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

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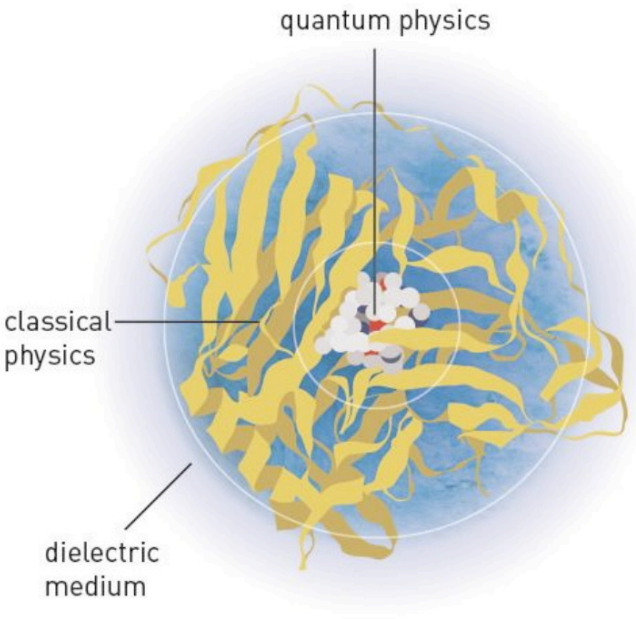
Michael Levitt

Photo: Wikimedia Commons

Arieh Warshel



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

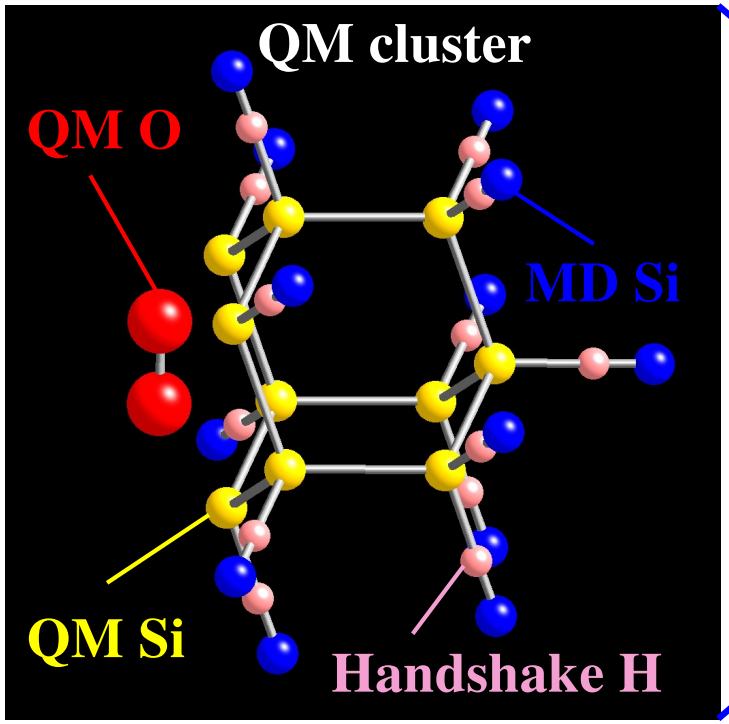
The Nobel Prize in Chemistry 2013 was awarded jointly to Martin Karplus, Michael Levitt and Arieh Warshel *"for the development of multiscale models for complex chemical systems"*.

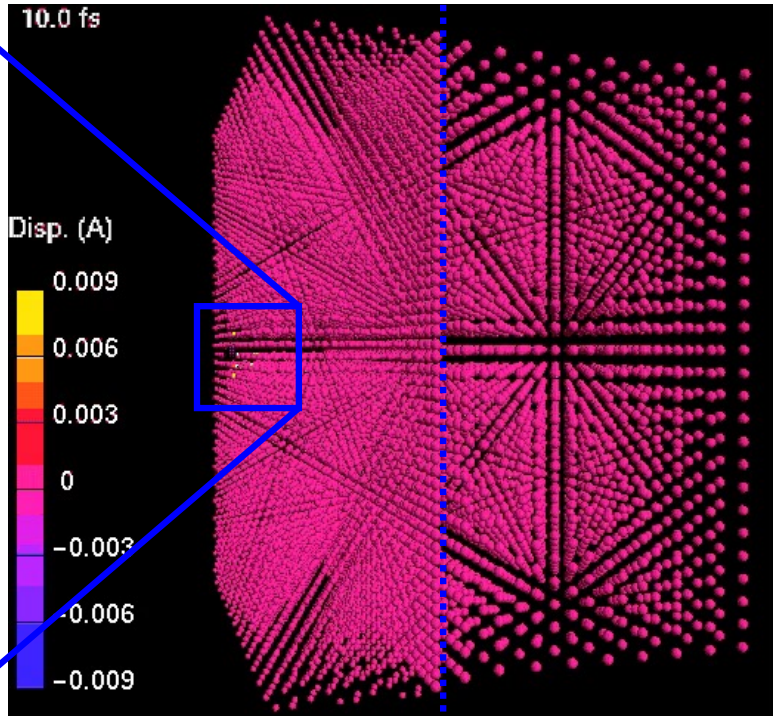
A. Warshel & M. Karplus, *J. Am. Chem. Soc.* **94**, 5612 ('72)
A. Warshel & M. Levitt, *J. Mol. Biol.* **103**, 227 ('76)

Find multiscale modeling in your area!

Adaptive Multiscale Dynamics

QMD MD FED





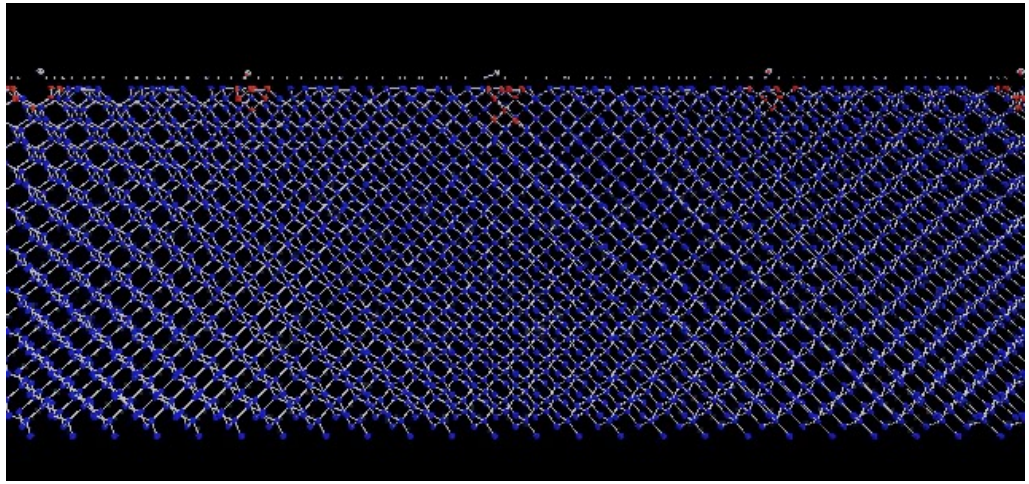
High-energy beam oxidation of Si (SIMOX)

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

Oxidation of Si

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

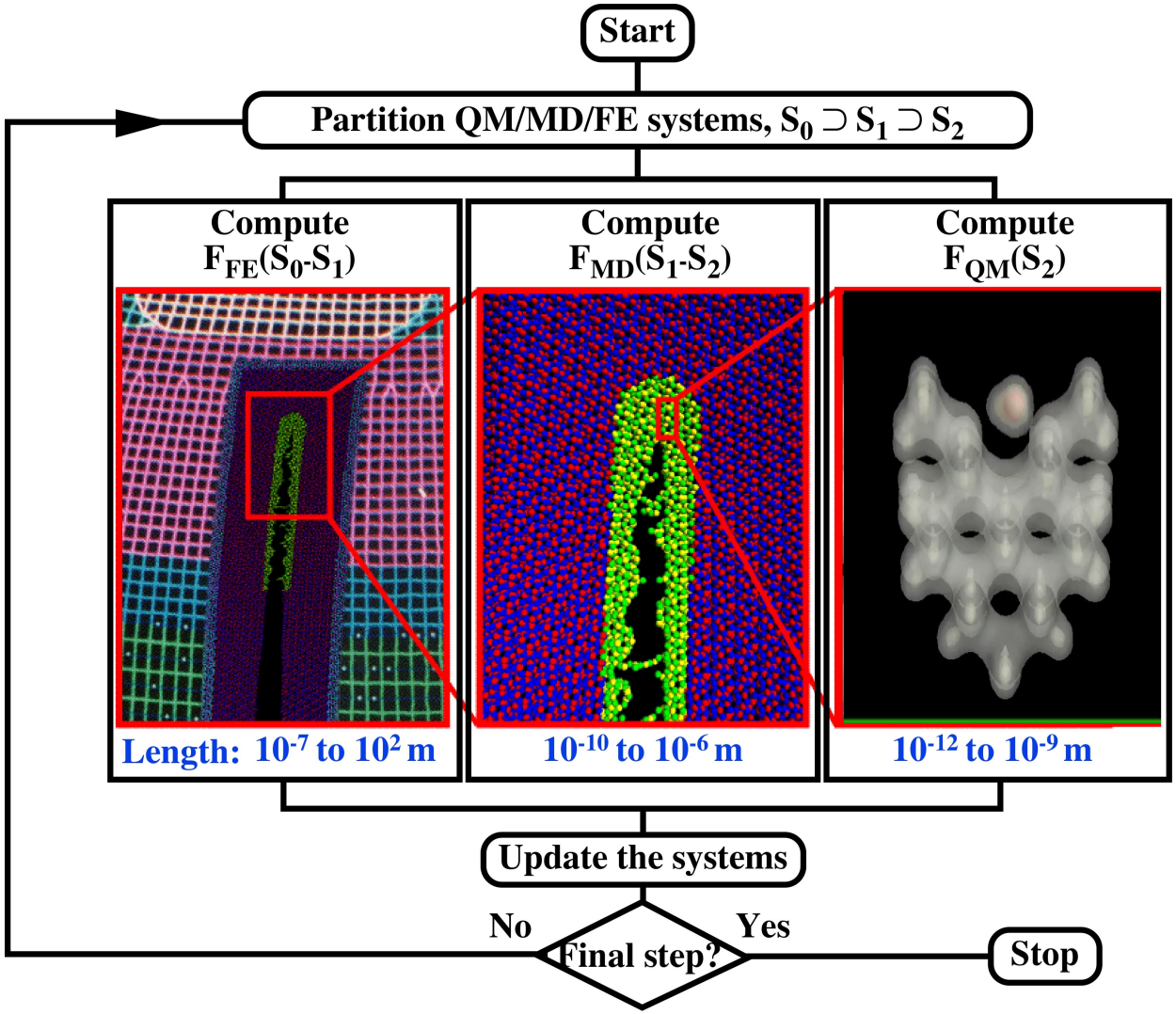
L. Lidorikis *et al.*, *Phys. Rev. Lett.* **87**, 086104 ('01)



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)

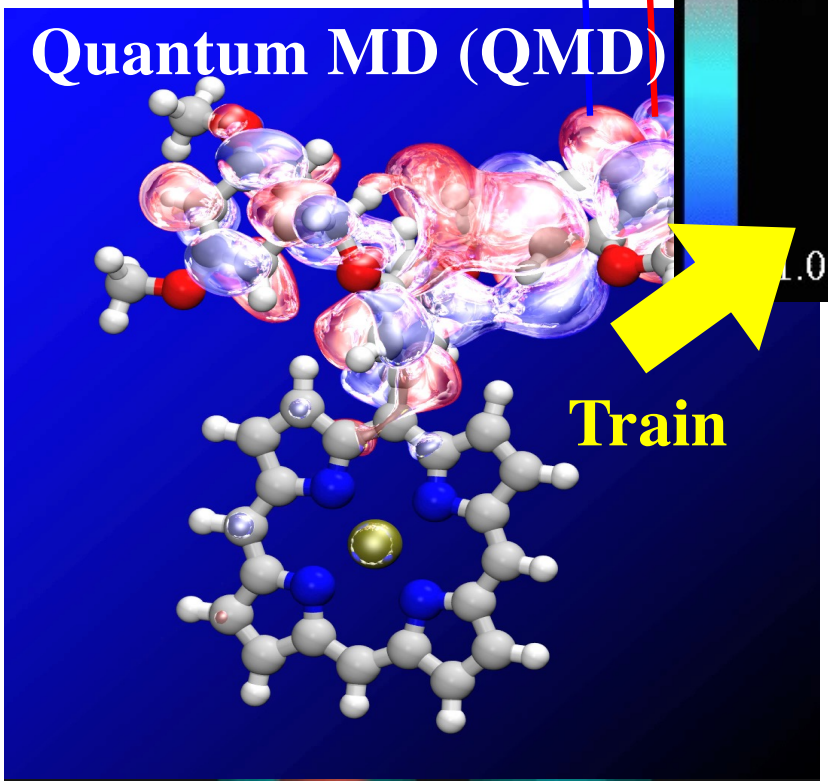
Hierarchical Atomistic Simulation Methods

$$m_i \frac{d^2\mathbf{r}_i}{dt^2} = -\frac{\partial}{\partial \mathbf{r}_i} E_{MD}(\{\mathbf{r}_i\})$$

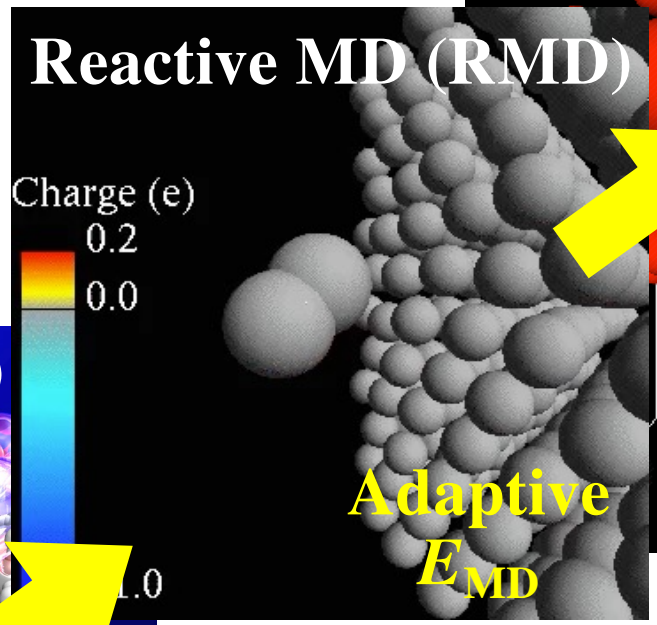
Atom

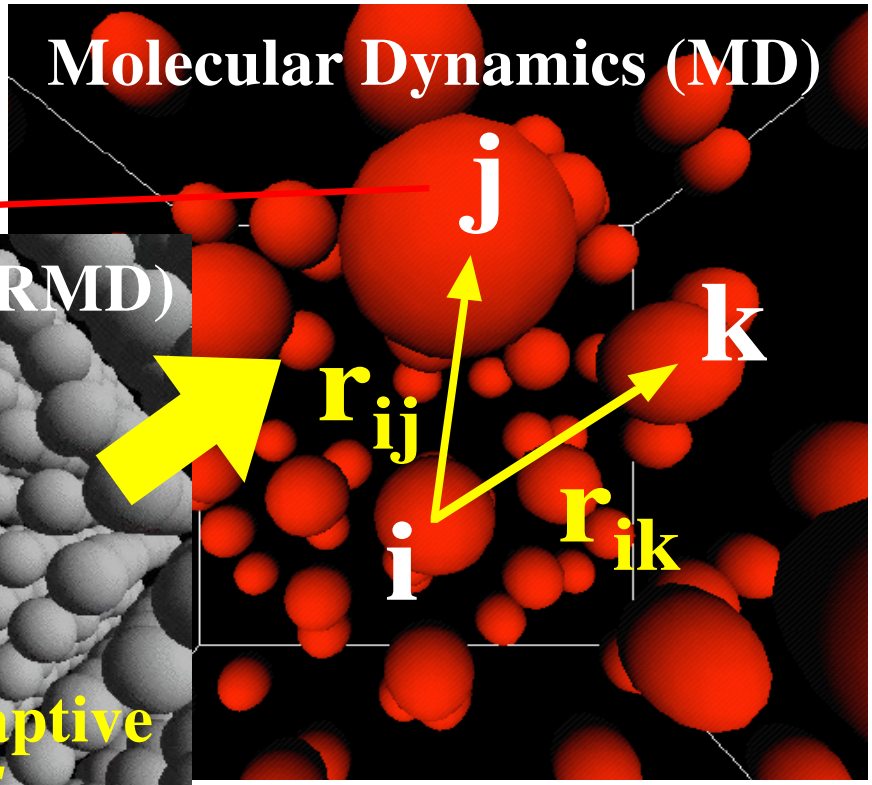
Electron wave function

$$\min E_{\mathrm{QM}}(\{\psi_n(\mathbf{r})\})$$



Train





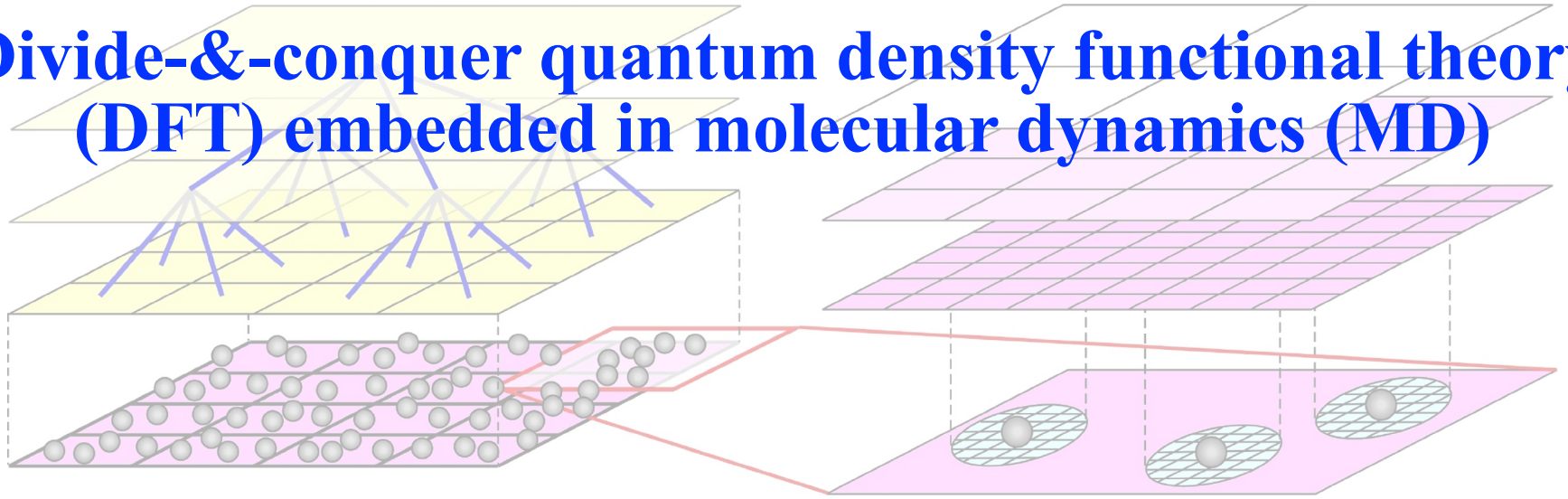
First principles-based reactive force-fields

- **Reactive bond order $\{BO_{ij}\}$**
  → **Bond breakage & formation**
- **Charge equilibration (QEq) $\{q_i\}$**
  → **Charge transfer**

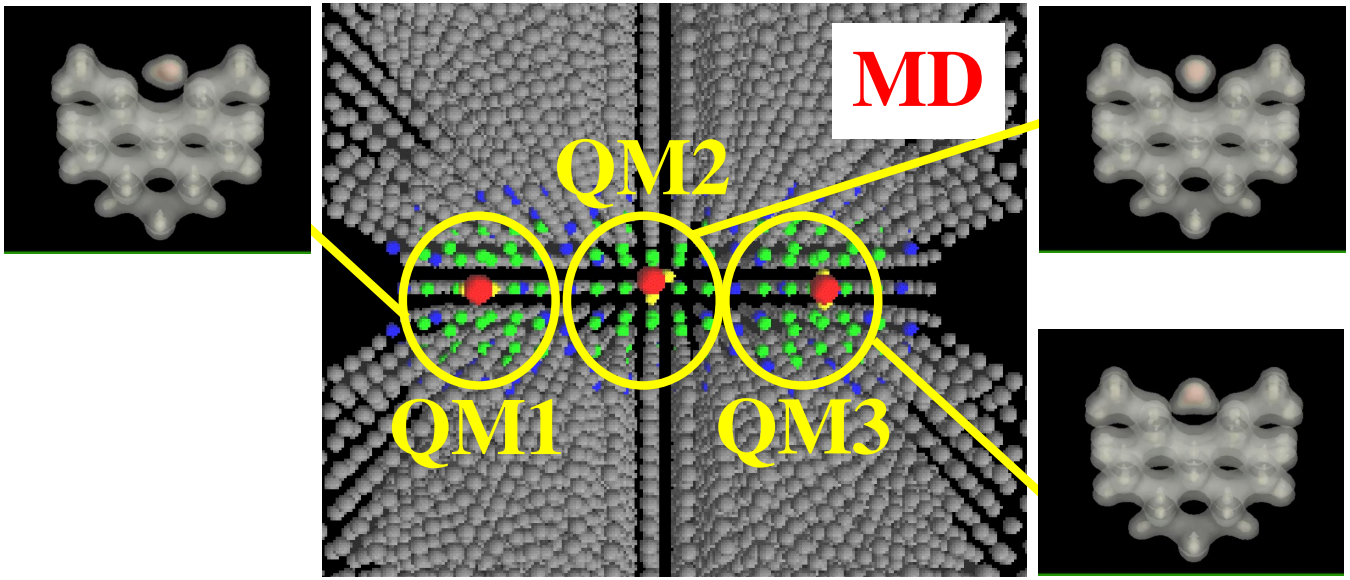
[Brenner; Streitz & Mintmire; van Duin & Goddard; Vashishta *et al.*]

DC Multiscale MD/QM (DFT)

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



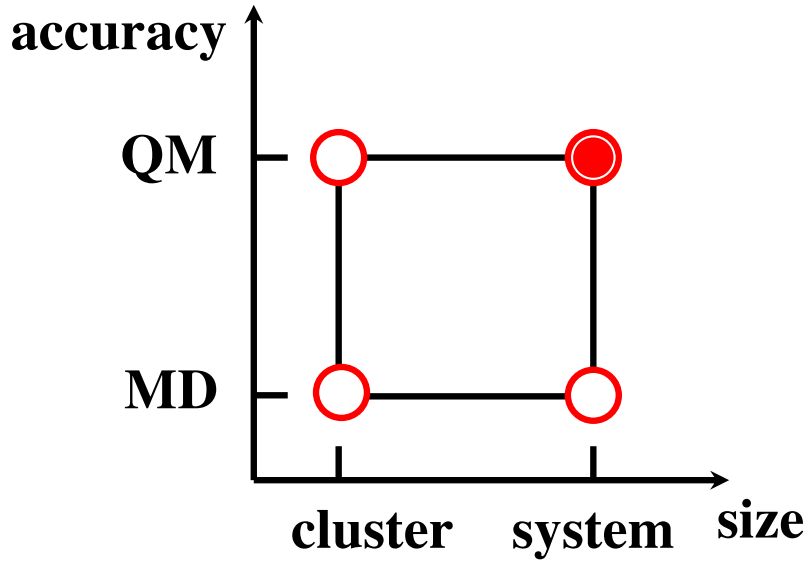
$$E_{\mathrm{MD/QM}} = E_{\mathrm{MD}}^{\mathrm{system}} + \sum_{\mathrm{cluster}} [E_{\mathrm{QM}}^{\mathrm{cluster}}(\{\mathbf{r}_{\mathrm{QM}}\},\{\mathbf{r}_{\mathrm{HS}}\}) - E_{\mathrm{MD}}^{\mathrm{cluster}}(\{\mathbf{r}_{\mathrm{QM}}\},\{\mathbf{r}_{\mathrm{HS}}\})]$$



Additive Hybridization

Additive hybridization [Morokuma *et al.*, '96]

- **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**

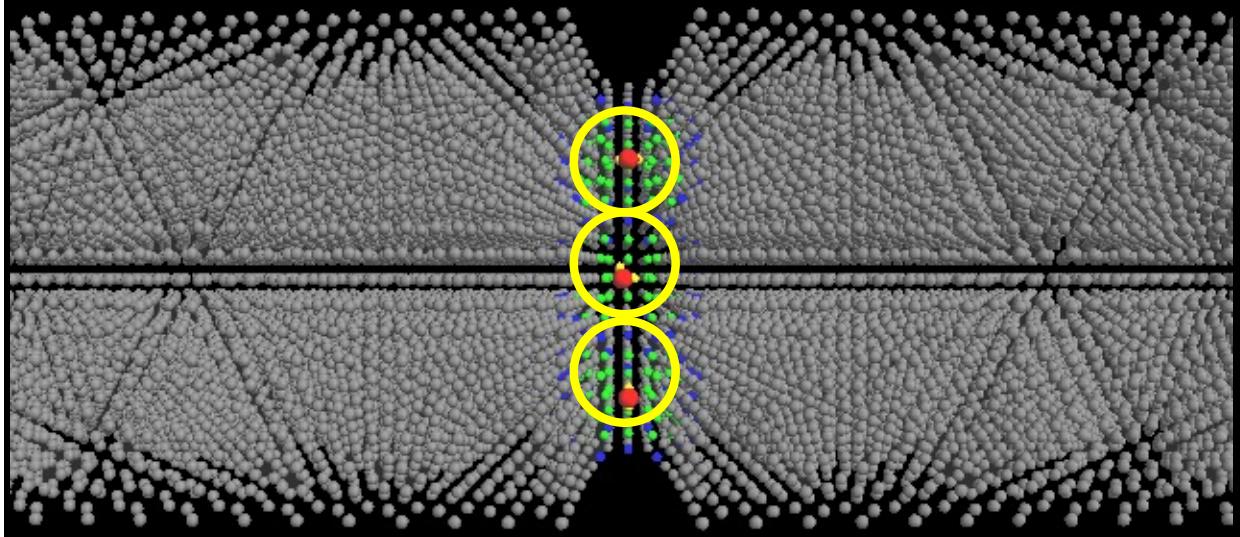


$$E_{\mathrm{QM}}(s) - E_{\mathrm{QM}}(c) \cong E_{\mathrm{MD}}(s) - E_{\mathrm{MD}}(c)$$

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

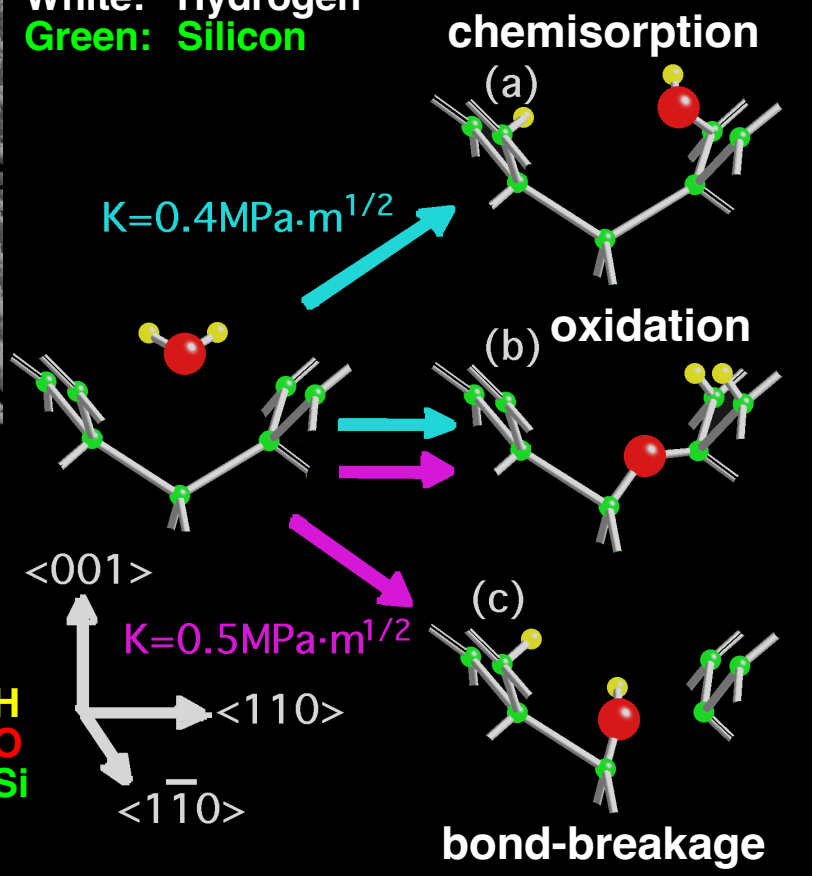
Environmental Effect on Fracture

Reaction of H_2O molecules at a Si crack tip



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

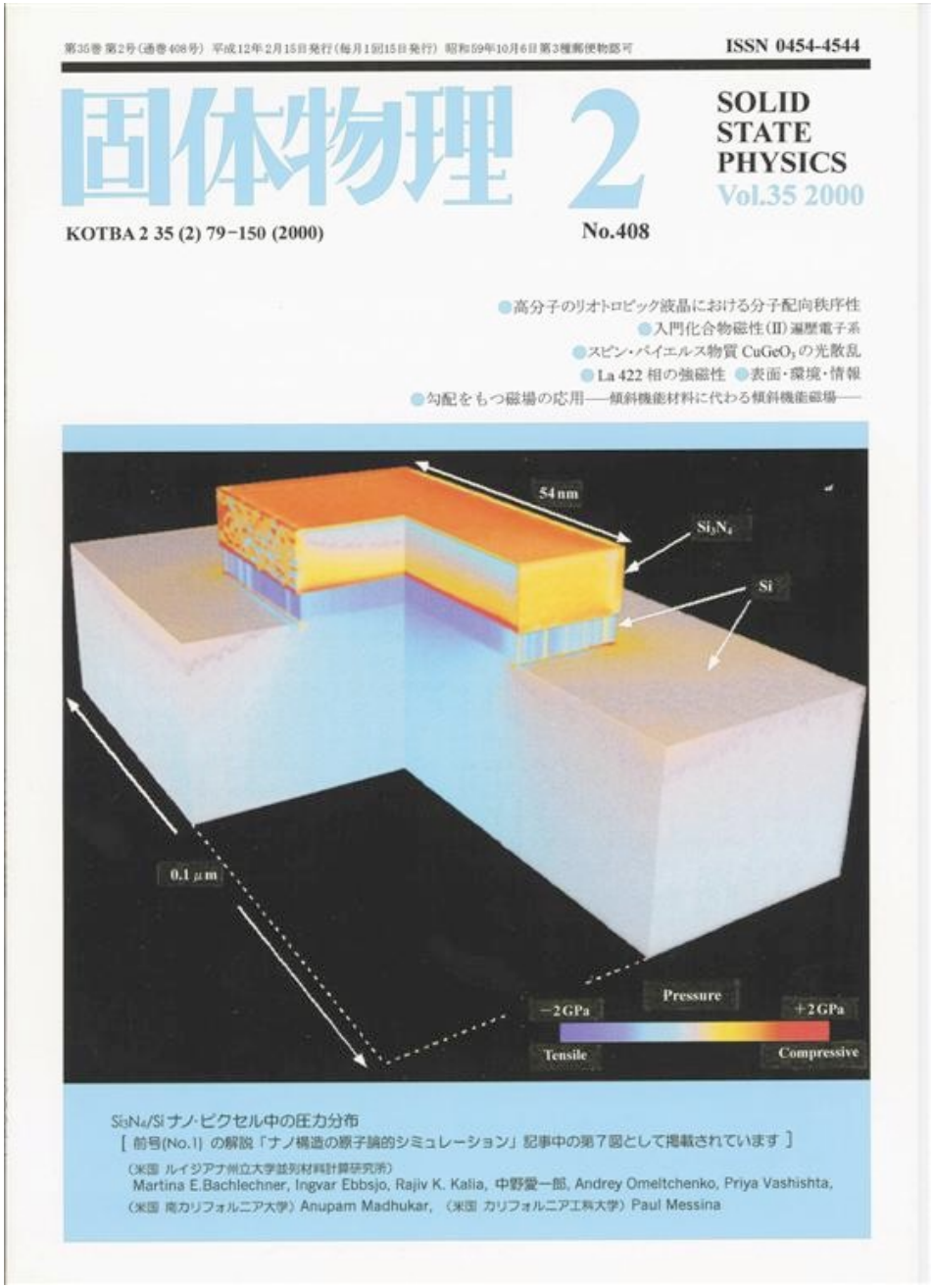
Blue: Oxygen
White: Hydrogen
Green: Silicon



Yellow: H
Red: O
Green: Si

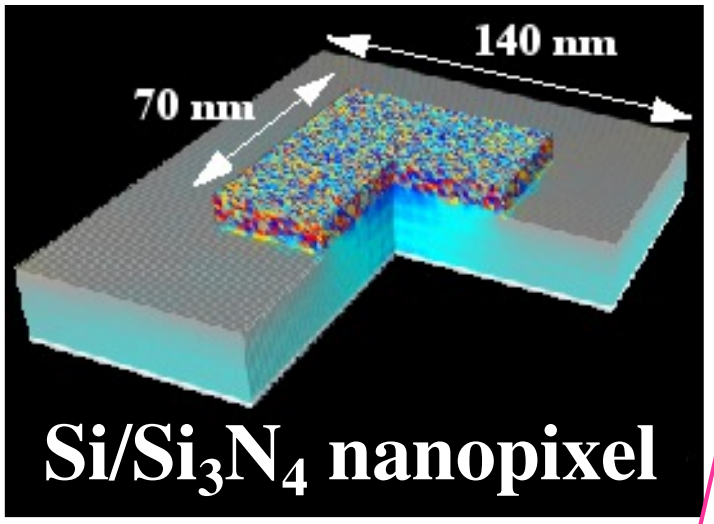
Significant dependence of the reaction on stress intensity factor

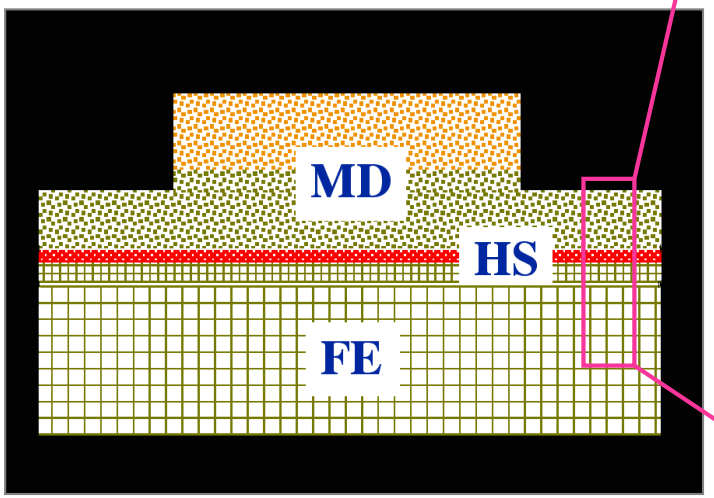
Atomistic Simulations of Nanodevices

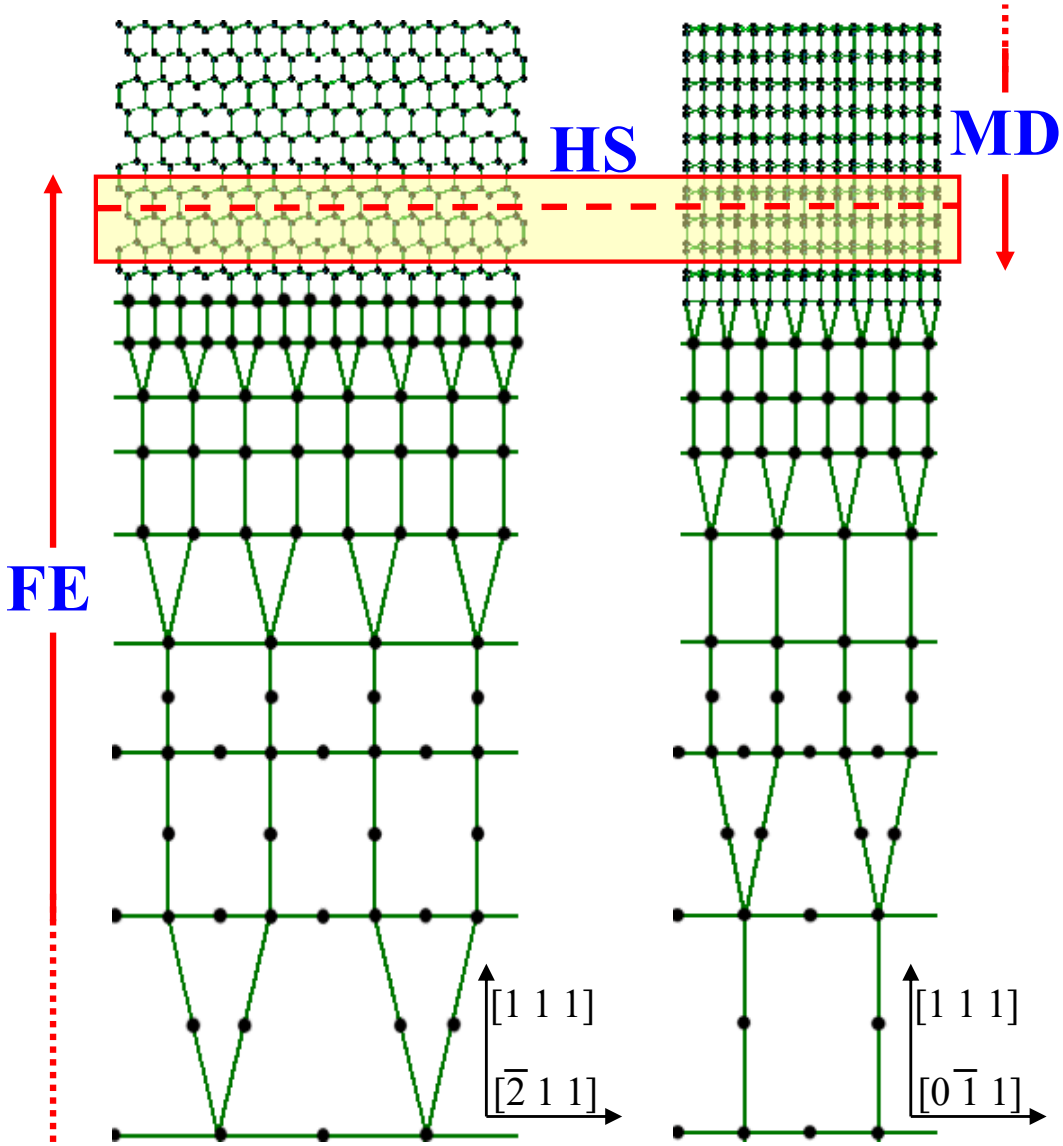


Hybrid FE/MD Algorithm

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

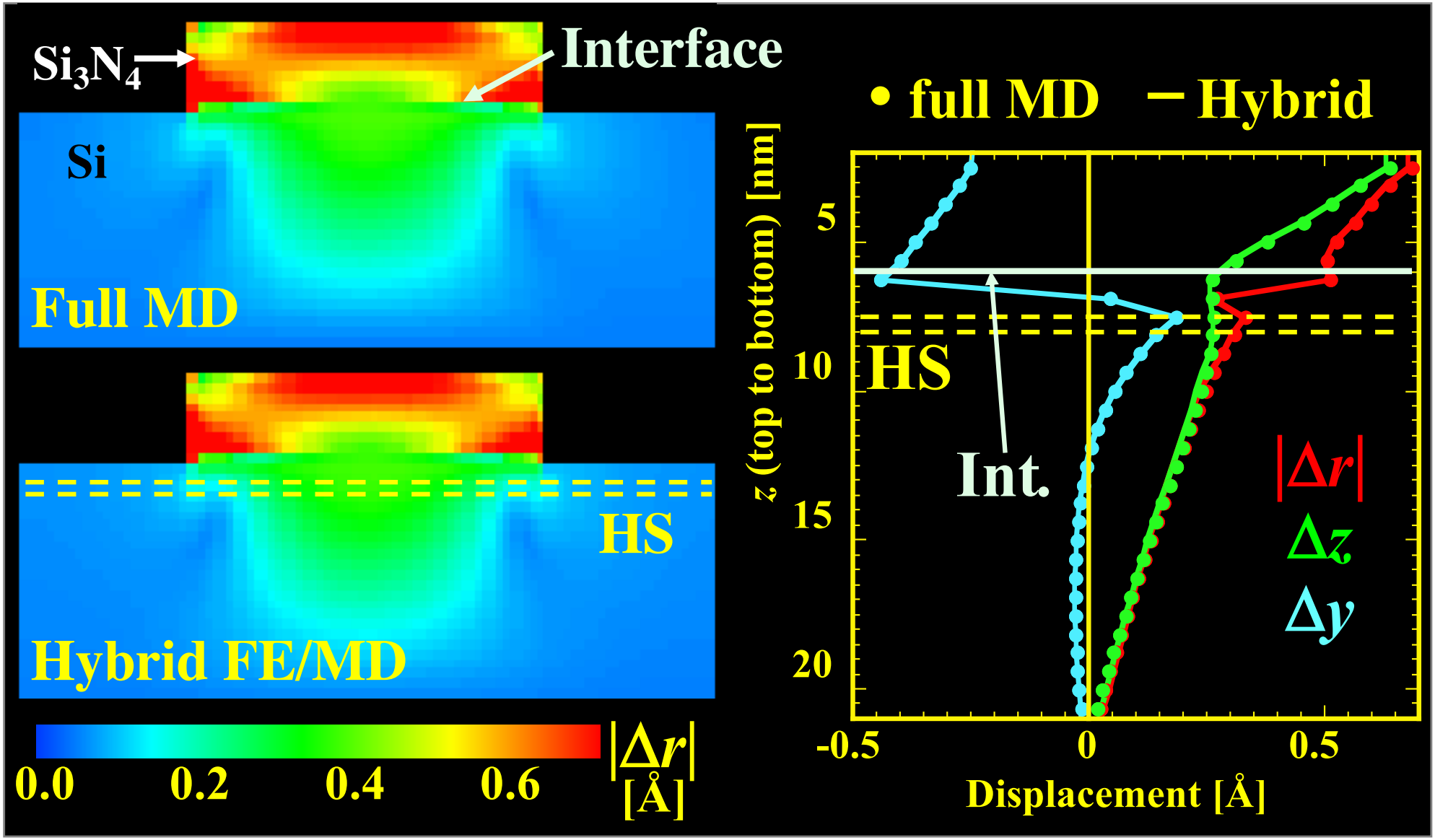




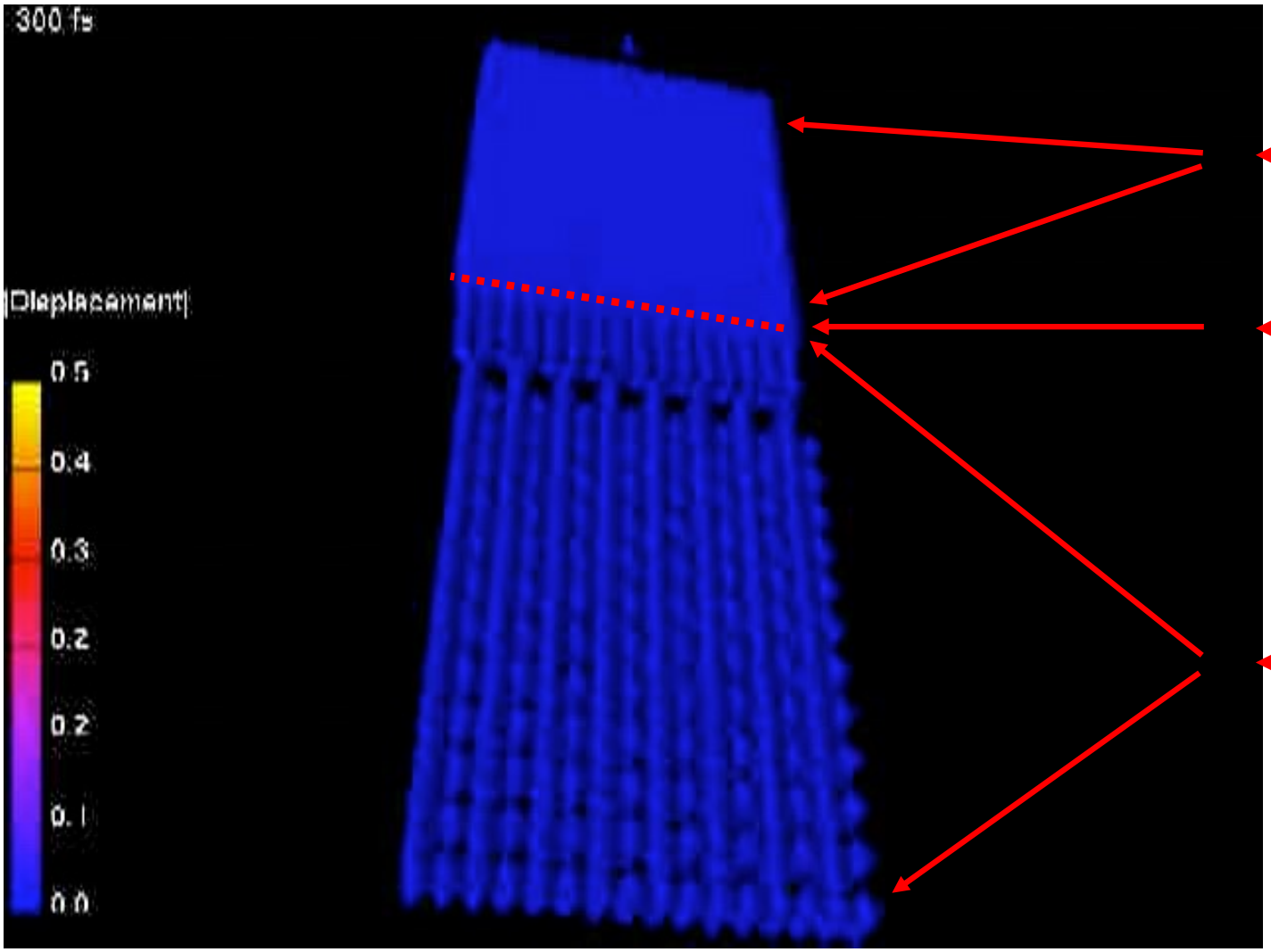


$Si(111)/Si_3N_4(0001)$ Nanopixel

Displacement from equilibrium positions



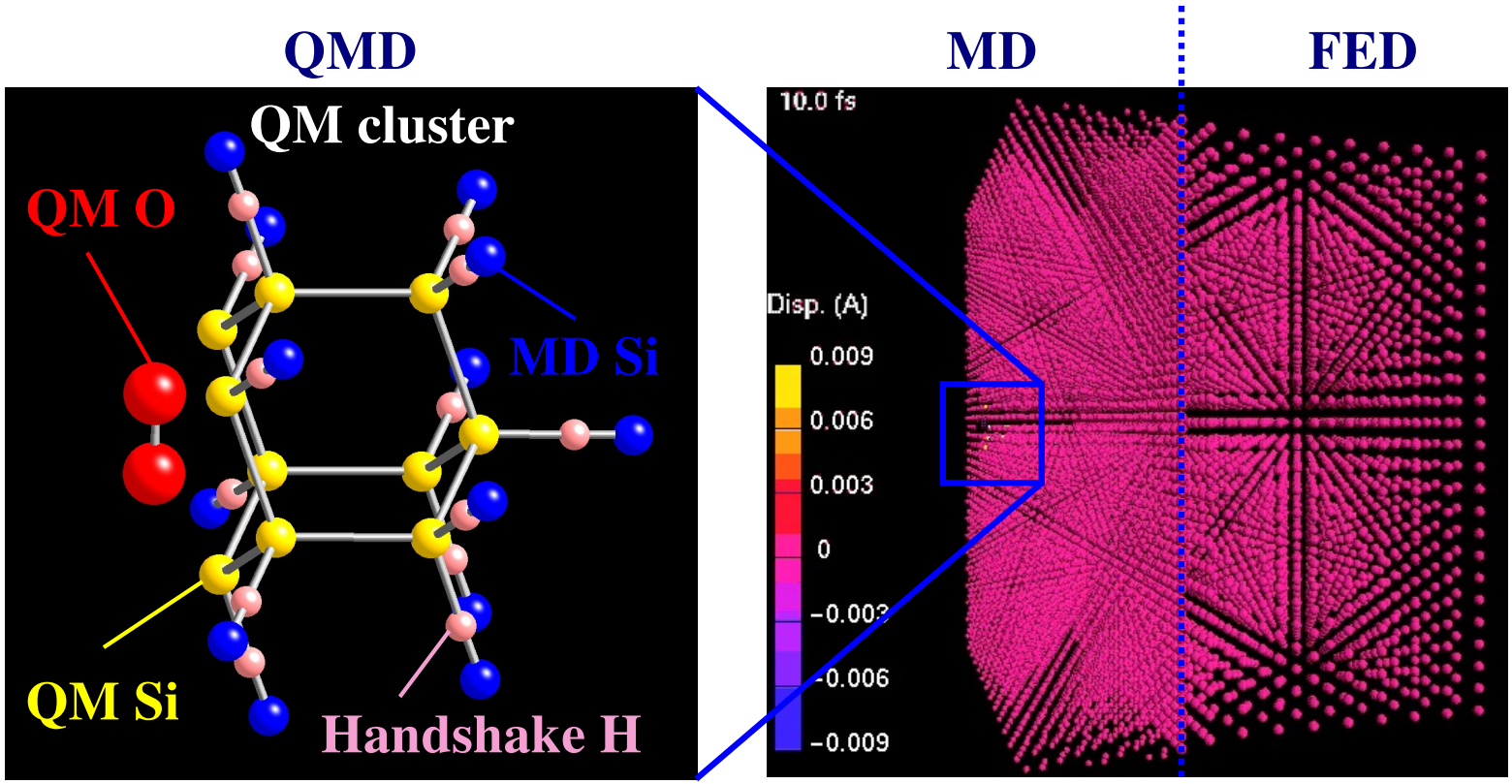
Dynamics Test & Demonstration: Projectile Impact on Silicon



Waves propagate seamlessly into the FE region

Application of Multiscale Simulations

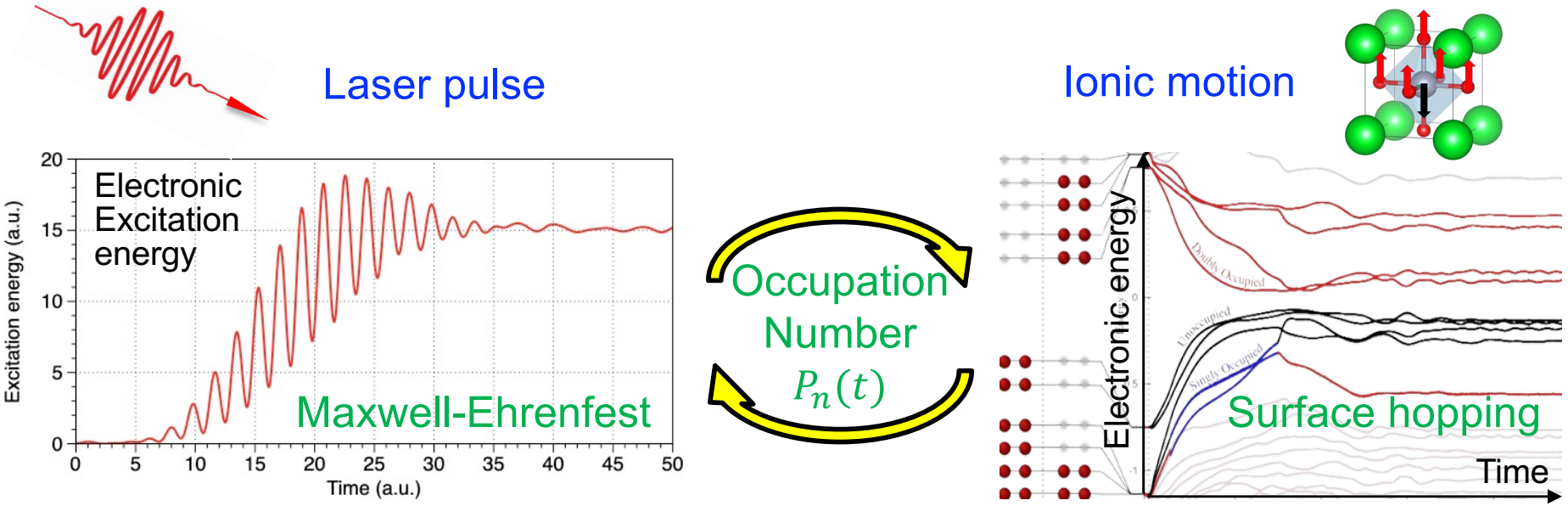
Oxidation dynamics on Si surface



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

Light-Matter Interaction: DC-MESH

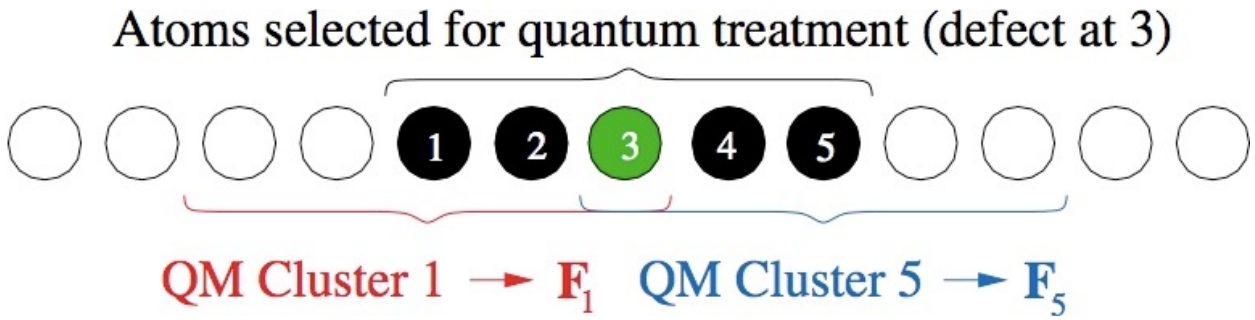
- **DC-MESH (divide-&-conquer Maxwell + Ehrenfest + surface-hopping): *O*(*N*) algorithm to simulate photo-induced quantum materials dynamics**
- **LFD (local field dynamics): Solve Maxwell equations for light & real-time time-dependent density functional theory (RT-TDDFT) equations for electrons to 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**



T. Linker *et al.*, *Science Advances* **8**, eabk2625 ('22)

"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**



G. 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] J. Behler & M. Parrinello, *Phys. Rev. Lett*. **98**, 146401 ('07); *IJQC* **115**, 1032 ('15)
[2] A. P. Bartok *et al*., *Phys. Rev. Lett*. **104**, 136403 ('10)
[3] L. Zhang *et al*., *Phys. Rev. Mater*. **3**, 023804 ('19)
[4] J. 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**

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

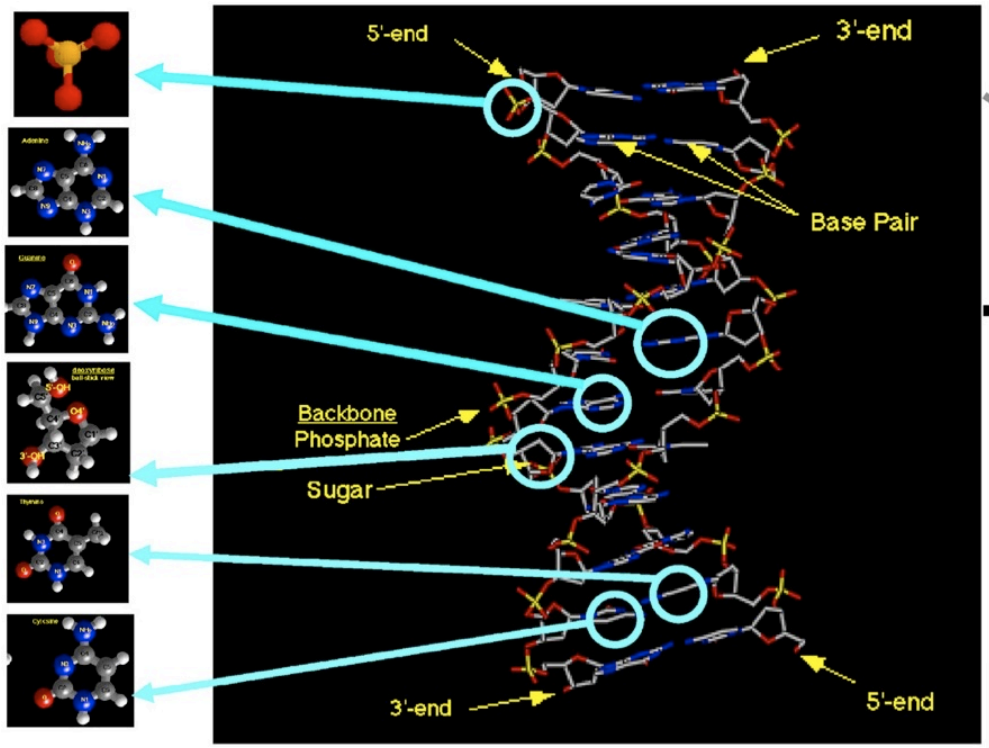
$$= \int d\mathbf{x}_\mu d\mathbf{p}_\mu H_{MD} e^{-\beta H_{MD}} \Delta / Z,$$

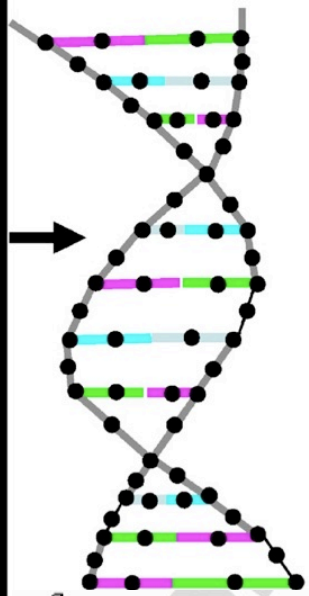
$$\Delta = \prod_j \delta\left(\mathbf{u}_j - \sum_\mu \mathbf{u}_\mu f_{j\mu}\right) \quad \delta\left(\dot{\mathbf{u}}_j - \sum_\mu \frac{\mathbf{p}_\mu f_{j\mu}}{m_\mu}\right),$$

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





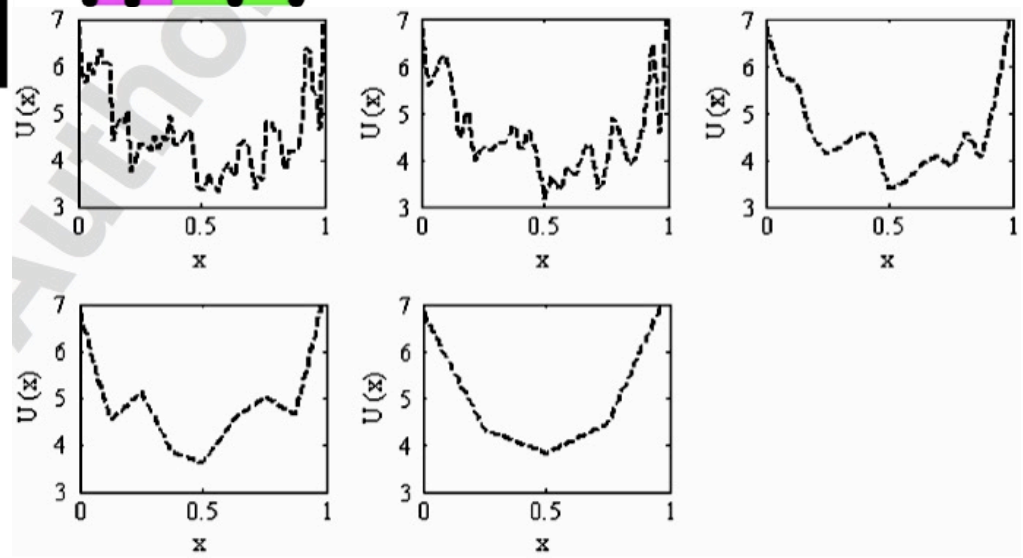


Fig. 11. Effective bond potential based on wavelet multi-scale projection of fine scale bond potential.

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

Quasicontinuum Method

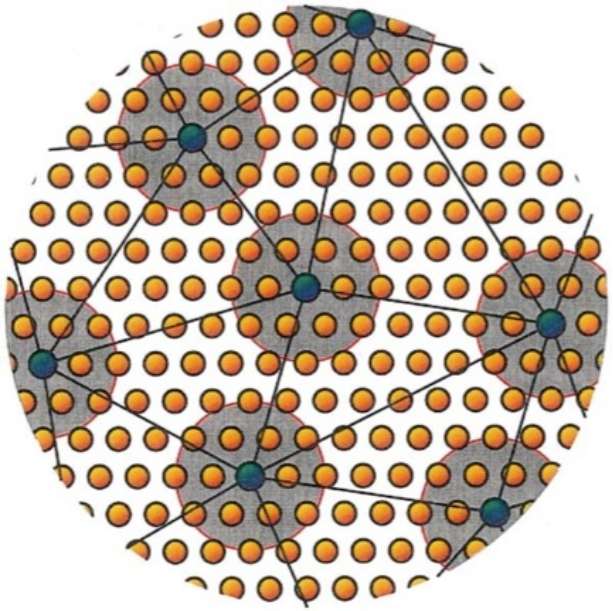
- **Piecewise interpolation of the deformation of a selected subset of atoms**

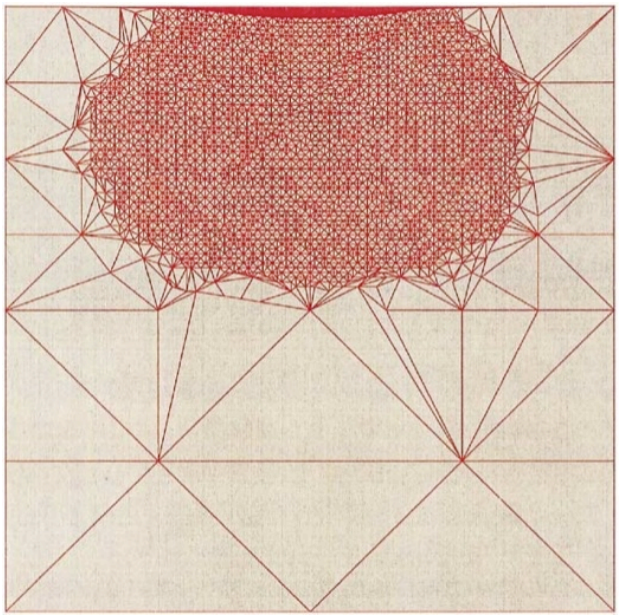
$$\mathbf{x}_\mu = \sum_j N_j(\mathbf{x}_\mu)\,\mathbf{x}_j$$

- **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"**





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

Linear-Scaling MD/FE Optimization

Iterate:

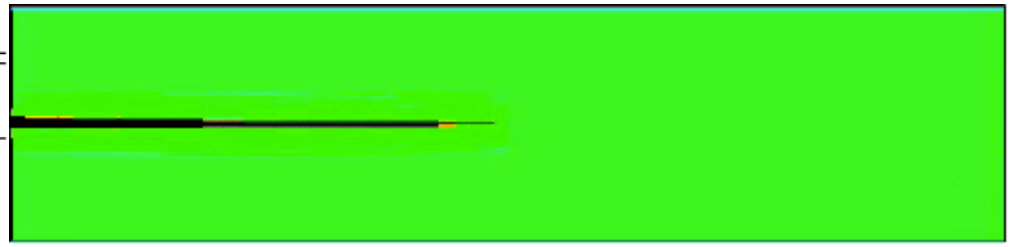
1. Perform a small # of steps of conventional (*e.g.*, CG) energy minimization
2. Perform one step where solid is described by elastic theory
   a. Calculate the forces acting on the atoms
   b. Transfer the atomic forces onto a computational grid
   c. Solve linear elastic eq. with a multigrid method 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	↔	117	5586.0	1147.0

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

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



Tensile stress

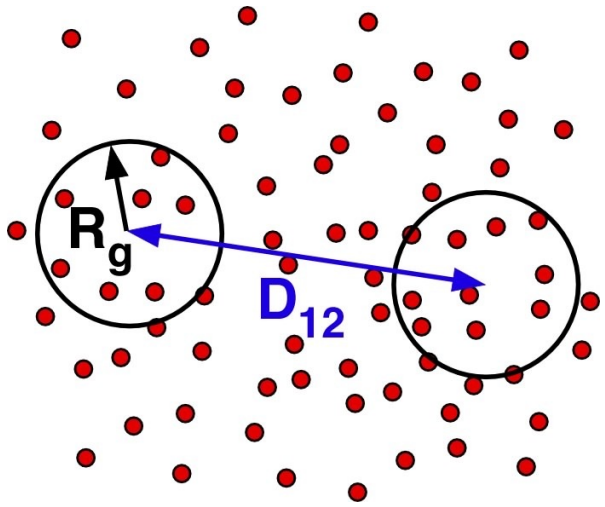
Shear stress

Dissipative Particle Dynamics

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

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

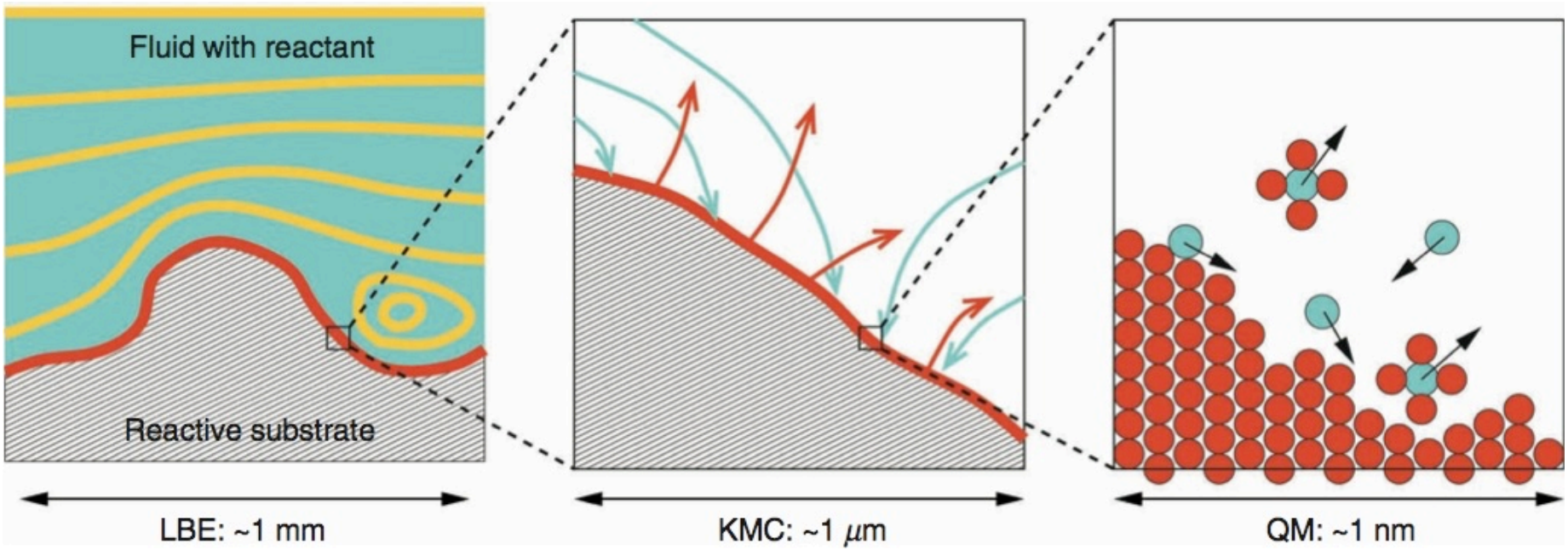
- **Generalized Langevin equation (Liouville equation & Mori-Zwanzig projection operator) for first-principles derivation of coarse forces**
  [T. Kinjo & S. Hyodo, *Phys. Rev. E* **75**, 051109 ('07)]



R. D. Groot & P. B. Warren, *J. Chem. Phys.* **107**, 4423 ('97)

Lattice Boltzmann + Atomistic

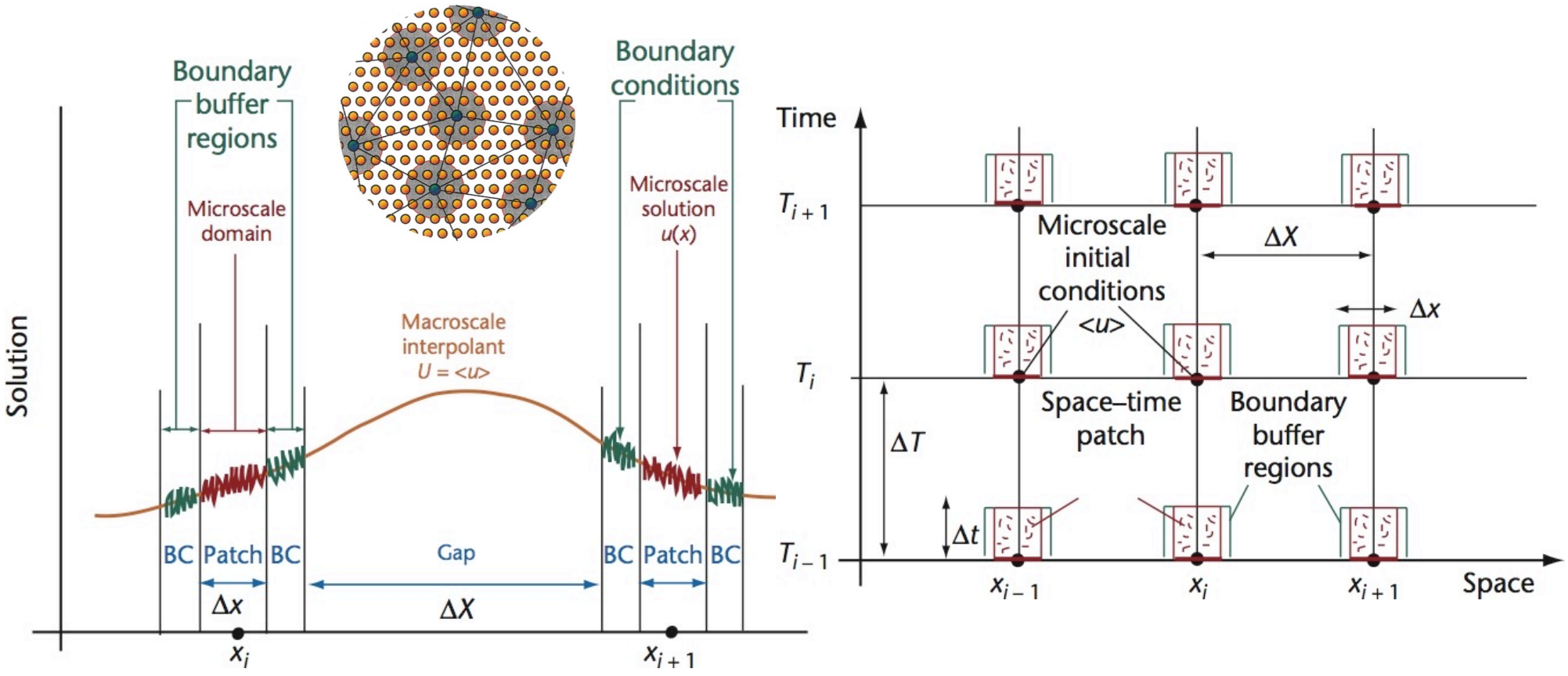
- **Coupling fluid dynamics + atomistics/chemical reactions**
- **Possible breakdown of hydrodynamics at small length scales: Boltzmann equation for $f(x,v,t)$**



S. Succi, O. Filippova, G. Smith & E. Kaxiras, *Comp. Sci. Eng.* **3**(6), 26 ('01)
Y. 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**



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