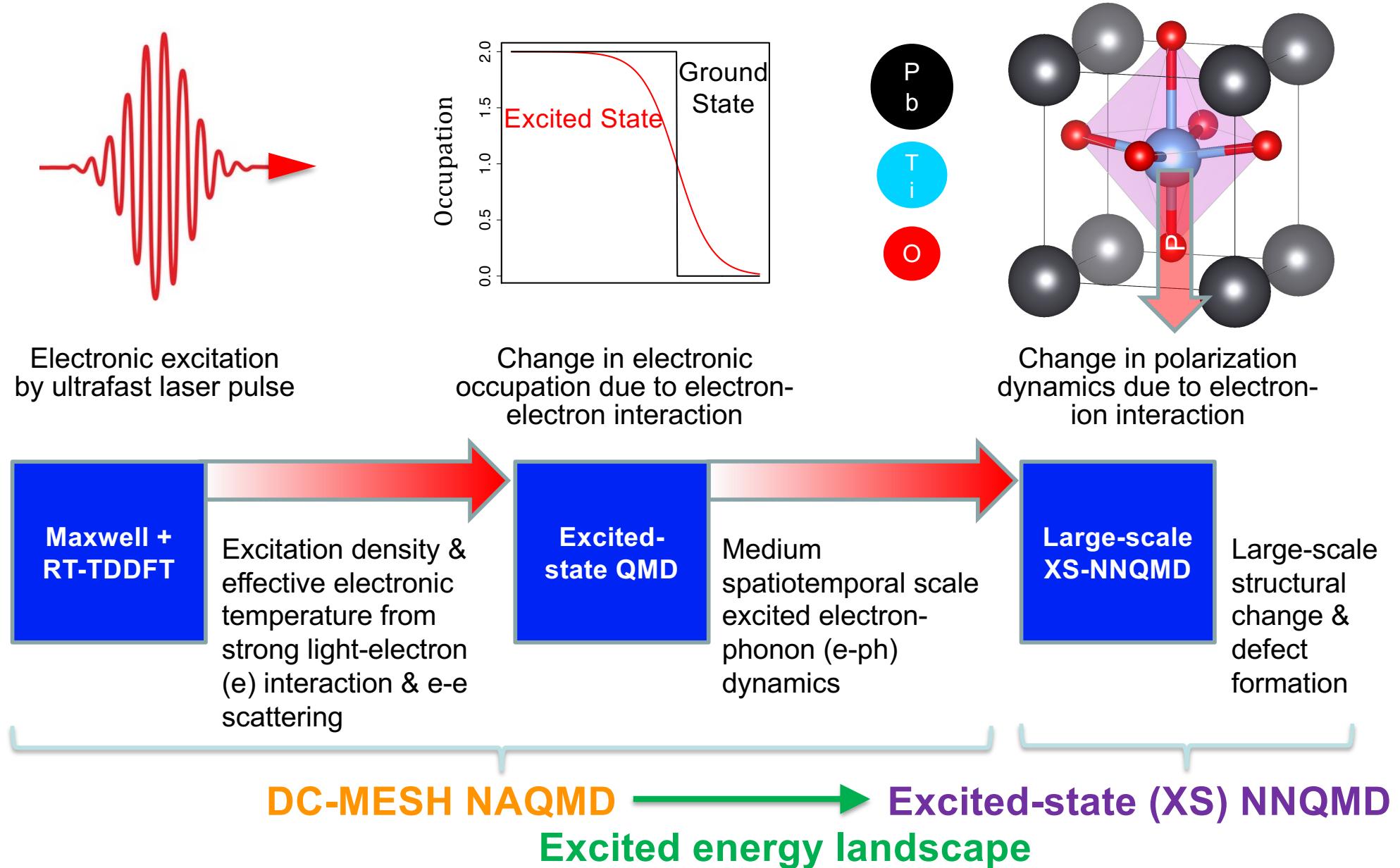
QMD/MD/FED:
Quantum molecular dynamics/molecular dynamics/finite-element dynamics

Light-Matter Interaction: DC-MESH

- DC-MESH (divide-&-conquer Maxwell + Ehrenfest + surface-hopping): $O(N)$ algorithm to simulate photo-induced quantum materials dynamics
- LFD (local field dynamics): Solve Maxwell equations for light & real-time time-dependent density functional theory (RT-TDDFT) equations for electrons to describe light-matter interaction
- QXMD (quantum molecular dynamics with excitation): Describe nonadiabatic coupling of excited electrons & ionic motions *via* surface-hopping approach *Nature Commun.* 8, 1745 ('17); *Nature Photon.*, 13, 425 ('19)
- LFD-QXMD handshaking *via* electronic occupation numbers



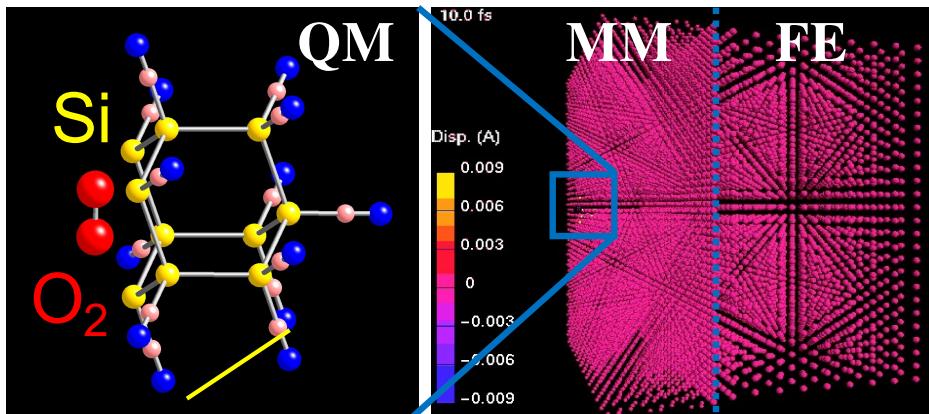
Multiscale DC-MESH + XS-NNQMD



Multiscale XN/NN

- **Multiscale quantum challenge:** Complex response of ferroelectric topological defects to external stimuli encompasses picosecond-to-nanosecond time & nanometer-to-micrometer length scales
- **QM/MM:** Overcame the challenge taking cue from multiscale quantum-mechanics (QM)/molecular mechanics (MM) approach (2013 Nobel chemistry prize)

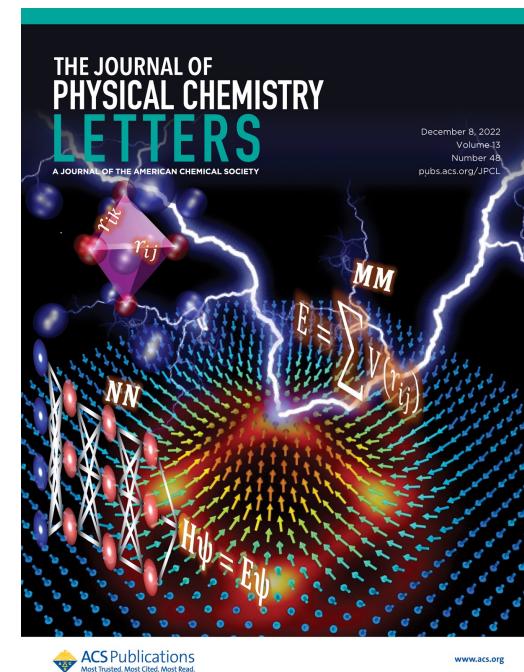
Warshel, *Angew. Chem.* **53**, 10020 ('14)



QM/MM/FE (finite-element method)

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

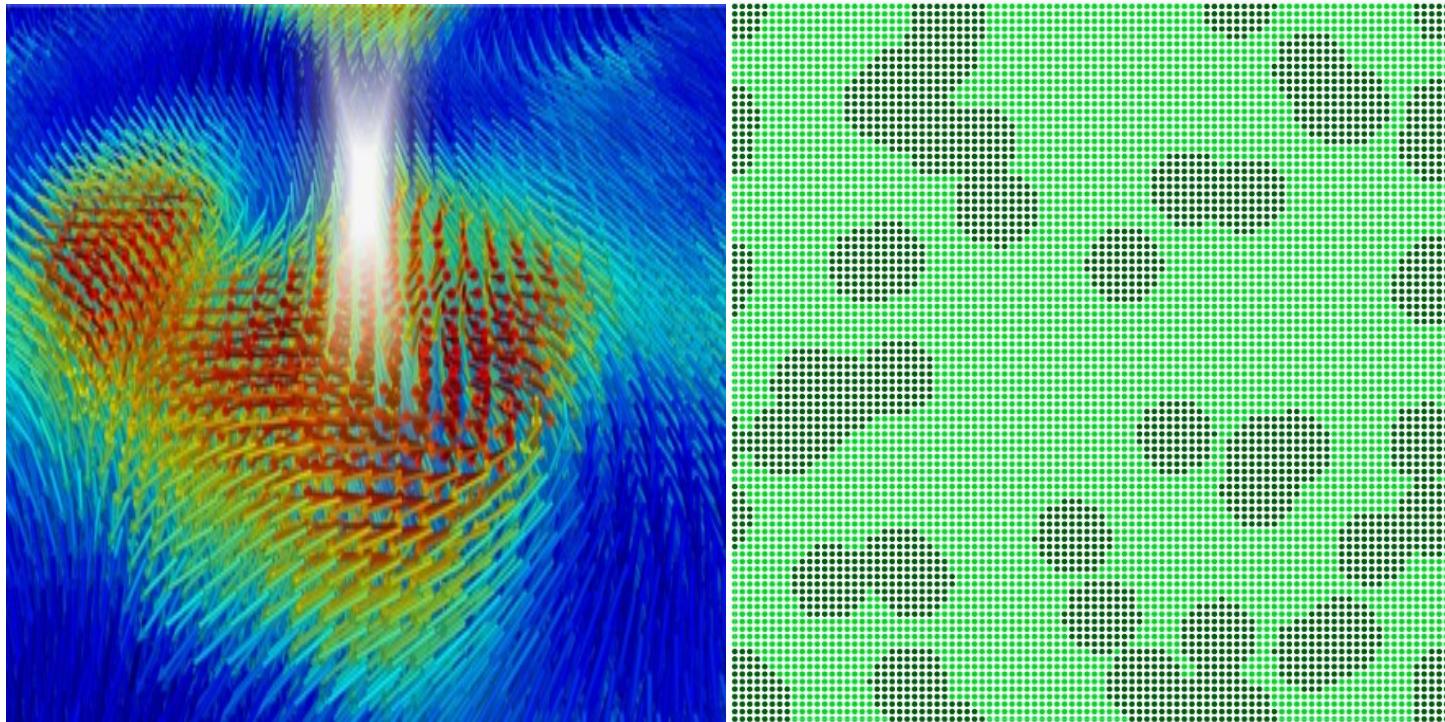
- **NN/MM:** NNQMD for ferroelectric (PbTiO_3 : PTO) embedded in MM for paraelectric (SrTiO_3 : STO) to apply appropriate strain boundary condition



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

- **XN/NN:** Fine-tuned excited-state NNQMD model is added perturbatively to pretrained Allegro-FM ground-state NNQMD model

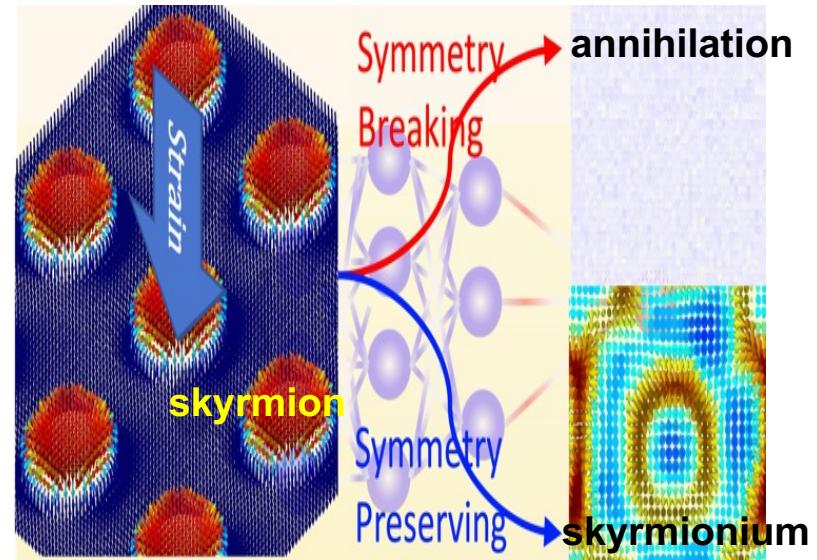
Application: Ferroelectric Opto-Toptronics



*ML for
large scale!*

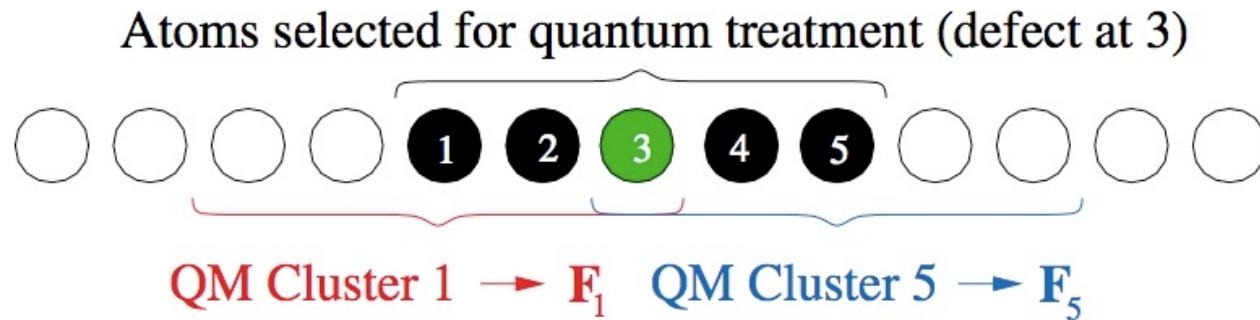
- Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications
- Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)
- Symmetry-controlled skyrmion-to-skyrmionium^{*} switching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

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



“Learning on the Fly” MD/QM

- Use parameterized interatomic potential, with parameters varying atom-by-atom
- Runtime refit of the parameters in chemically reactive regions by performing small quantum-mechanical calculations



Csanyi *et al.*, *Phys. Rev. Lett.* **93**, 175503 ('04); *Nature* **455**, 1224 ('08)

- Use expressive machine-learning (ML) potential like neural network (NN)¹ or Gaussian approximation potential (GAP)²
- Active learning to use uncertain quantification (UQ) of the ML potential to re-train the model only when needed^{3,4}

¹ Behler & Parrinello, *Phys. Rev. Lett.* **98**, 146401 ('07); *IJQC* **115**, 1032 ('15)

² Bartok *et al.*, *Phys. Rev. Lett.* **104**, 136403 ('10)

³ Zhang *et al.*, *Phys. Rev. Mater.* **3**, 023804 ('19)

⁴ Vandermause *et al.*, *arXiv:1904.02042v1a* ('19)

Coarse Grained Molecular Dynamics

- Coarse graining: $\mathbf{u}_j = \sum_{\mu} f_{j\mu} \mathbf{u}_{\mu}$, cf. wavelet smoothing
- Reduced (constrained) Hamiltonian → equations-of-motion for \mathbf{u}_j 's

$$E(\mathbf{u}_k, \dot{\mathbf{u}}_k) = \langle H_{MD} \rangle_{\mathbf{u}_k, \dot{\mathbf{u}}_k}$$

$$= \int d\mathbf{x}_{\mu} d\mathbf{p}_{\mu} H_{MD} e^{-\beta H_{MD}} \Delta / Z,$$

$$\Delta = \prod_j \delta\left(\mathbf{u}_j - \sum_{\mu} \mathbf{u}_{\mu} f_{j\mu} \right) \delta\left(\dot{\mathbf{u}}_j - \sum_{\mu} \frac{\mathbf{p}_{\mu} f_{j\mu}}{m_{\mu}} \right),$$

R. E. Rudd & J. Q. Broughton, *Phys. Rev. B* **58**, R5893 ('98)
R. Kobayashi et al., *Int'l J. Num. Method Eng.* **83**, 249 ('10)

cf. J. Fish *et al.*, *Comput. Methods Appl. Mech. Eng.* **196**, 908 ('07)

Coarse Graining Using Wavelets

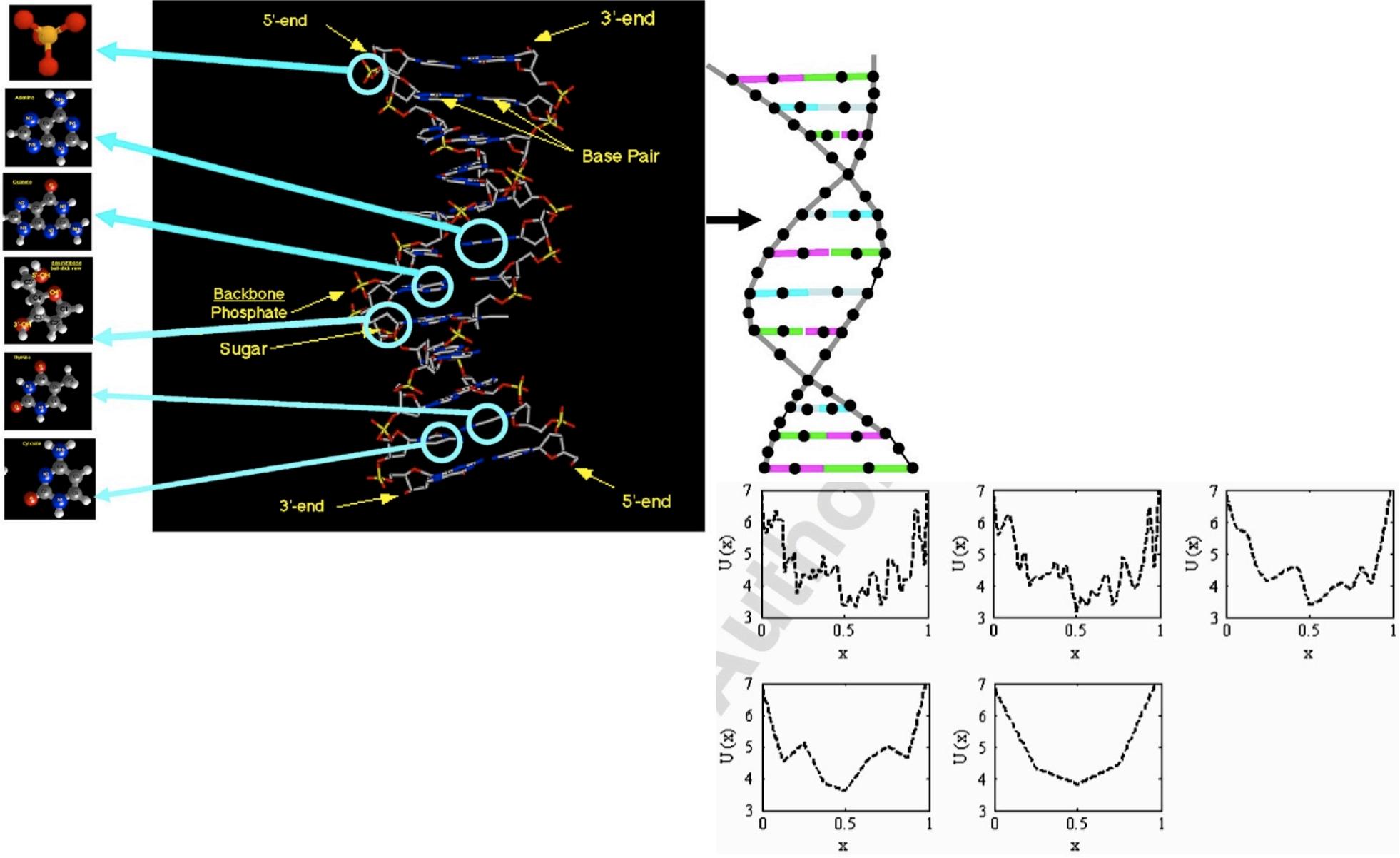


Fig. 11. Effective bond potential based on wavelet multi-scale projection of fine scale bond potential.

J.-S. Chen et al., *Finite Elements in Analysis & Design* 43, 346 ('07)

Quasicontinuum Method

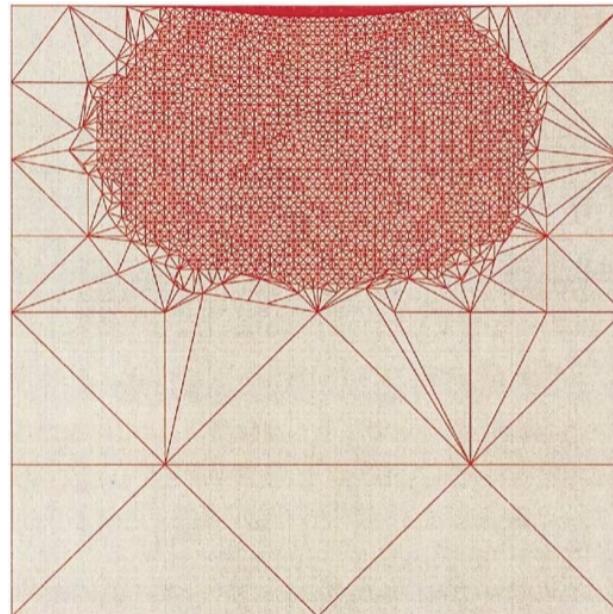
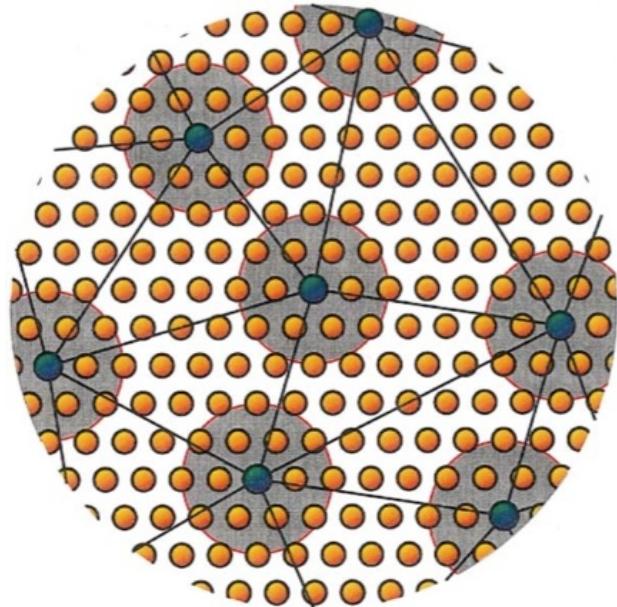
- Piecewise interpolation of the deformation of a selected subset of atoms

$$\mathbf{x}_\mu = \sum_j N_j(\mathbf{x}_\mu) \mathbf{x}_j$$

- Coarse graining as numerical quadrature

$$E_{\text{total}} = \sum_j w_j E_j$$

- Summation weight locally determined by a cluster of atoms centered at the “representative atoms”



Shenoy *et al.*, *J. Mech. Phys. Solids* **47**, 611 ('99)
Knap & Ortiz, *J. Mech. Phys. Solids* **49**, 1899 ('01)

Linear-Scaling MD/FE Optimization

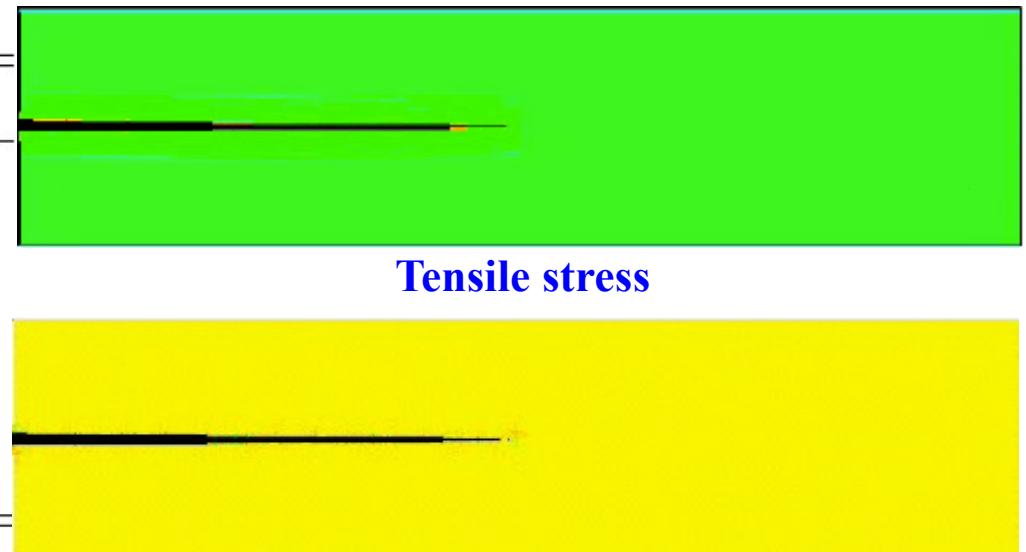
Iterate:

1. Perform a small # of steps of conventional (e.g., CG) energy minimization
2. Perform one step where solid is described by elastic theory
 - a. Calculate the forces acting on the atoms
 - b. Transfer the atomic forces onto a computational grid
 - c. Solve linear elastic eq. with a **multigrid method** to obtain the displacement field
 - d. Get the atomic displacements by interpolating the displacement field
 - e. Move the atoms along the displacement directions

TABLE I. Number of force evaluations n_f and CPU time T in seconds for the conjugate gradient (CG) and the linear scaling (SC) method for a divacancy in silicon.

Number of atoms	n_f (CG)	n_f (LS)	T (CG)	T (LS)
510	102	106	0.41	0.50
998	124	106	0.90	0.93
1726	146	109	1.7	1.6
4094	184	115	5.1	4.2
13822	260	115	24.0	14.0
110592	502	115	373.0	135.0
884734	934 \leftrightarrow	117	5586.0	1147.0

**100 million-atom molecular dynamics simulation
of a crack tip in GaAs**



Goedecker *et al.*,
Phys. Rev. B 64, 161102(R) ('01)

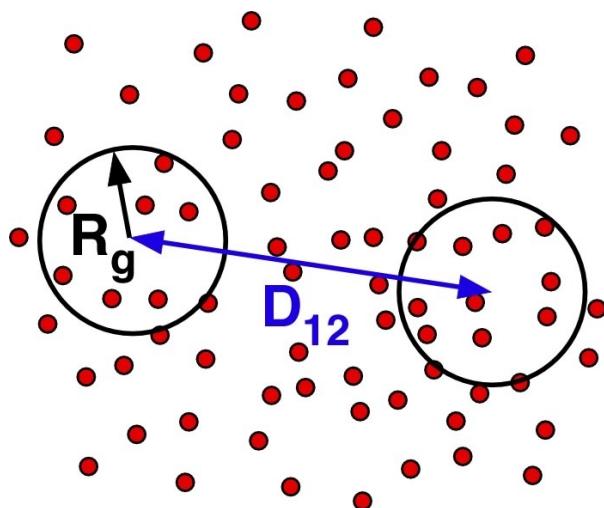
Dissipative Particle Dynamics

$$\frac{d^2 \mathbf{r}_i}{dt^2} = \sum_{j(\neq i)} \left(\mathbf{f}_{ij}^C + \mathbf{f}_{ij}^D + \mathbf{f}_{ij}^R \right)$$

$$\begin{cases} \mathbf{f}_{ij}^C = a_{ij} (1 - r_{ij}) \hat{\mathbf{r}}_{ij} \Theta(1 - r_{ij}) & \text{coarse interaction} \\ \mathbf{f}_{ij}^D = -g(1 - r_{ij})^2 \Theta(1 - r_{ij}) (\mathbf{v}_{ij} \bullet \hat{\mathbf{r}}_{ij}) \hat{\mathbf{r}}_{ij} & \text{friction} \\ \mathbf{f}_{ij}^R = \sqrt{2gk_B T} (1 - r_{ij}) \Theta(1 - r_{ij}) \text{rnd}_{ij} \hat{\mathbf{r}}_{ij} & \text{random force} \end{cases}$$

- Generalized Langevin equation (Liouville equation & Mori-Zwanzig projection operator) for first-principles derivation of coarse forces

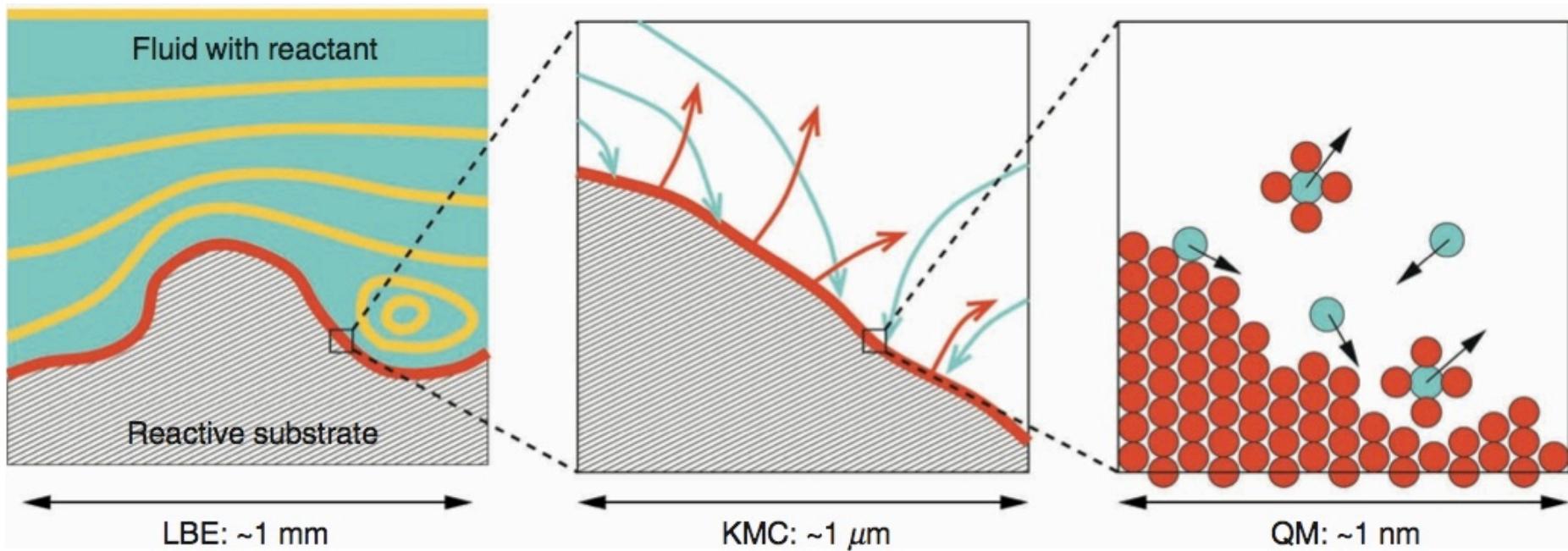
Kinjo & Hyodo, *Phys. Rev. E* 75, 051109 ('07)



Groot & Warren, *J. Chem. Phys.* 107, 4423 ('97)

Lattice Boltzmann + Atomistic

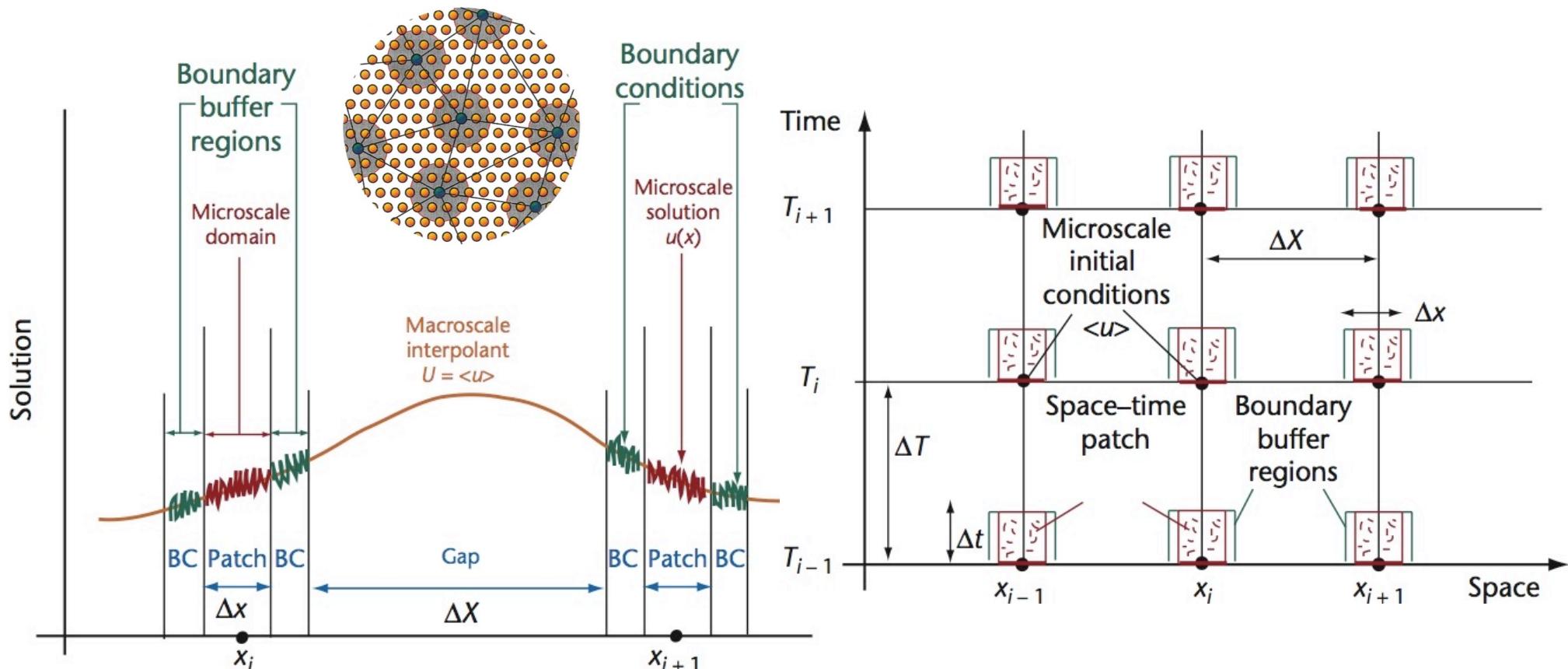
- Coupling fluid dynamics + atomistics/chemical reactions
- Possible breakdown of hydrodynamics at small length scales:
Boltzmann equation for $f(x, v, t)$



Succi, O. Filippova, Smith & Kaxiras, *Comp. Sci. Eng.* 3(6), 26 ('01)
Kwak et al., *Int'l J. Comput. Sci.* 3, 579 ('09)

Spatio-Temporal Interpolation

- Interpolation in both space & time (coarse model can be “equation free”)
- Fine simulations only in small space-time patches



Kevrekidis, C. W. Gear & Hummer, *AIChE J.* **50**, 1346 ('04)
Hyman, *Comp. Sci. Eng.* **7**(3), 47 ('05)