

Hybrid Particle-Continuum Simulation

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Collaboratory for Advanced Computing & Simulations

Department of Computer Science

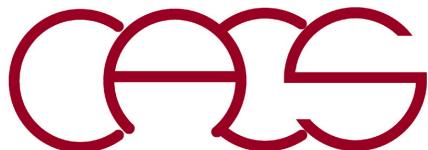
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Adaptively manage accuracy-cost trade-off;
coarse-graining by heuristics
(*i.e.*, switching to different abstract)



Multiscale Modeling

The Nobel Prize in Chemistry 2013



© Nobel Media AB

Martin Karplus



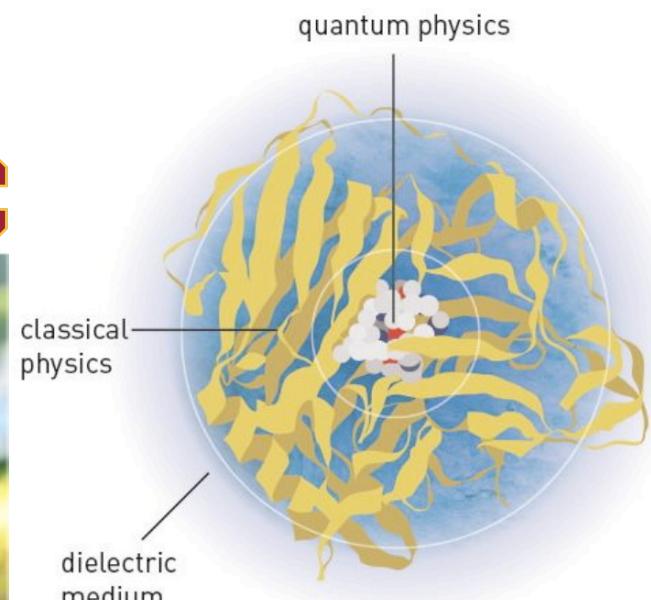
Photo: Keilana via
Wikimedia Commons

Michael Levitt



Photo: Wikimedia
Commons

Arieh Warshel



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

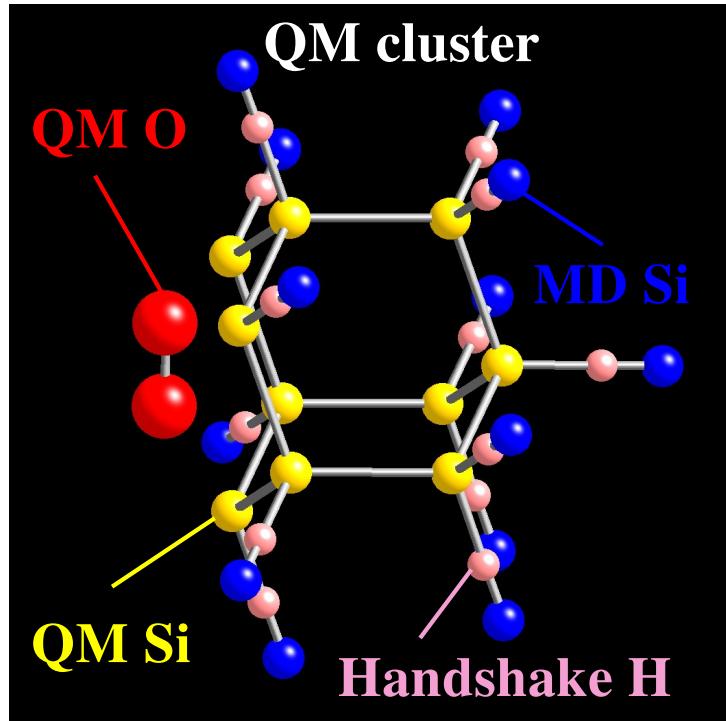
The Nobel Prize in Chemistry 2013 was awarded jointly to Martin Karplus, Michael Levitt and Arieh Warshel *"for the development of multiscale models for complex chemical systems"*.

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

Find multiscale modeling in your area!

Adaptive Multiscale Dynamics

QMD

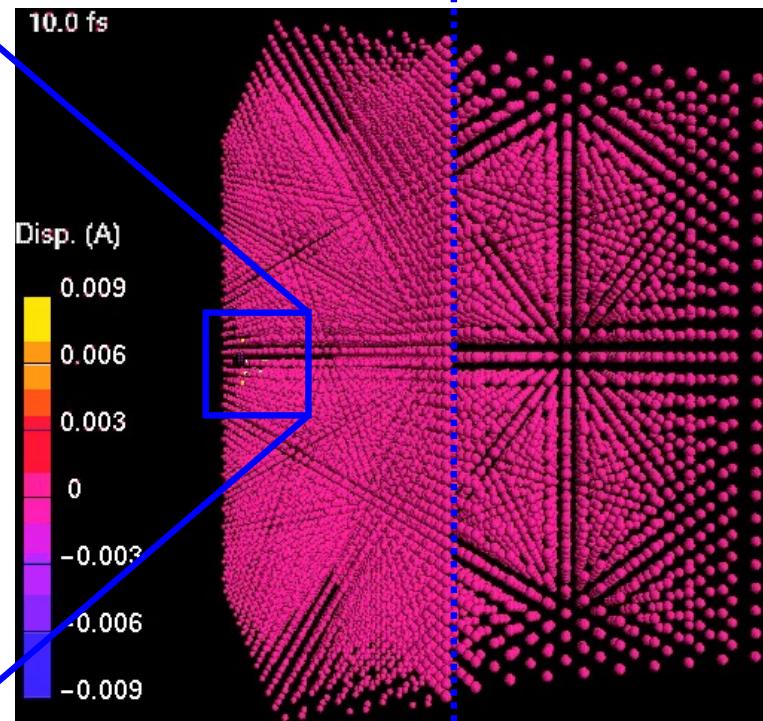


Oxidation of Si

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

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

MD



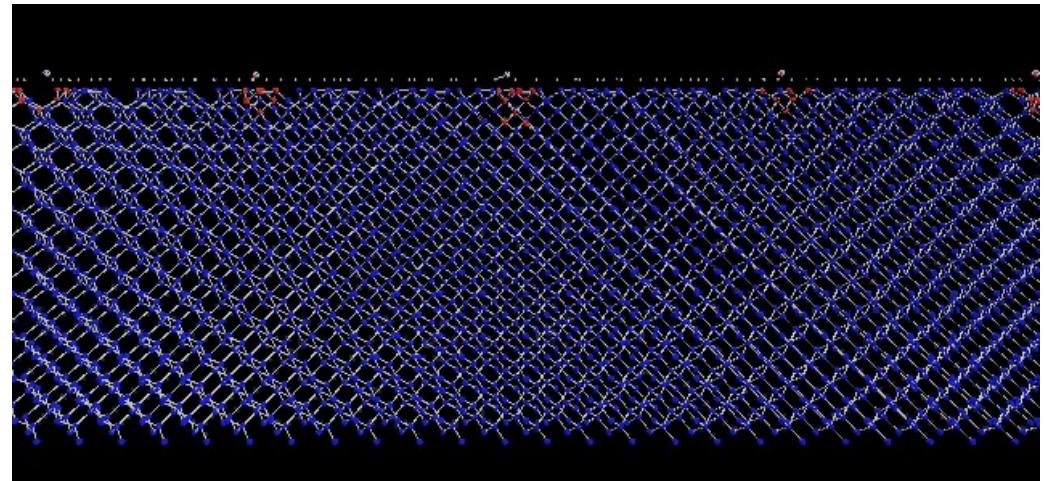
FED



High-energy
beam oxidation
of Si (SIMOX)

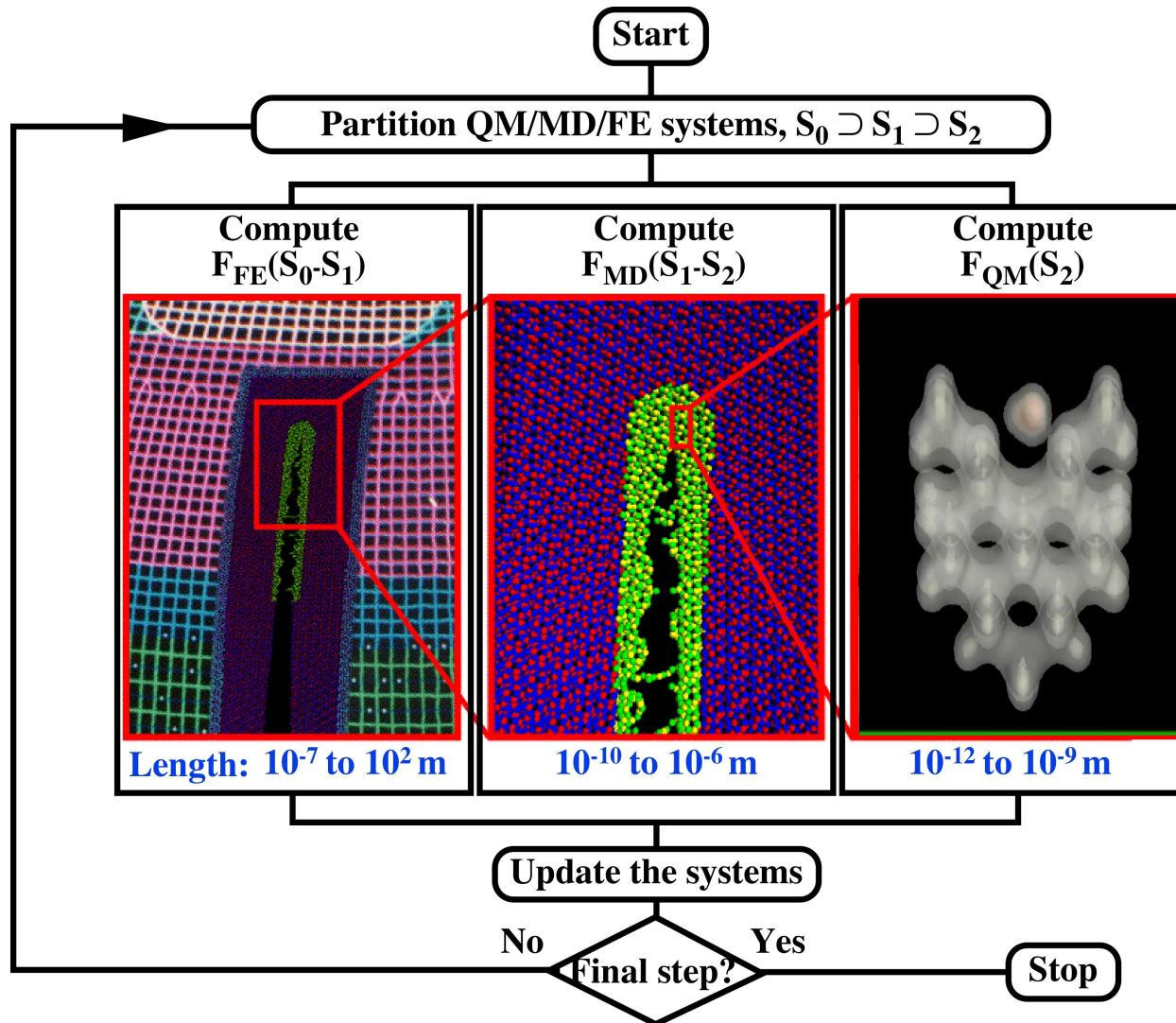
H. Takemiya *et al.*,
IEEE/ACM Supercomputing (SC06)

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:

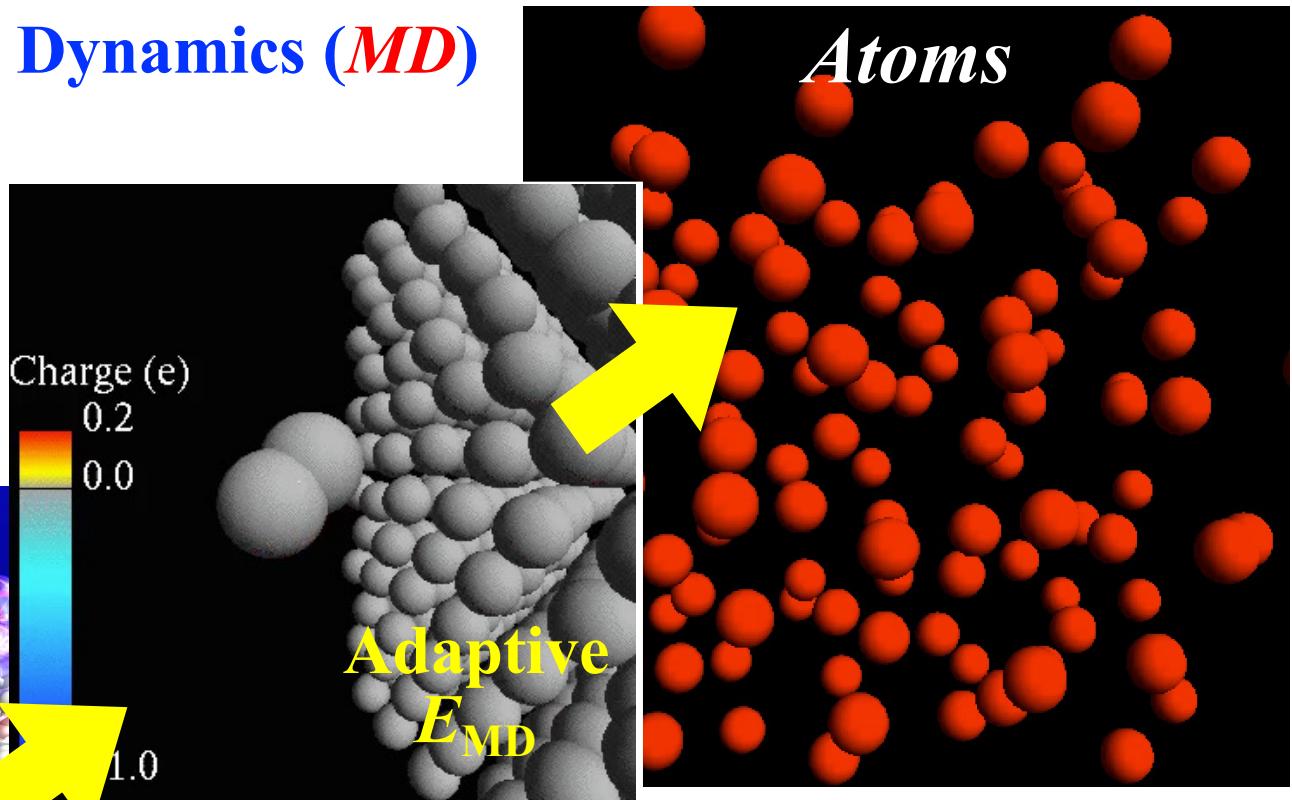
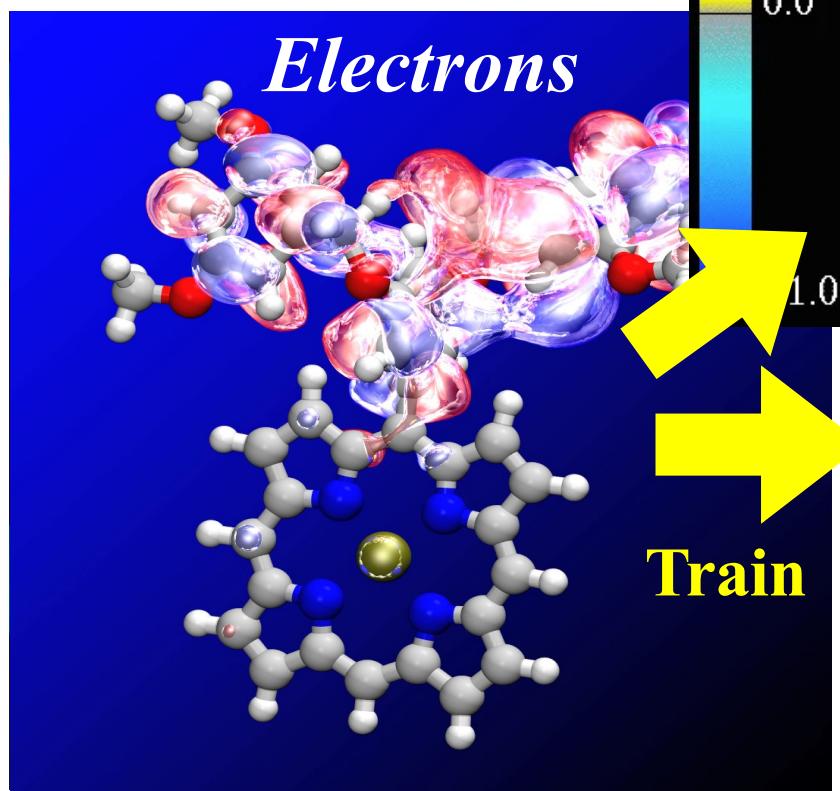
- Finite element (FE) dynamics based on continuum elasticity
- Atomistic molecular dynamics (MD) simulation
- Quantum molecular dynamics (QMD) based on the density functional theory (DFT)

Molecular Dynamics & Machine Learning

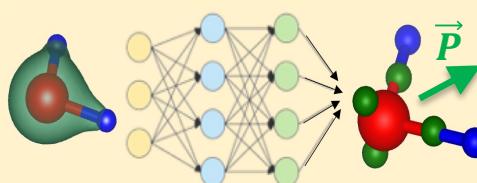
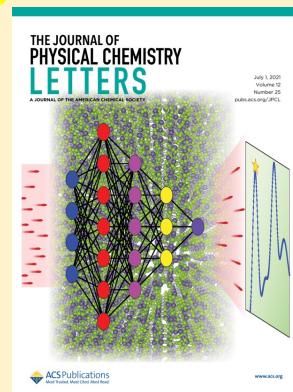
Molecular Dynamics (*MD*)

Reactive MD (*RMD*)

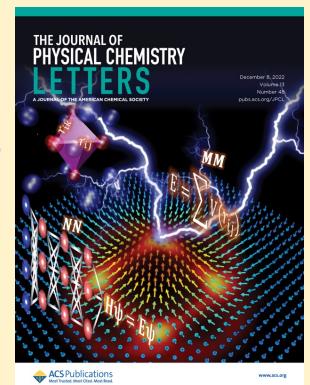
Nonadiabatic quantum MD (*NAQMD*)



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

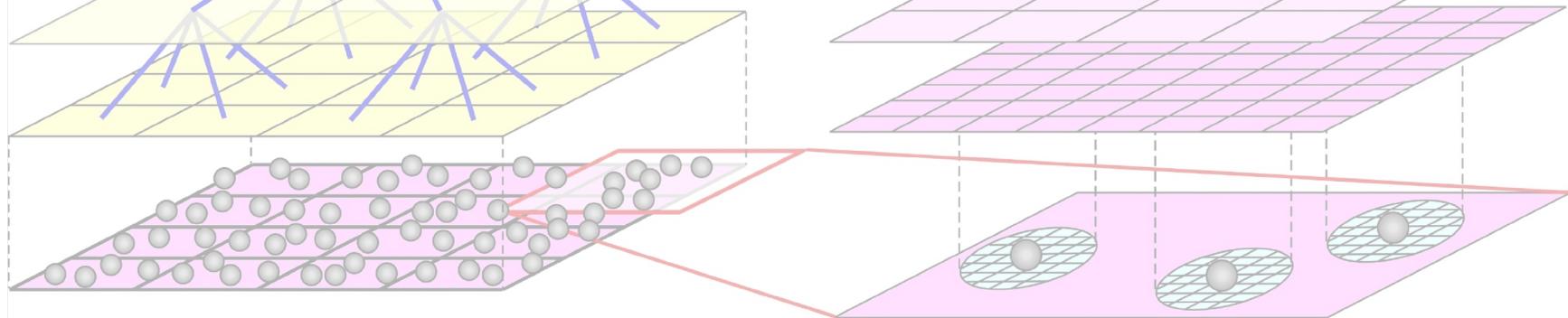


Physical Review Letters
Editor's choice
(May 25, '21)

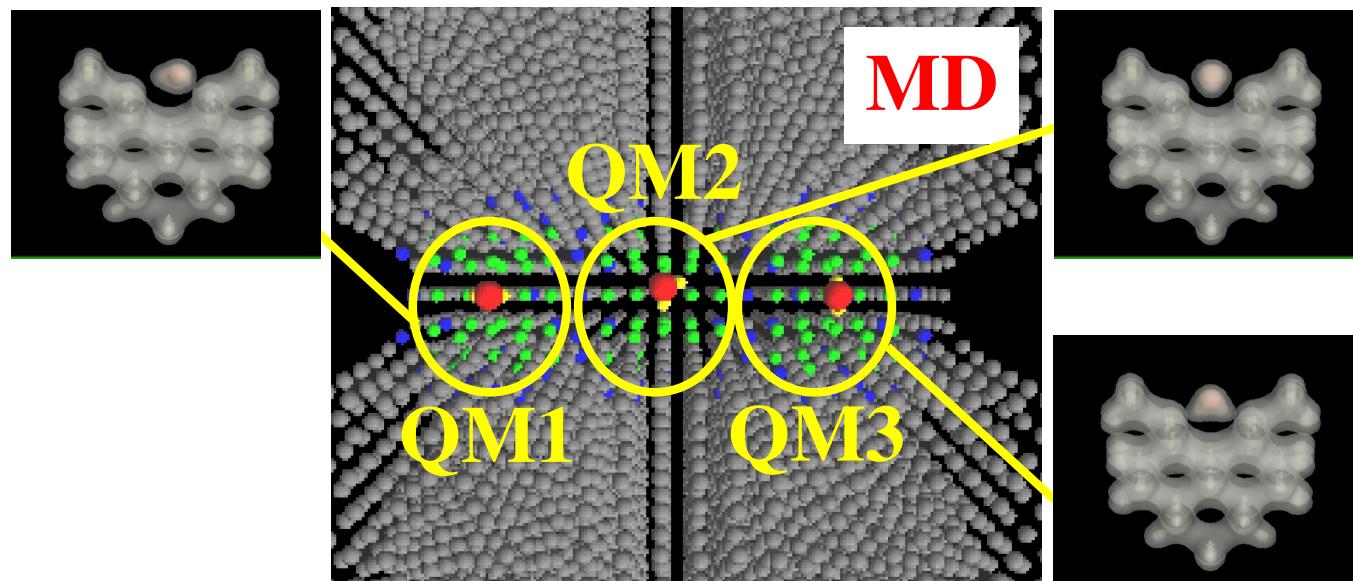


DC Multiscale MD/QM (DFT)

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



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

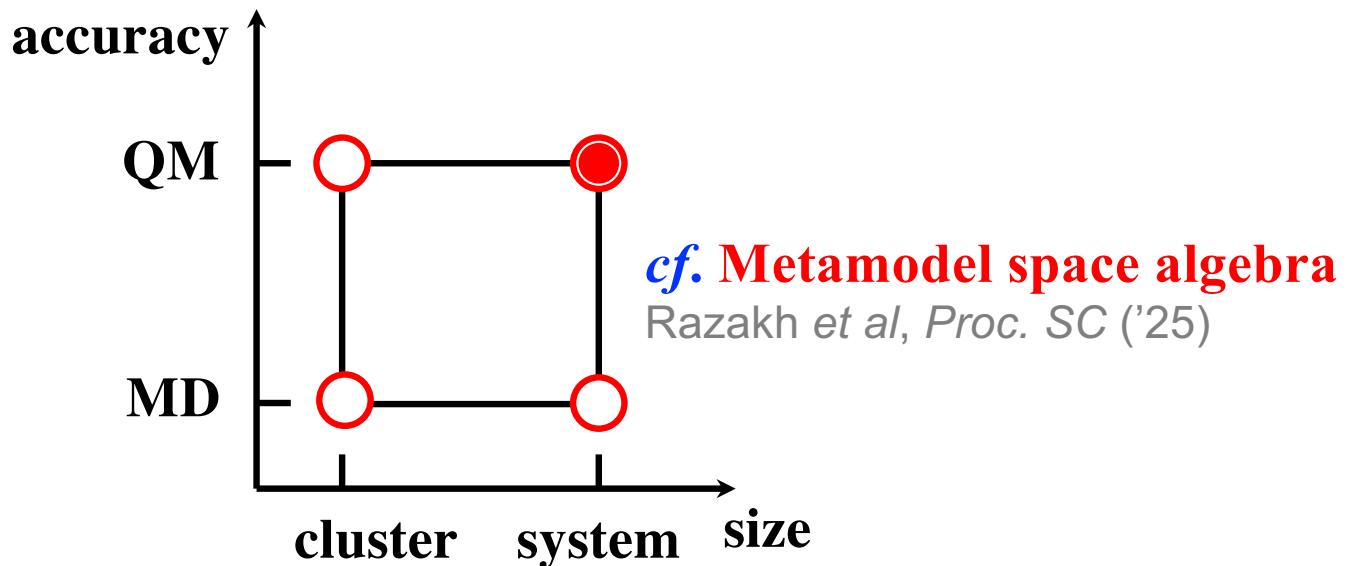


Additive Hybridization

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

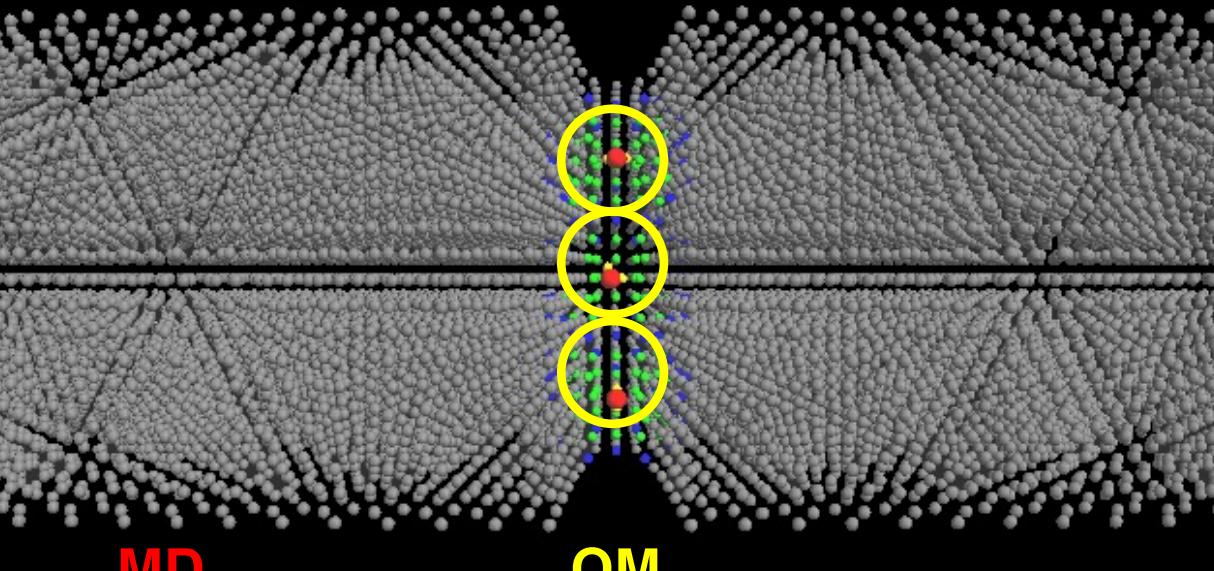


$$E_{\text{QM}}(s) - E_{\text{QM}}(c) \cong E_{\text{MD}}(s) - E_{\text{MD}}(c)$$

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

Environmental Effect on Fracture

Reaction of H₂O molecules at a Si crack tip

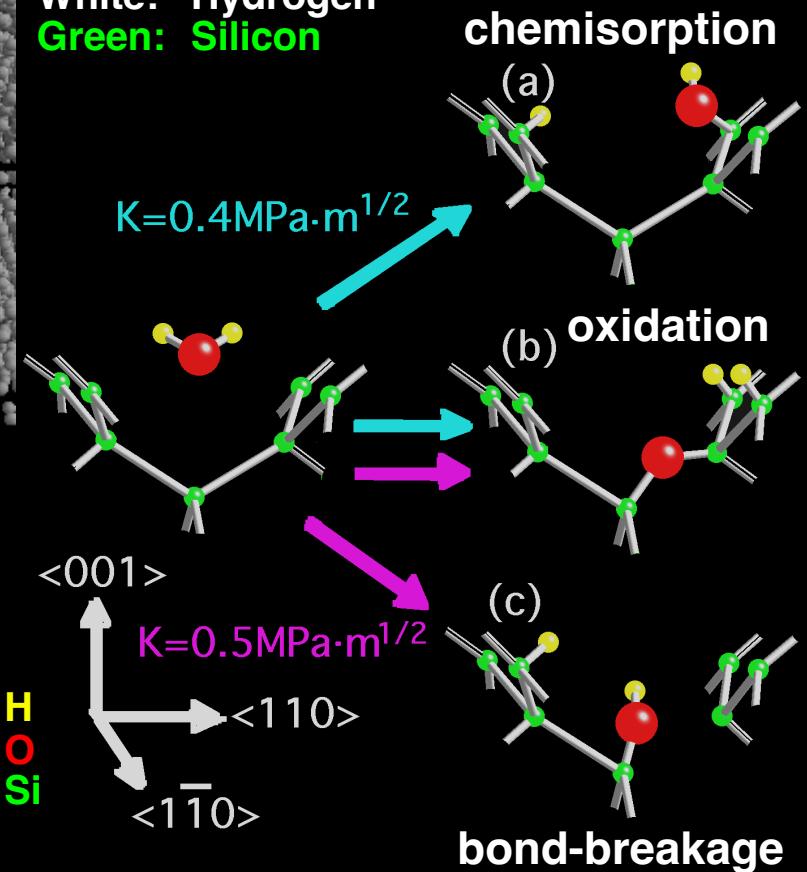


MD

QM

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

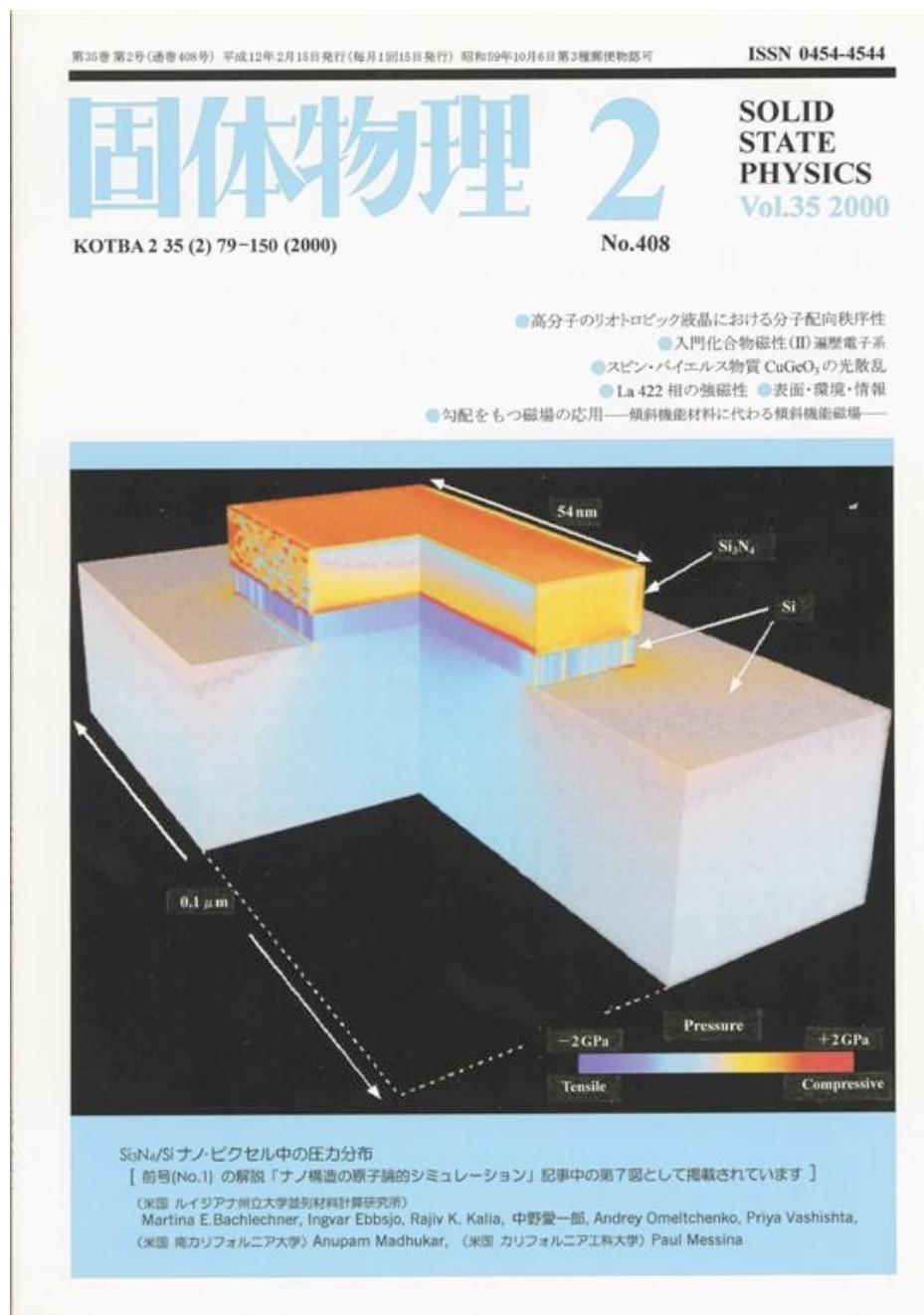
Blue: Oxygen
White: Hydrogen
Green: Silicon



Yellow: H
Red: O
Green: Si

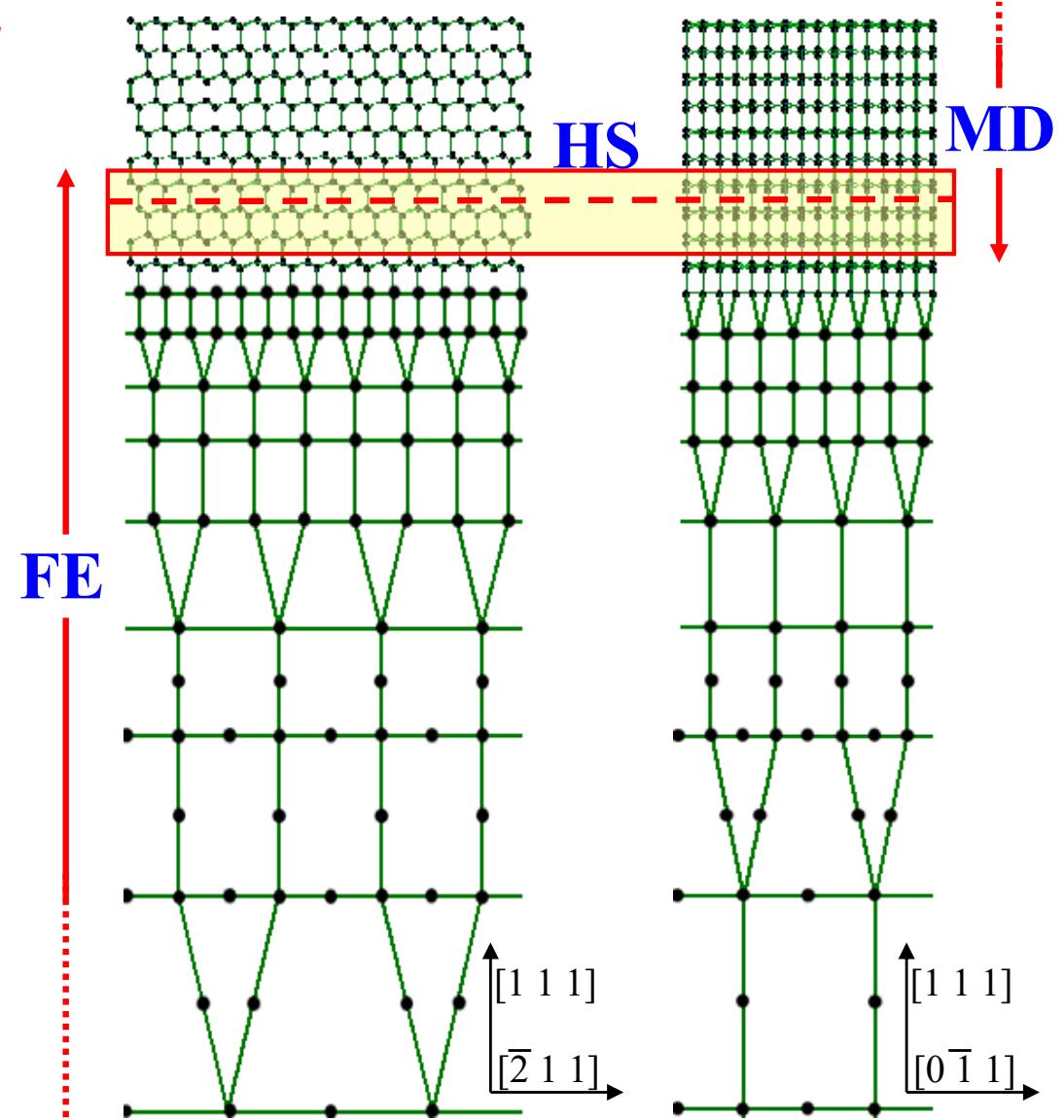
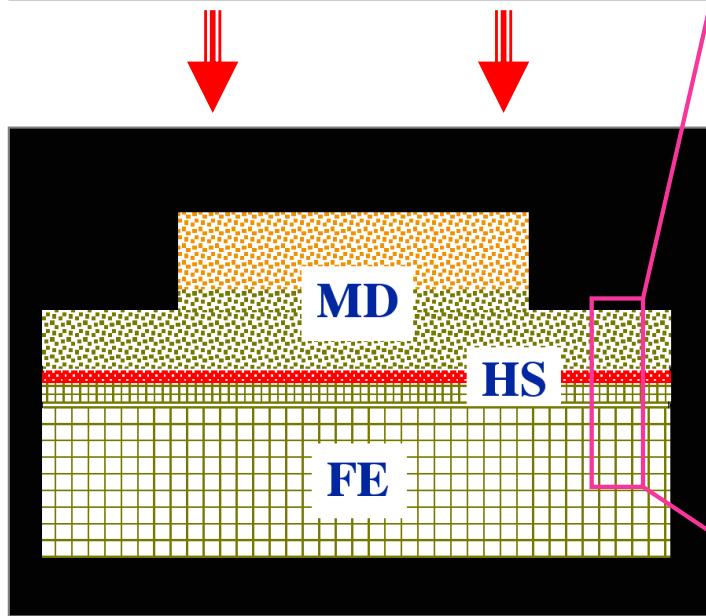
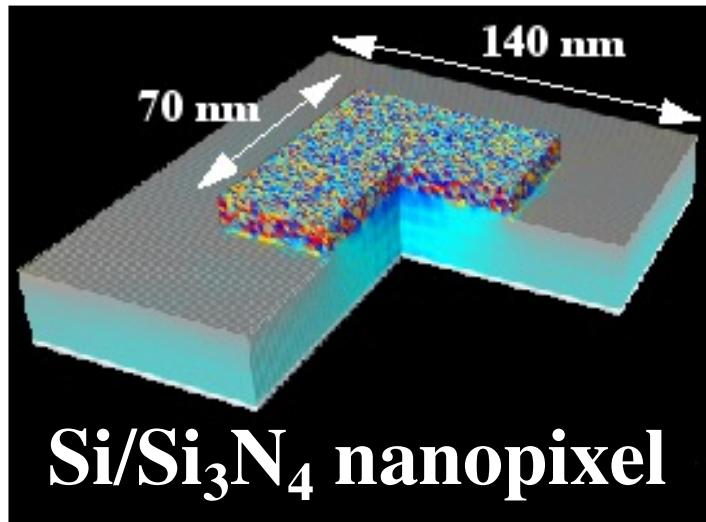
Significant dependence of the
reaction on stress intensity factor

Atomistic Simulations of Nanodevices



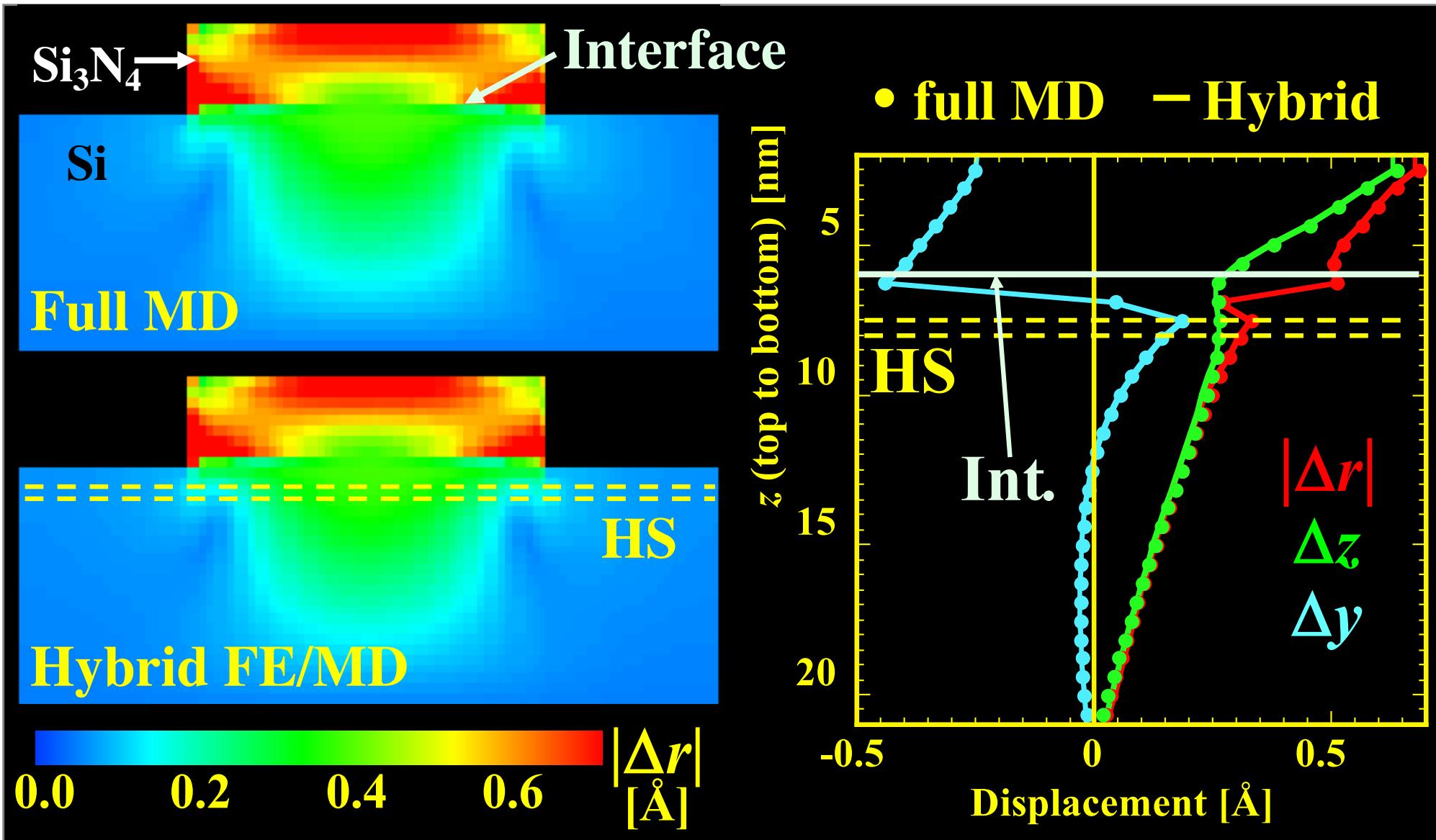
Hybrid FE/MD Algorithm

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

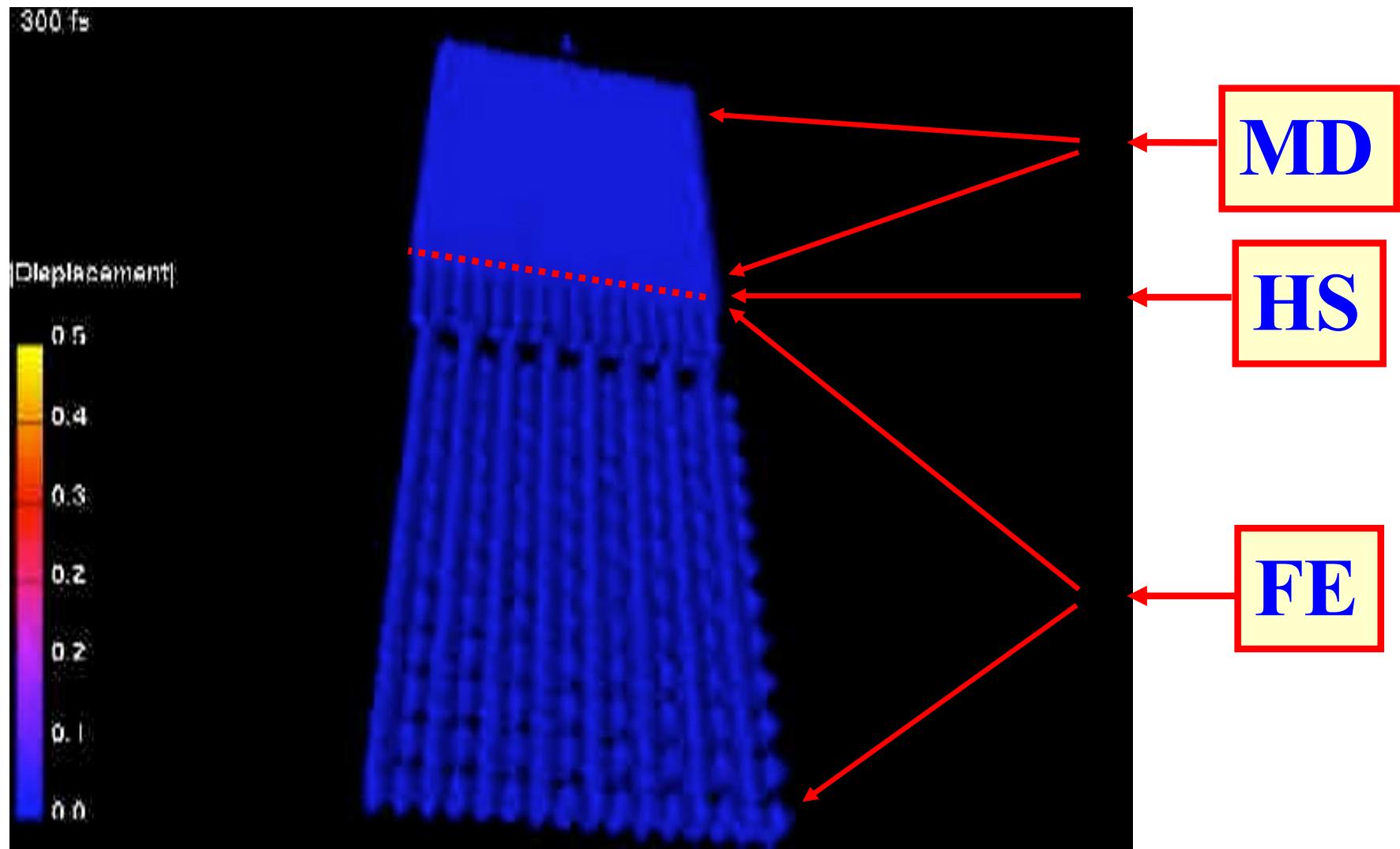


Si(111)/Si₃N₄(0001) Nanopixel

Displacement from equilibrium positions



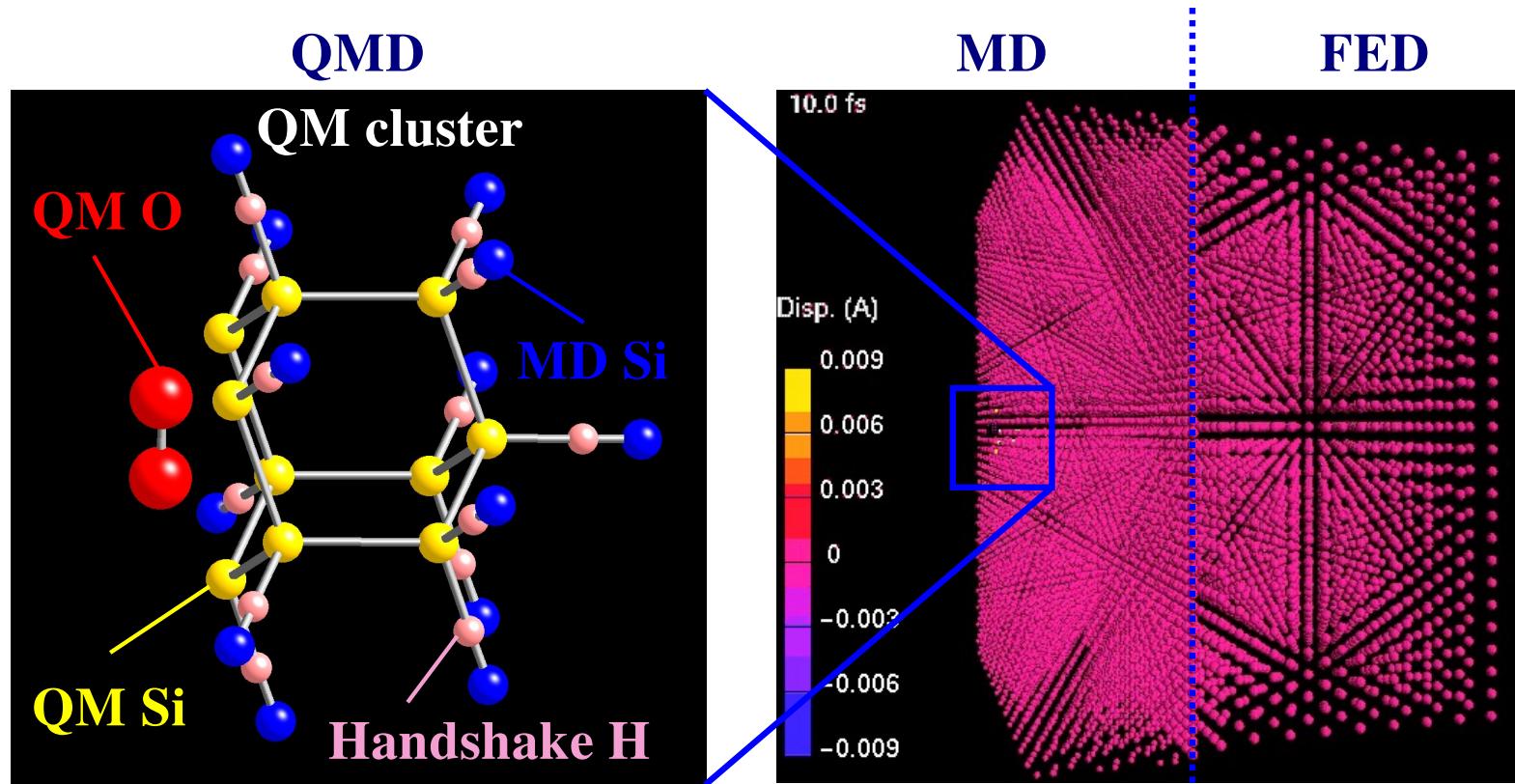
Dynamics Test & Demonstration: Projectile Impact on Silicon



Waves propagate seamlessly into the FE region

Application of Multiscale Simulations

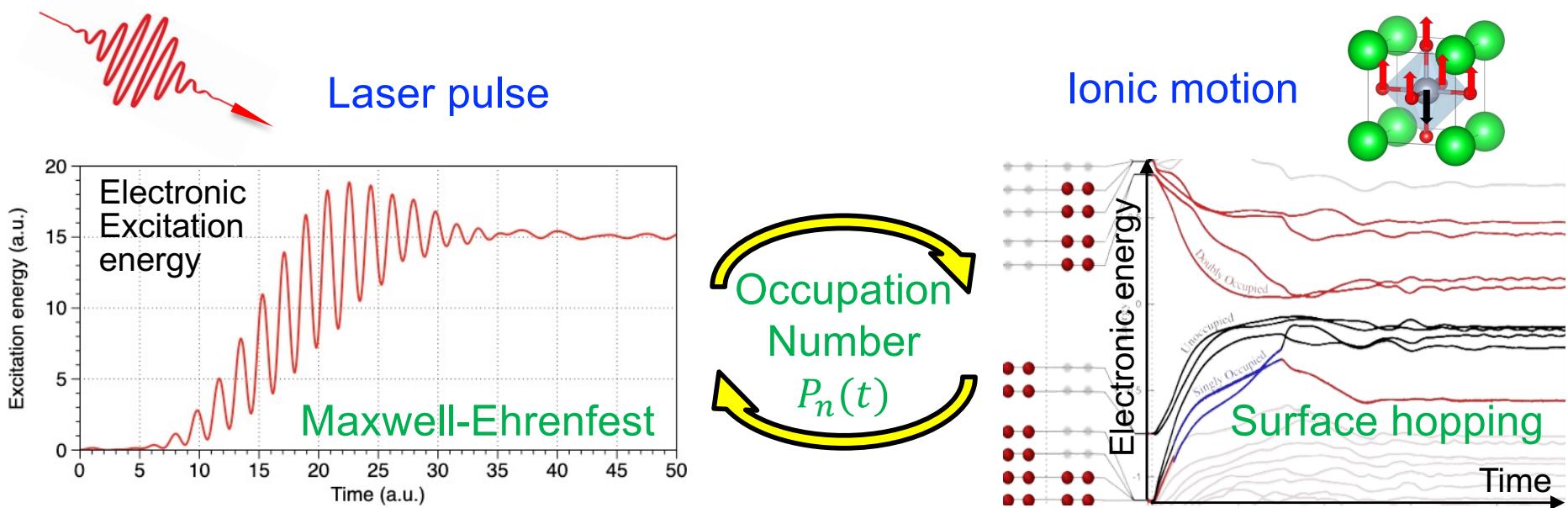
Oxidation dynamics on Si surface



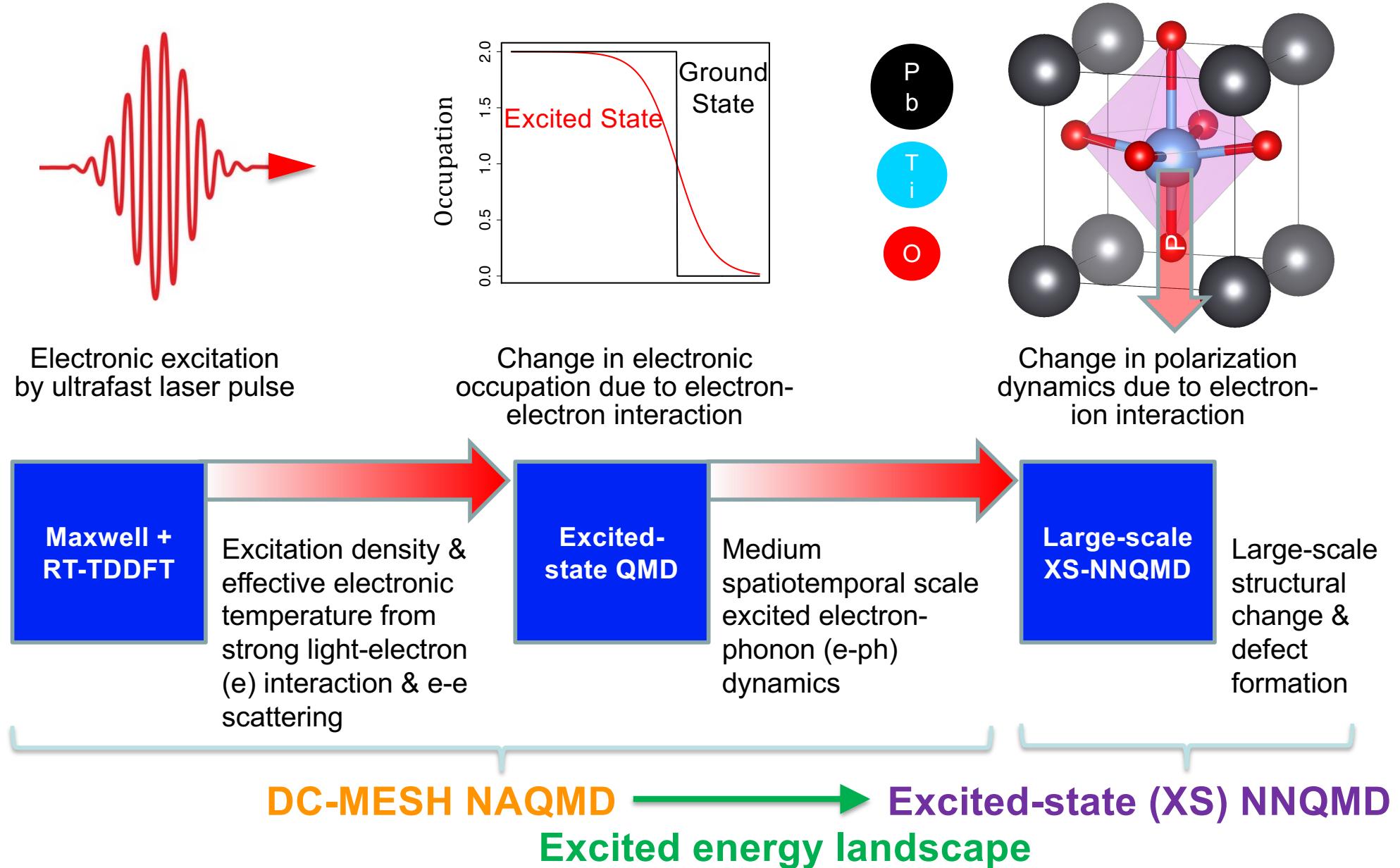
QMD/MD/FED:
Quantum molecular dynamics/molecular dynamics/finite-element dynamics

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



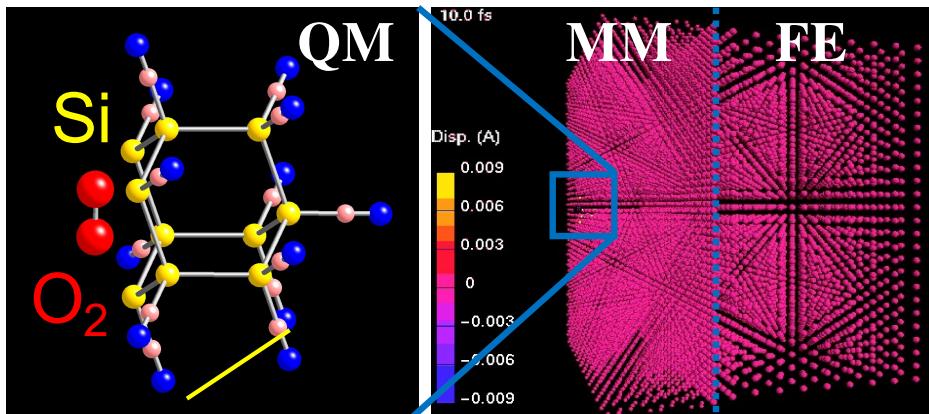
Multiscale DC-MESH + XS-NNQMD



Multiscale XN/NN

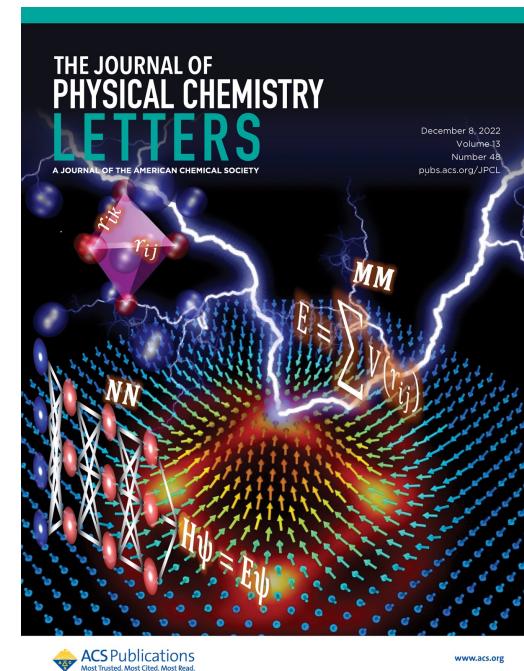
- **Multiscale quantum challenge:** Complex response of ferroelectric topological defects to external stimuli encompasses picosecond-to-nanosecond time & nanometer-to-micrometer length scales
- **QM/MM:** Overcame the challenge taking cue from multiscale quantum-mechanics (QM)/molecular mechanics (MM)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

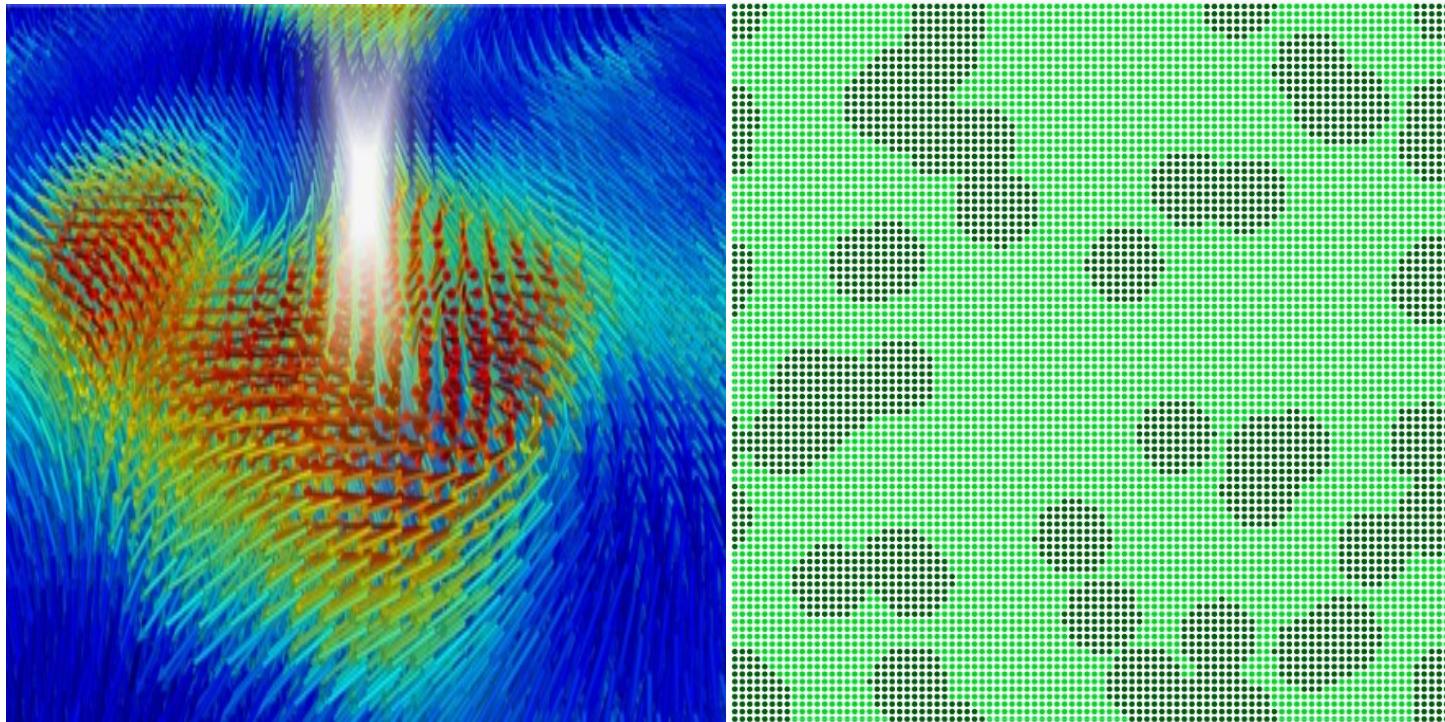
- **NN/MM:** NNQMD for ferroelectric (PbTiO_3 : PTO) embedded in MM for paraelectric (SrTiO_3 : STO) to apply appropriate strain boundary condition



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

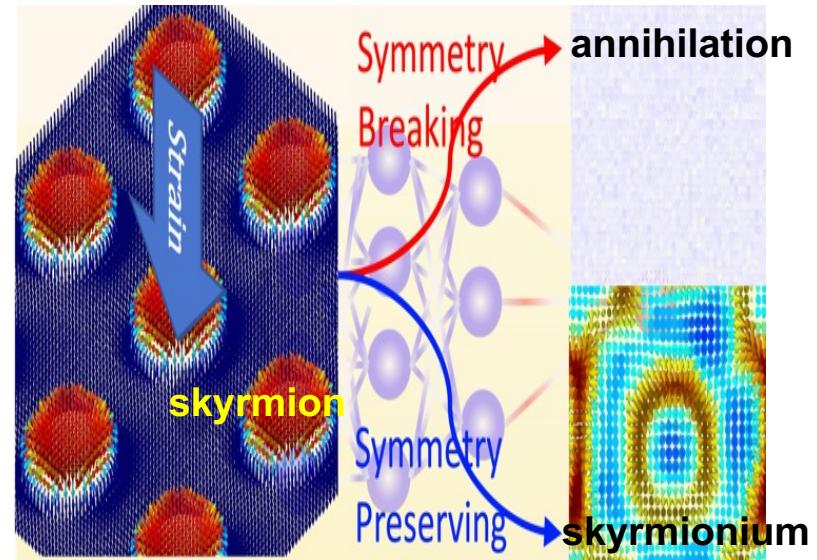
Application: Ferroelectric Opto-Toptronics



*ML for
large scale!*

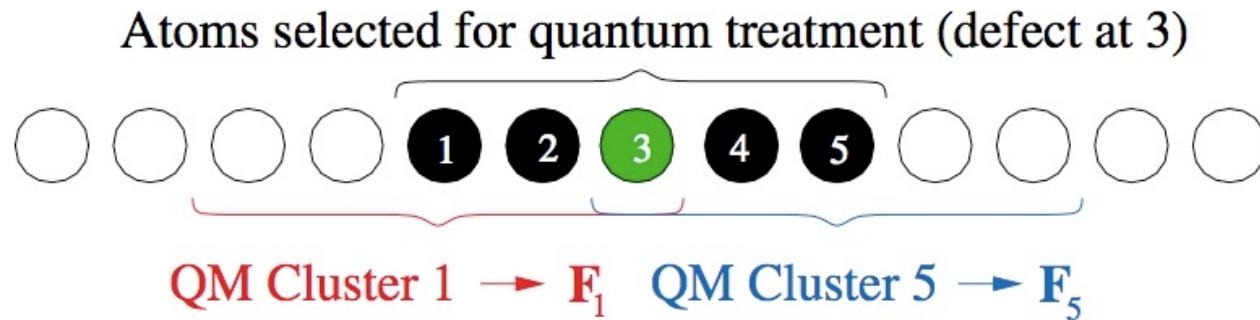
- Quantized ferroelectric topology is protected against thermal noise → future ultralow-power opto-electronics applications
- Billion-atom NNQMD revealed photo-induced topological phase-transition dynamics (*cf.* Kibble-Zurek mechanism in cosmology)
- Symmetry-controlled skyrmion-to-skyrmionium^{*} switching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

$$\begin{aligned} E(\mathbf{u}_k, \dot{\mathbf{u}}_k) &= \langle H_{MD} \rangle_{\mathbf{u}_k, \dot{\mathbf{u}}_k} \\ &= \int d\mathbf{x}_{\mu} d\mathbf{p}_{\mu} H_{MD} e^{-\beta H_{MD}} \Delta / Z, \\ \Delta &= \prod_j \delta\left(\mathbf{u}_j - \sum_{\mu} \mathbf{u}_{\mu} f_{j\mu}\right) \delta\left(\dot{\mathbf{u}}_j - \sum_{\mu} \frac{\mathbf{p}_{\mu} f_{j\mu}}{m_{\mu}}\right), \end{aligned}$$

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

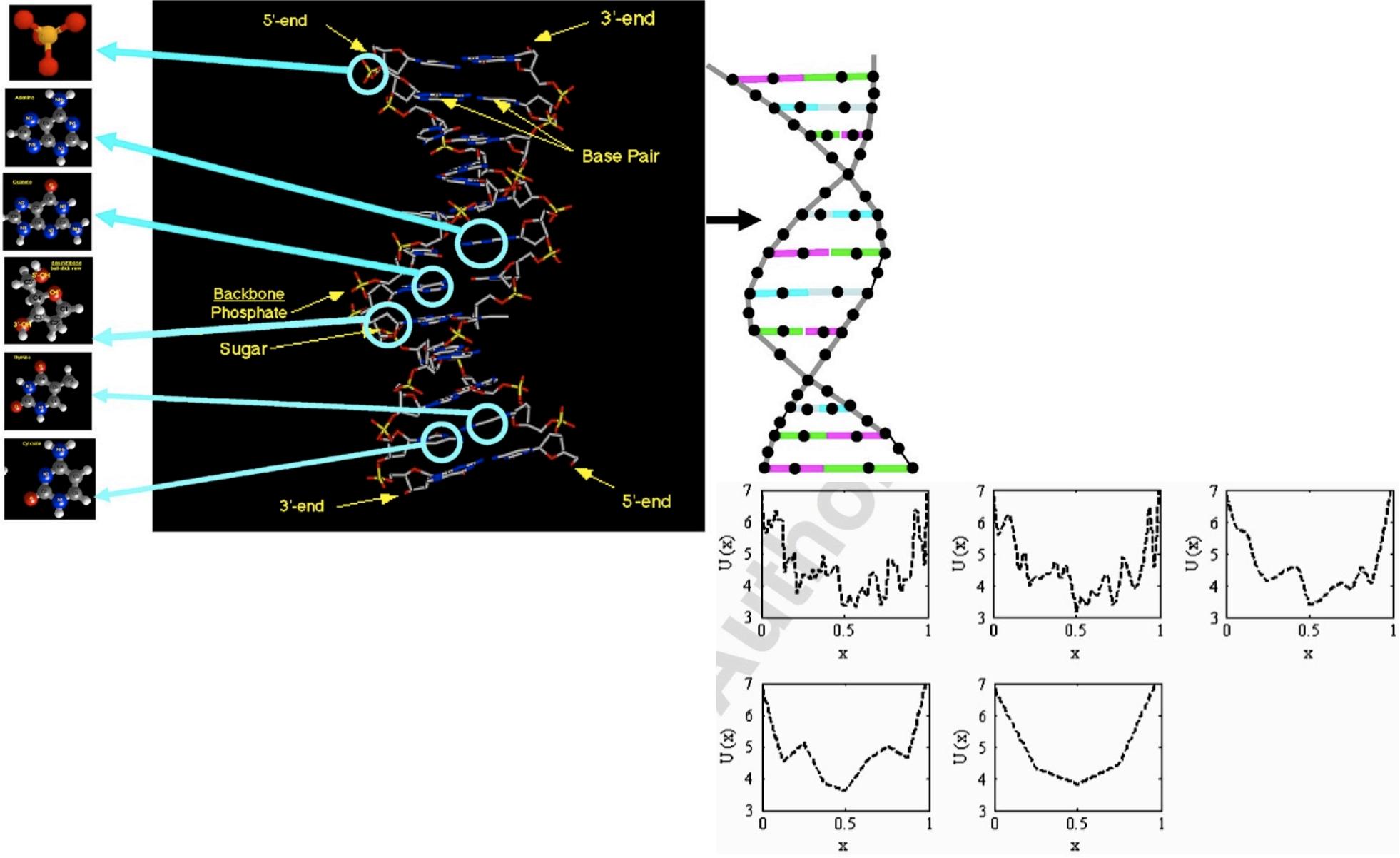


Fig. 11. Effective bond potential based on wavelet multi-scale projection of fine scale bond potential.

Quasicontinuum Method

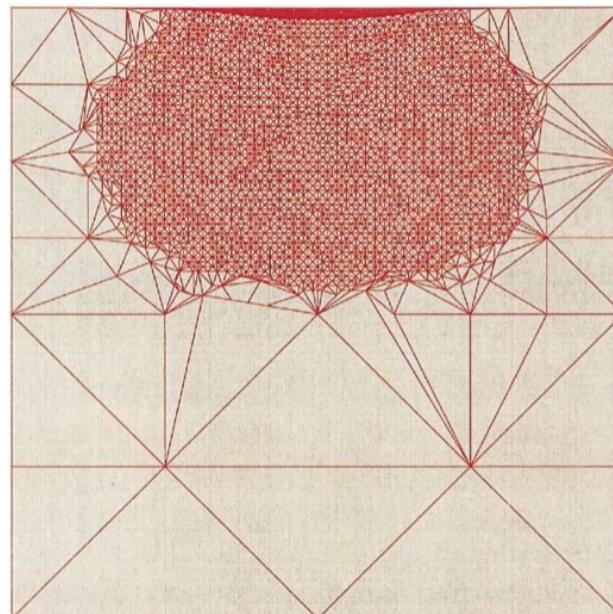
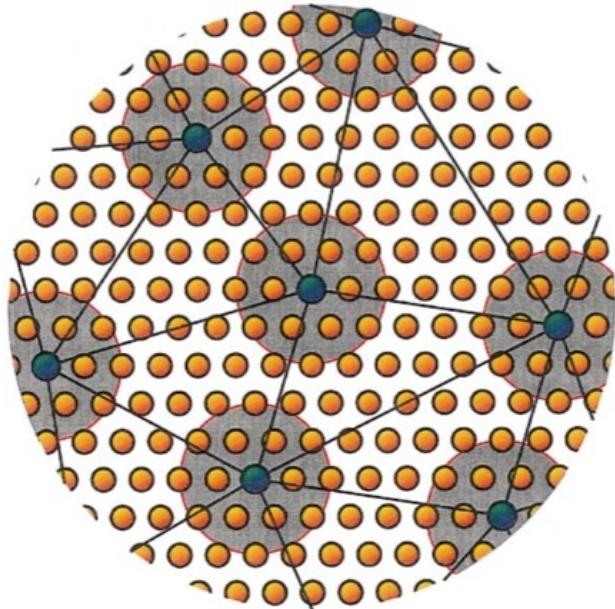
- Piecewise interpolation of the deformation of a selected subset of atoms

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

- 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

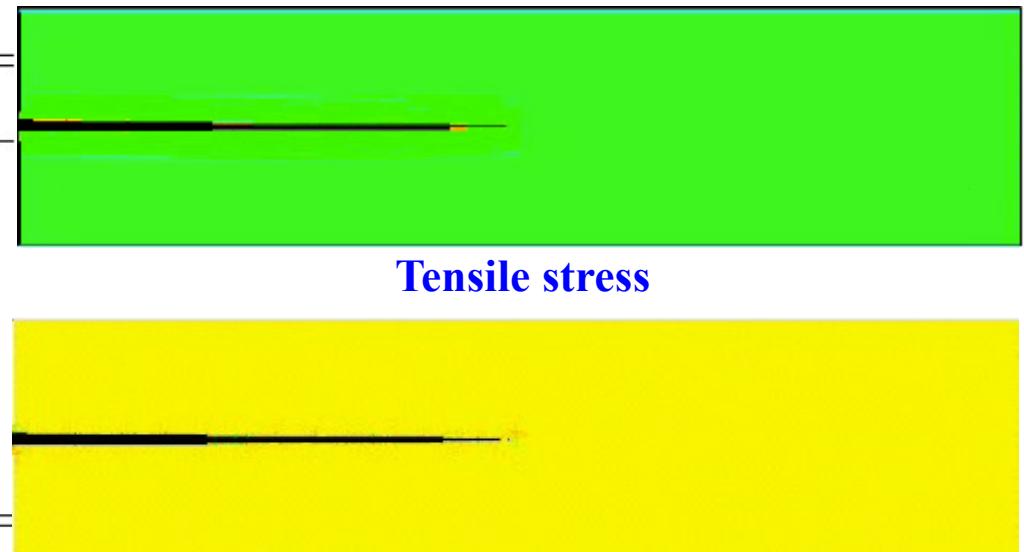
Iterate:

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)

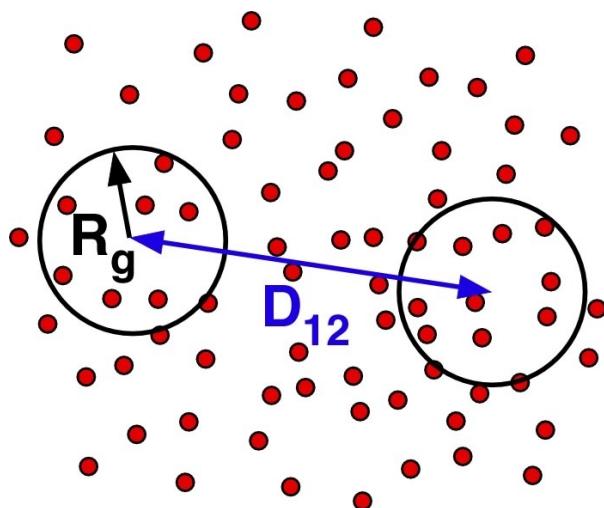
Dissipative Particle Dynamics

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

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

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

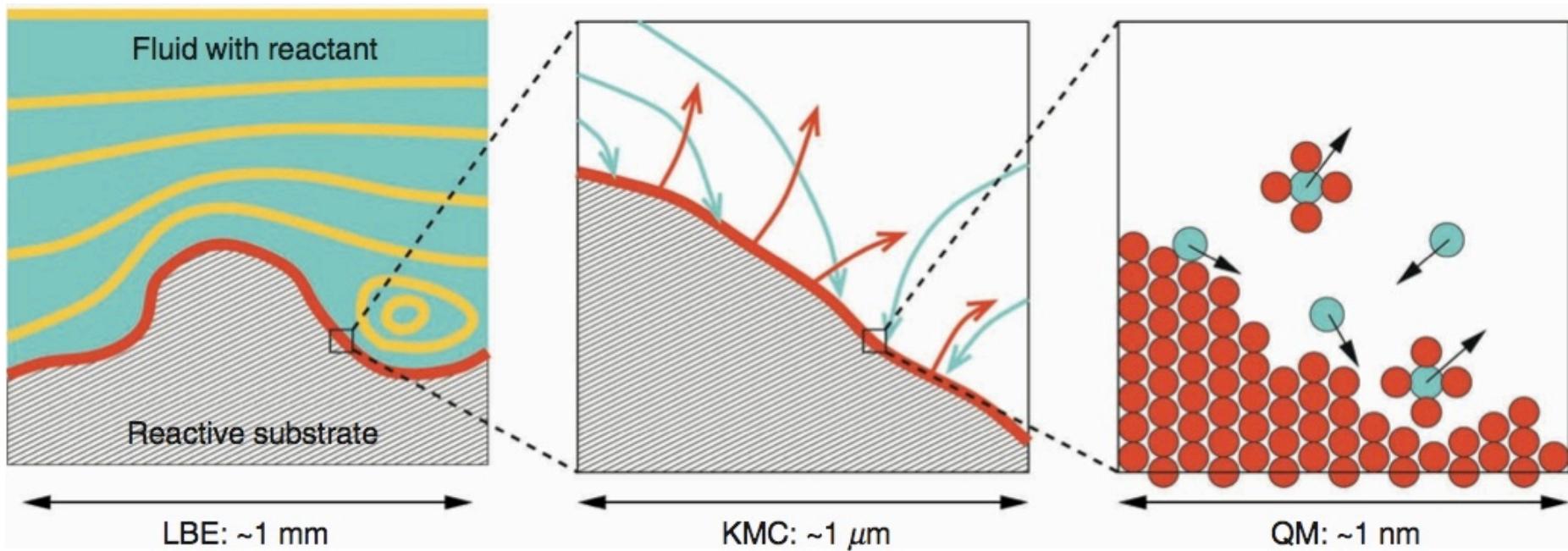
Kinjo & Hyodo, *Phys. Rev. E* 75, 051109 ('07)



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

Lattice Boltzmann + Atomistic

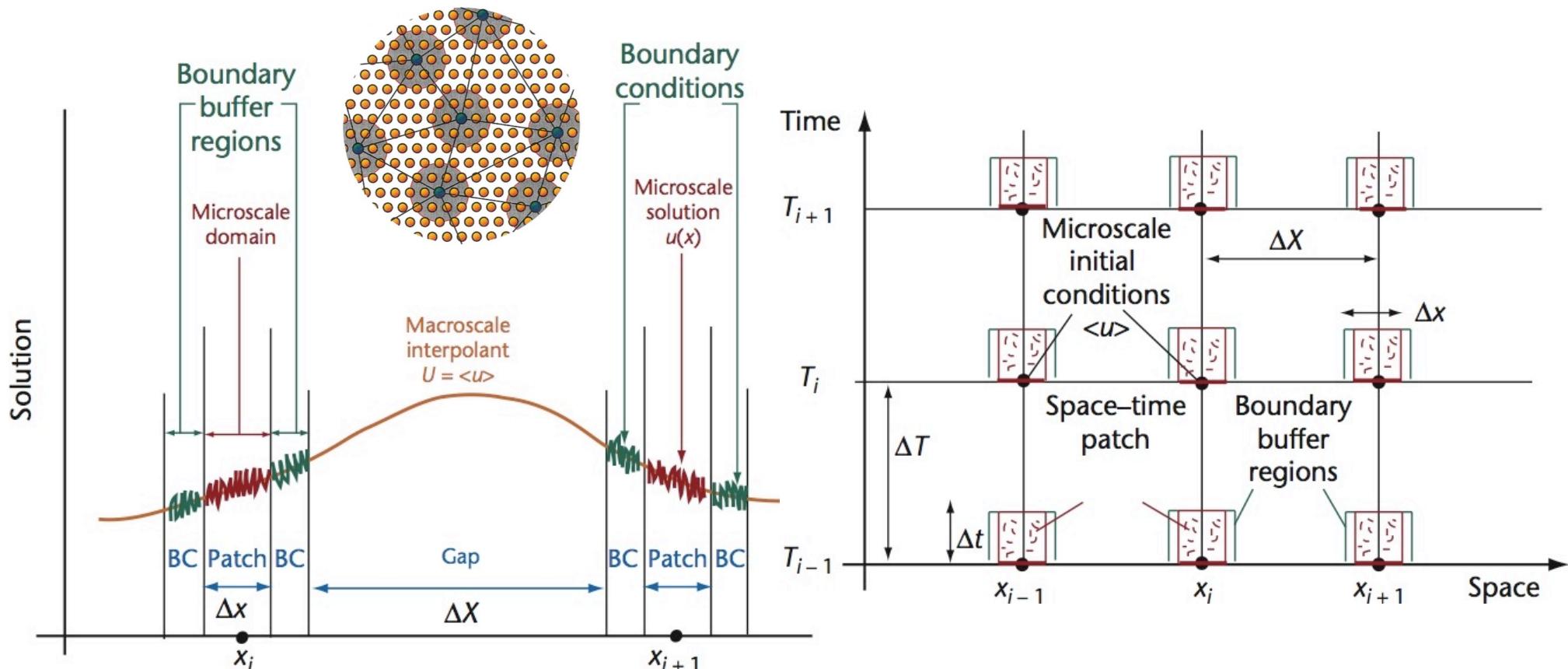
- Coupling fluid dynamics + atomistics/chemical reactions
- Possible breakdown of hydrodynamics at small length scales:
Boltzmann equation for $f(x, v, t)$



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)