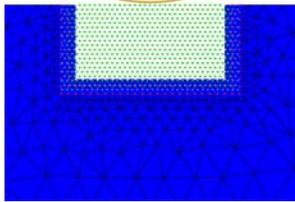


Coupling Atomistic and Continuum Descriptions in Multiscale Modeling

Mark Robbins, Johns Hopkins University

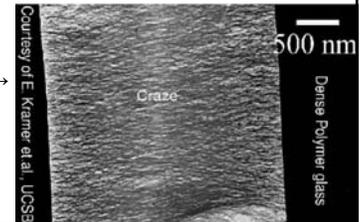
Collaborators: B. Luan, X. Nie, S. Hyun, J. Rottler
S. Chen, J. F. Molinari, J. A. Harrison, N. Bernstein
Sponsor: National Science Foundation DMR and CMS
2007 DOE Summer School in Multiscale Mathematics and High Performance Computing, Corvallis, OR, July 1, 2007



Why Multiscale?

Behavior controlled by processes on wide range of lengths and times
0.1 to 1 nm – chemical bond, atomic size, polymer diameter, ...
1 nm to 1 μ m – size of colloidal particle, polymer, actin, ...
size of defect structures: dislocations, fibrils, domain walls, ...
1 μ m to 1 mm – microstructure – collections of defects, domains, scale of cells, grains, pores, ...
large scales – homogeneous continuum with complex geometry

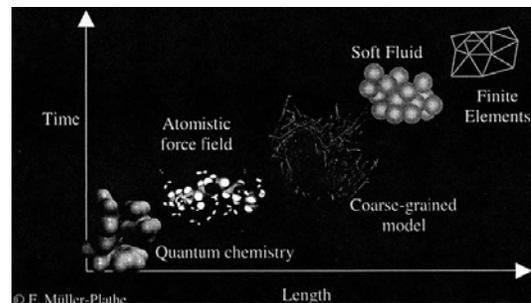
Polymer fracture:
nm polymer \rightarrow 10 nm fibril \rightarrow
10 μ m craze \rightarrow 10 mm crack



Why Multiscale?

- Behavior controlled by processes on wide range of lengths and times
- Want to include relevant physics from all scales
- Usual approach \rightarrow Choose scale of interest and use appropriate method for phenomena at that scale. Model at that scale often chosen phenomenologically or fit to experiment.
- Multiscale approach \rightarrow Couple calculations at different scales
Two basic paradigms:
Sequential or hierarchical – Do separate calculations at each scale, pass the results between scales \rightarrow coarse-graining or fine-graining
Best when clear separation between scales, homogeneous at large
Concurrent – Do simultaneous coupled calculations with different resolutions and physical description in different regions
Best when continuous transition between scales and direct interaction between them.

Wide range of single scale methods



Multiscale methods couple descriptions either sequentially or concurrently

Main focus today on coupling from atomistic to continuum

Single Scale Methods

- Quantum Monte Carlo – Full quantum treatment of electron interactions, and perhaps even quantum treatment of nuclei. Exponential increase in computational effort with number of particles \rightarrow limited to 10-100 quantum particles.
- Electronic structure calculations – Treat quantum mechanical nature of electrons finding ground state for fixed nuclear positions. Algorithms that are linear in # of electrons allow thousands of atoms to be treated for picoseconds. Tight-binding treatment – larger systems and times
- Classical Molecular Dynamics and Monte Carlo – Coarse-grain to follow only the motion of atoms, including effect of electrons through a classical potential energy that depends only on the positions of nuclei. Depending on complexity of potential energy can treat 10^7 to 10^9 atoms for nanosecond and longer times. Coarse-grained potentials lump atoms together, follow these groups
Example: Lump carbon and hydrogens into a single “united atom” or represent a protein by the carbon atoms along its backbone.

Single Scale Methods – coarser scales.

- Particles representing entire polymers, fluid elements, galaxies, ...
Solve dynamics with MD or on lattice (Lattice Boltzmann Method)
Know that if equations obey conservation laws, particles will follow hydrodynamic equations \rightarrow solve Navier-Stokes, magnetohydrodynamics
- Slip-link models for polymer dynamics
- Discrete defect dynamics – Identify lines or surfaces where there is a defect in the order parameter: dislocation, interface, ...
Average out atoms, follow dynamics of defect
- Phase field models – Define local free energy that depends on order parameter: concentration, magnetization, nematic director, ...
Follow dynamics of order parameter assuming overdamped dynamics
- Purely continuum treatment:
Elasticity of solids, Navier-Stokes equation for fluid, ...

Coarse Graining in Sequential Coupling

Coarse-graining: Formally want to integrate out fine-scale degrees of freedom (DOF) as in Renormalization Group

Divide DOF into coarse – labeled I, and fine – labeled i

For equilibrium partition function can formally write:

$$Z = \sum_{I_i} \exp(-\beta U(u_i, u_i)) = \sum_I \exp(-\beta U'(u_i))$$

$$\text{where } \exp(-\beta U'(u_i)) \equiv \sum_i \exp(-\beta U(u_i, u_i))$$

However:

Summation can not be done analytically in most cases of interest, numerical approximations are difficult

Effective potential U' usually longer range, depends on T, ρ , etc.

while want potential that is transferable to different cases

Dynamic treatments even harder

Curtarolo & Ceder → Migdal-Kadanoff bond moving RG

Broughton & Rudd → KE of removed atoms and approx entropy

Common Method of Sequential Coupling

Calculate some quantity with fine-scale description

– eg. pair distribution function, energy, viscosity, elastic constants, pressure-volume curve, Kuhn length, order parameter

Adjust parameters of coarser description to reproduce fine-scale data

→ Bad parametrization may miss physics

Best if few parameters fixed by symmetry, conservation that can calculate directly – i.e. viscosity, elastic moduli, boundary cond.

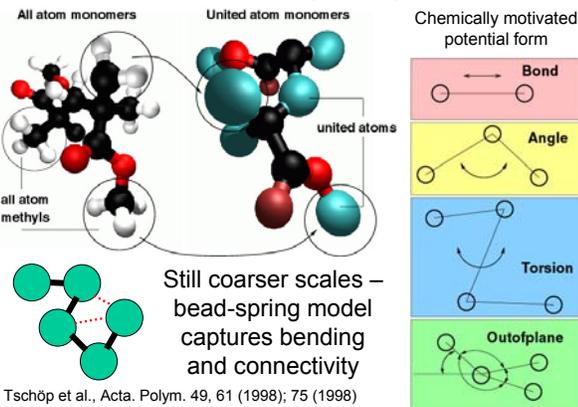
→ Quantities not included in fit may be inaccurate

→ Very different parameters may fit fine-scale data equally well

(Historically this approach was often followed using experimental data to calibrate the description)

Fine-graining requires way of reinserting fine-scale structure – not unique in general, so must allow re-equilibration. Can equilibrate at coarse scale, reinsert atoms to find polarizability, ...

Coarse- and Fine-Graining of Polymer Models



Connecting Atomic Simulations to Continuum Theory

Continuum mechanics needs:

Boundary conditions (BC) + Constitutive Relations
 velocity or stress stress vs. strain (rate)
 slip, friction, adhesion viscous, elastic, plastic

Traditionally assume simple forms:

No-slip for fluids, Linear viscosity, elasticity
 friction \propto load, ... Simple analytic functions

Want to replace assumptions by calculations. Need to know:

→ Down to what scale do continuum equations apply?

→ Is there new mesoscopic behavior between atomic and bulk?

→ Do usual assumptions for BC and constitutive laws work?

Continuum assumes homogeneity, short-range correlations

Simple molecules → Continuum good at a (3-10) diameters

Near phase transition, correlation length sets larger scale

Polymers → New behavior on entanglement scale

Interfacial behavior may be more complicated than assumed

Molecular Dynamics Study of the Stress Singularity at a Corner

O. Vafek and M. O. Robbins Phys. Rev. E60, 12002 (1999)

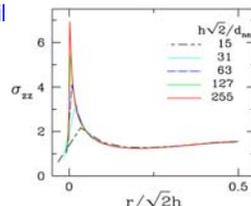
Continuum mechanics predicts stress singularity at corners between dissimilar materials ⇒ important for initiation of failure
 Simulations show:

⇒ Singularity has continuum form at large scales, but is cut off at molecular scale by discreteness, anharmonicity or plastic flow

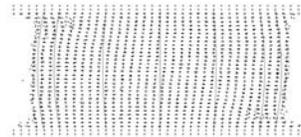
⇒ Maximum stress increases as a power of the system size

⇒ small bonds less likely to fail

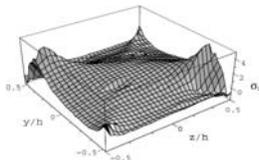
Geometry Stress from thermal expansion



For large enough system, yield stress exceeded, stress relieved by dislocation motion



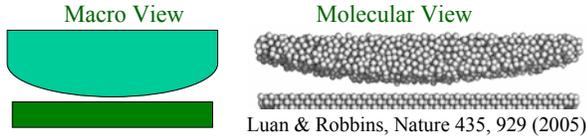
Stress release is anisotropic, inconsistent with many continuum models



What are limits of Continuum Theory for Solids?

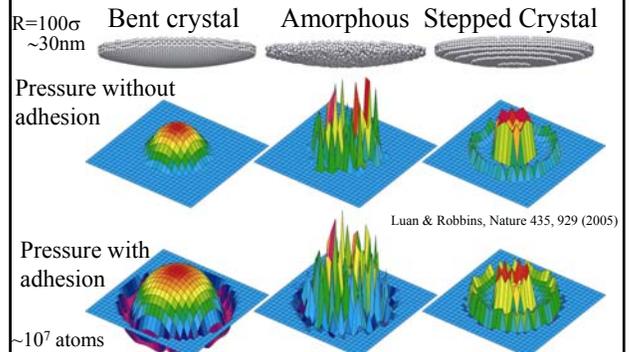
Continuum theories: Hertz, Johnson-Kendall-Roberts
 Assume: 1) continuous displacements, bulk elastic const.
 2) smooth surface (often spherical) at small scales
 Only tested for atomically flat mica bent into cylinders
 and elastomers with liquid behavior on small scales

Find (1) valid down to a few atomic diameters, but atomic
 scale roughness causes failure of continuum theories.
 Important for small contacts between rough surfaces
 and ideal single asperities: scanning probe or nanoindenter



Pressure distribution for sphere on flat

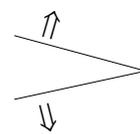
Atomic scale roughness qualitatively changes pressure, yield
 Bent crystal agrees with Hertz/JKR, more realistic tips do not



Single Asperity Conclusions

- Bulk elastic modulus describes stress/strain to $\sim 3\sigma$
 Atomic roughness \Rightarrow deviations from continuum theory
- Molecular scale geometry has little effect on normal displacement vs. force curves
 \rightarrow Moduli from continuum fits are accurate
- Contact areas, morphologies and pressures are changed
 \rightarrow Yield stress, areas, pull-off force off by factor ~ 2
 \rightarrow Adhesive energy off by factor ~ 5
- Lateral stiffness and friction vary by more than order of magnitude with atomic geometry
 \rightarrow Contact stiffness dominated by interface
 \rightarrow Friction scales with real contact area for bent or amorphous tips, but not stepped tips
 \rightarrow Shear stresses from continuum fits too high

Toughness or fracture energy $G \rightarrow$ Crack resistance



$G \equiv$ work / fractured area

Lower bound $G_{eq} = 2\gamma$

\rightarrow interfacial free energy change

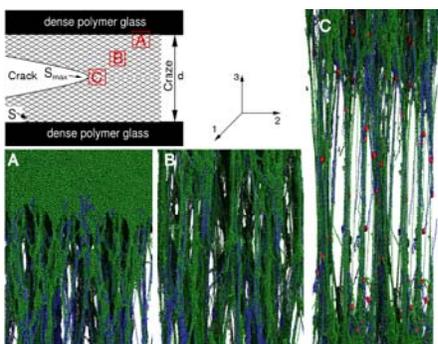
Glassy polymer: $G/2\gamma \sim 10^3 - 10^4$

$< 10^3$ for metals

How is $G - 2\gamma$ dissipated?

Fracture in Polymer Glasses \rightarrow Many Length Scales

cm crack \rightarrow μm craze \rightarrow 10nm fibril \rightarrow nm polymer
 Combine simulations in each region to get macro G



Interaction Potential \rightarrow Bead-Spring Model

Each polymer contains N spherical beads

..... All interact with Lennard-Jones potential

$$V_{LJ}(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6] \text{ for } r < r_c$$

$$\epsilon \sim 3\text{kJ/mol} = 30\text{meV}, \sigma \sim 0.5\text{nm}, \tau = (m/\epsilon)^{1/2} \sigma \sim 5\text{ps}$$



— Backbone

\rightarrow Empirical potential that breaks at force f_B

experiment $\Rightarrow f_B = 100 f_{LJ}$

Semiflexible chains \rightarrow add bond-bending terms

\rightarrow less flexible, smaller N_e

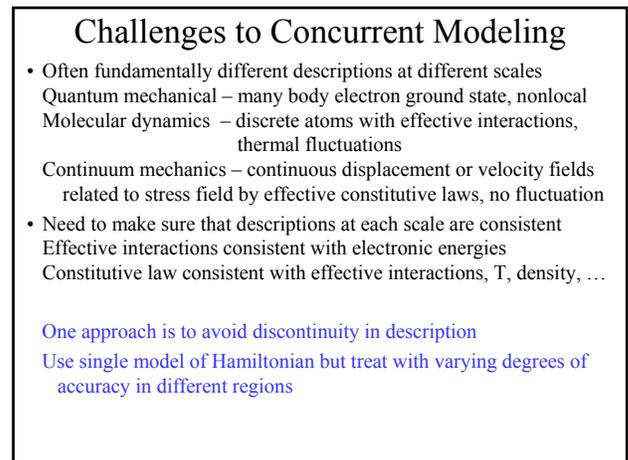
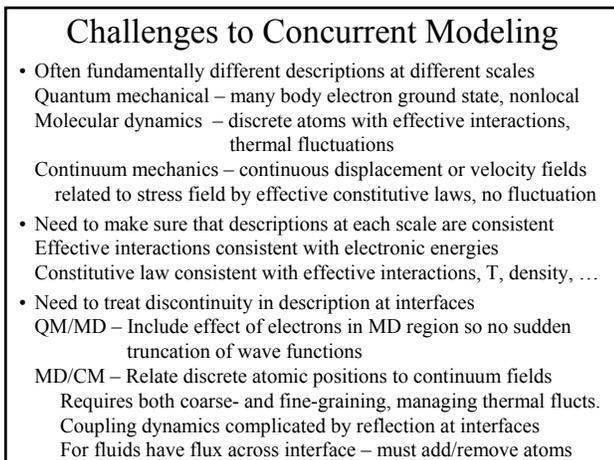
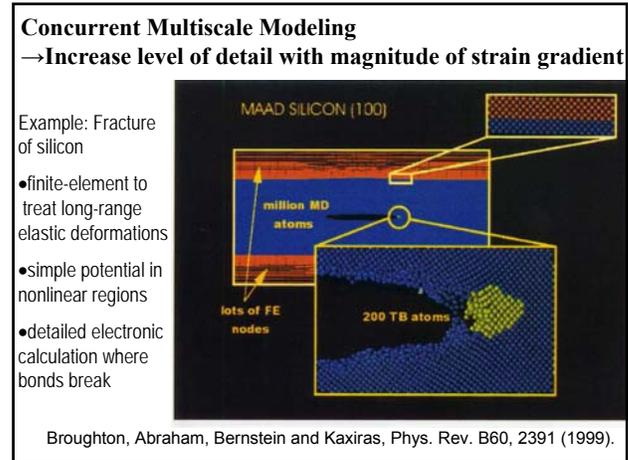
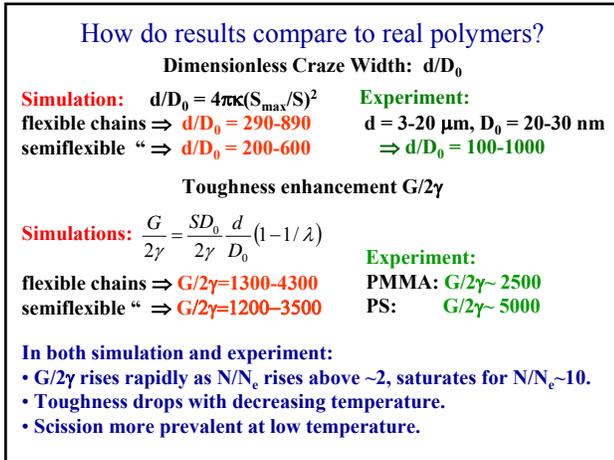
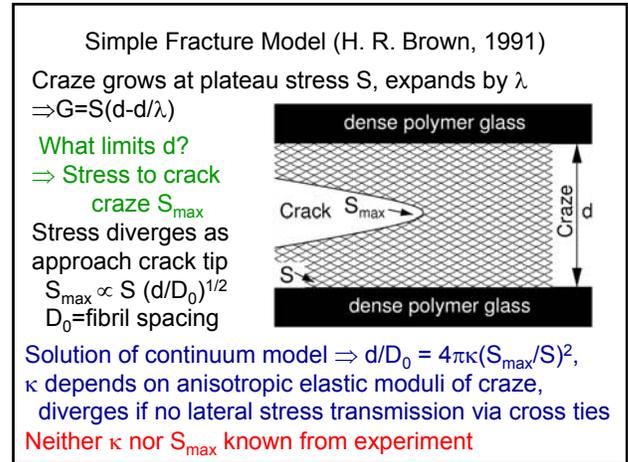
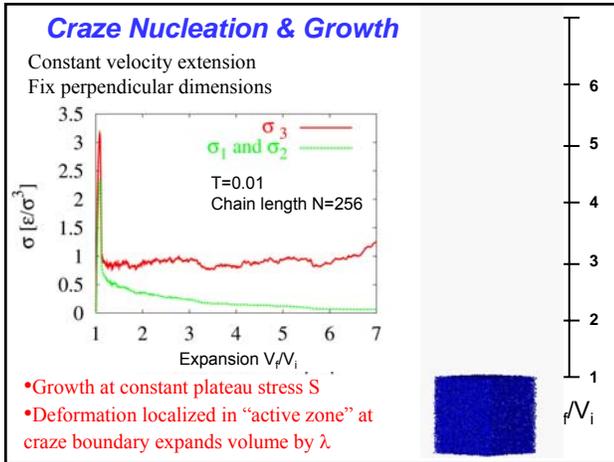
Control stress or strain, 3D periodic boundaries or walls

Vary: N/N_e

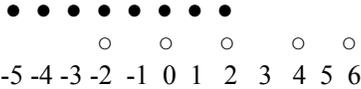
Flexibility of chains $- N_e$

Temperature: $T = 0.01$ to $0.3\epsilon/k_B$ ($T_g \approx 0.4\epsilon/k_B$)

Ratio of backbone and LJ bond-breaking forces f_B/f_{LJ}



What Happens at MD/FEM Interface?

- Alternative approach for **discontinuous** description
Finite elements larger than interaction range – local
Use overlap region so sharp interfaces outside region of interest
- 1D example MD ●, FEM ○


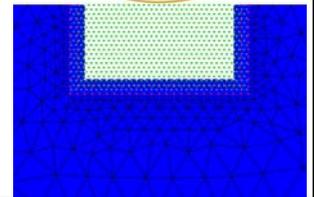
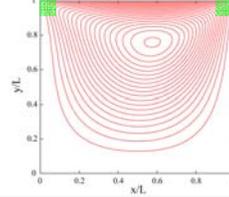
i = -5 -4 -3 -2 -1 0 1 2 3 4 5 6
- Two descriptions provide boundary conditions for each other at outer edge of overlap
Displacements for solid, velocities for fluid

Linking Atomistic and Continuum Regions

- Three overlap regions where solve both continuum and MD
Outermost → Continuum solution gives MD boundary condition
Innermost → MD gives continuum boundary condition
Middle → Two solutions equilibrate independently
- Fluids: Apply boundary conditions to velocities
Solids: Apply boundary conditions to displacements

Streamlines in L=100nm channel with moving top wall. Atomistic solution in <1% of area (green) removes continuum singularity

Model contact region atomistically, elastic deformations with finite-elements, constrain deformations in overlap region



Linking Atomistic and Continuum Regions

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Outermost → Continuum solution gives MD boundary condition
Innermost → MD gives continuum boundary condition
Middle → Two solutions equilibrate independently
- Fluids: Apply boundary conditions to velocities
Solids: Apply boundary conditions to displacements
- Fluids: S. T. O'Connell & P. A. Thompson, Phys. Rev. E52, R5792, (1995)

Why not use forces instead of displacements/velocities?

E. G. Flekkoy, G. Wagner & J. Feder, Europhys. Lett. 52, 271 (2000)

Fluids – Position of boundary is undetermined
→ drifts in response to fluctuations or systematic errors

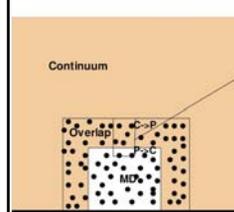
General – Any error in constitutive relation creates problems in overlap region
Less sensitive when match displacements
any global factor in stress is irrelevant

Hybrid Algorithm Applied to Fluids

- Continuum: Incompressible Navier-Stokes (Projection method)
- Atomistic: Molecular dynamics of Lennard-Jones atoms, no-slip
Potential: $U(r) = 4\epsilon((\sigma/r)^{12} - (\sigma/r)^6)$; Units ϵ, σ

$$\text{MD} \rightarrow \text{Continuum} \quad u_j = \frac{1}{N_j} \sum_{i=1}^{N_j} v_i$$

$$\text{Continuum} \rightarrow \text{MD} \quad \frac{1}{N_j} \sum_{i=1}^{N_j} v_i = u_j(t) \quad m\ddot{x}_i = F_i - \frac{1}{N_j} \sum_{k=1}^{N_j} F_k + m \frac{Du_j}{Dt}$$



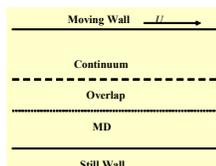
$F_i = -\frac{\partial}{\partial x} \sum_k V_{ik}^{LJ}$

Potential confines particles at y_3

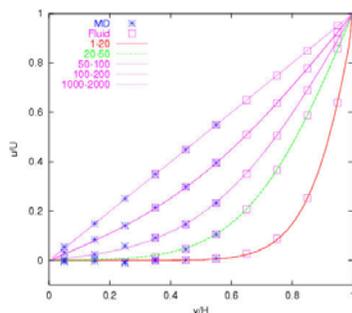
Insert/remove number of particles equal to net flux

Dynamic Couette Flow

Schematic of simulation



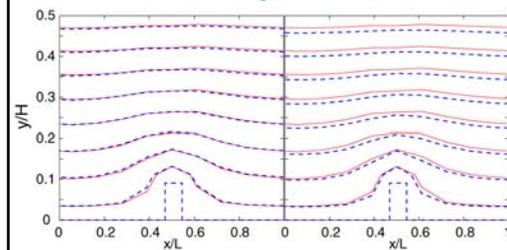
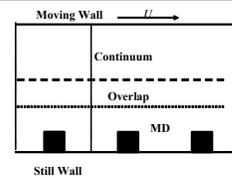
Hybrid solution (symbols) tracks full continuum (lines) as a function of time after motion starts



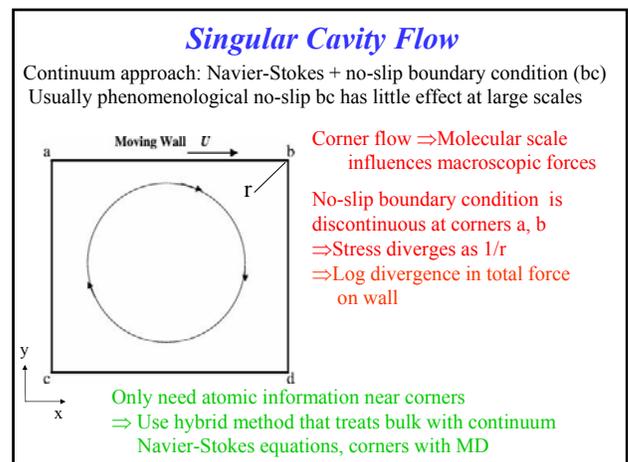
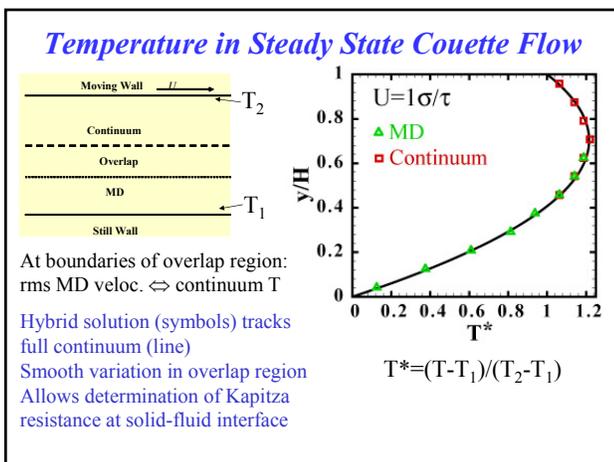
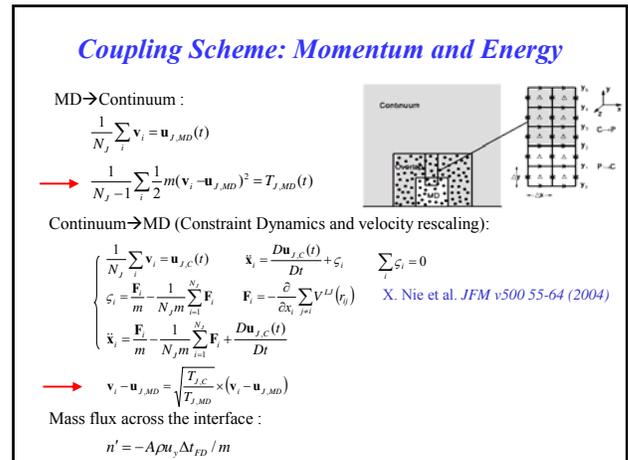
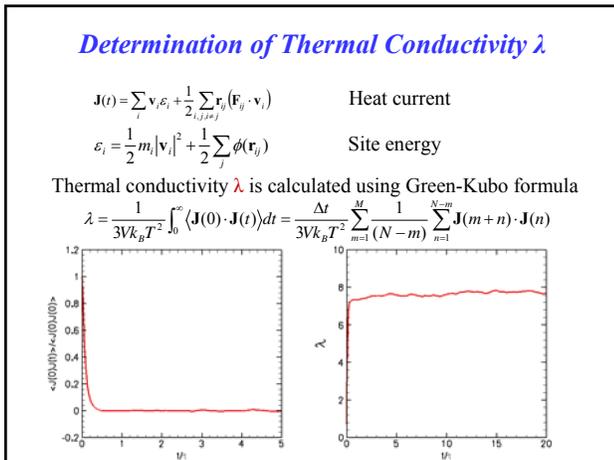
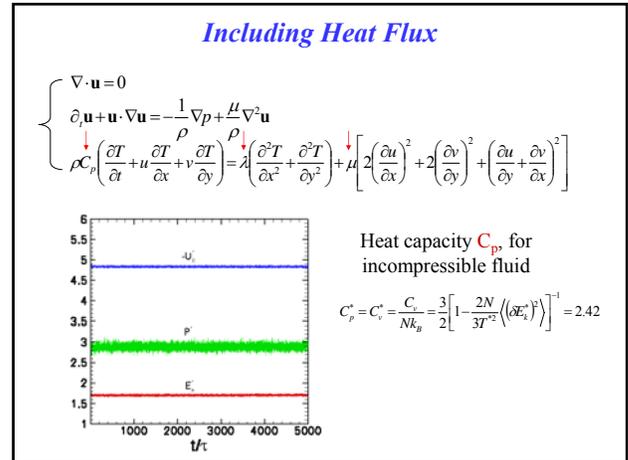
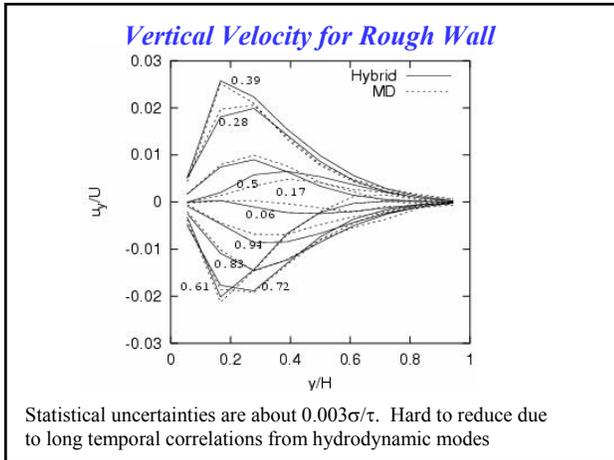
X. B. Nie, S. Y. Chen, W. N. E and M. O. Robbins, J. Fluid Mech. 2004.

Flow past a rough wall

- Streamlines from hybrid \approx MD includes flow between regions
- Continuum fails because doesn't match complex boundary condition around bump



Hybrid vs. MD Hybrid vs. Continuum



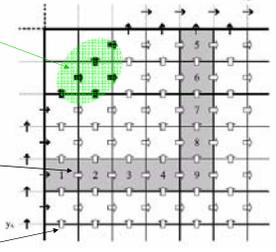
Coupling in Overlap Region

MD ⇒ Navier Stokes
 Mean atomic velocity gives boundary condition to NS eqs.

Continuum ⇒ MD
 1) Average tangential MD velocity in shadowed bins forced to NS value:

$$\bar{x}_i = \frac{F_i}{m} - \frac{1}{mN_j} \sum_{j=1}^{N_j} F_j + \frac{Du_i}{Dt}, F_j = -\frac{\partial}{\partial x} \sum_k V_{jk}$$

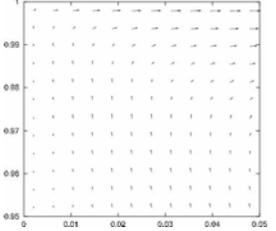
2) Normal MD velocity constrained by matching mass flux at boundary

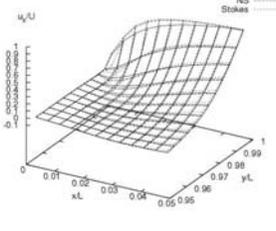


Have tested:
 Agrees with pure MD calculations.
 Independent of continuum grid 1, 3 and 6σ and specific set of constrained velocities (within MD noise)
 X.B. Nie, S.Y. Chen and M. R. Robbins, Physics of Fluids 2004.

NS and Hybrid Velocities Near Corners

hybrid





Effect *like* slip BC on scale S
 S is larger of $\sim 2\sigma$ and $U/0.1\sigma/\tau$

discreteness shear-thinning

Hard to use effective Navier BC: spatially varying, nonlinear

Treating Large Range of Length Scales

Problem: Size of atomistic region independent of system size L
 BUT time to equilibrate NS flow field grows with L.
 Initial approach limited to $L \sim 0.1\mu\text{m}$.

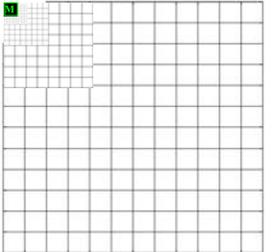
Solution: Multigrid and time approach
 Integrate to steady state at each scale with optimum time step.
 Iterate between scales till self-consistent (~ 10 times).

Result: Size limited only by onset of non-steady, turbulent flow
 Show results for 0.1mm cavities.
 > 10 orders of magnitude faster than fully atomistic
 ~ 20 minutes per iteration
 Use average over 16 MD representations to accelerate

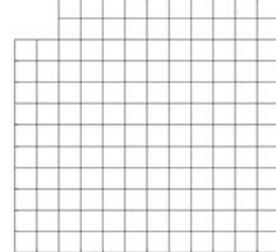
Nie, Robbins and Chen, Phys. Rev. Lett. **96**, 134501 (2006).

Schematic of Local Refinement

Coarse → Fine: Prolongation.

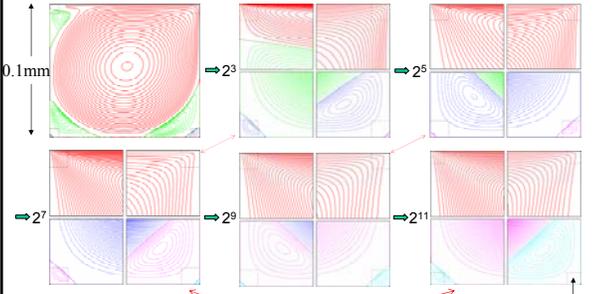


Fine → Coarse: Restriction.



Flow at each scale reaches steady state at its own characteristic time

Multiscale Solution for $Re=6400$ ($U=0.068\sigma/\tau$)

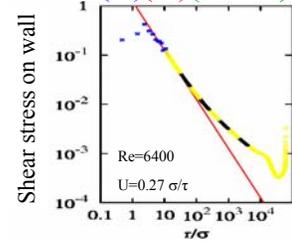


- Ten grid levels, largest 256x256, others 64x64, smallest mesh 0.95σ
- Dashed lines: the regions expanded in successive plots. Final plot → MD region
- Stokes equations → bottom corners self-similar under mag. by ~ 16 (red arrows)
 This scaling is cut off by atomic structure.
- Computational time saving more than 10^{10} over fully atomistic.

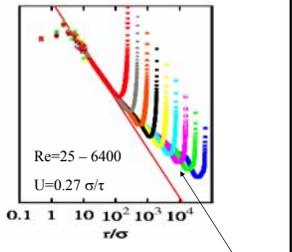
Stress along the moving wall

Three regions contribute to force F:
 Atomistic, Stokes, high Re

$Re=6400$
 $U=0.27\sigma/\tau$



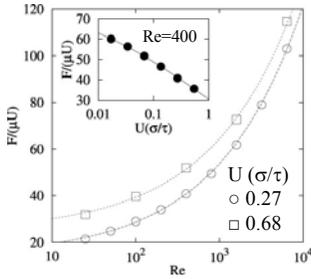
$Re=25-6400$
 $U=0.27\sigma/\tau$



Breakdown of Stokes for $r < S$ – atomistic or $r > R_1 \equiv \mu/\rho U$ – inertial
 Little change for $r < R_1$ as increase Re by increasing L
 Large r contribution gives change in F for fixed U, atomic props.

Total Force on the Moving Wall

Re - only parameter in continuum theory
Find strong variation with U at fixed Re, atomic model



$$\frac{F}{\mu U} = f_S + f_{Stokes} + f_{Re}$$

$$r < S \quad S < r < R_1 \quad R_1 < r$$

$$f_S = 4.3$$

$$f_{Stokes} = \frac{8\pi}{\pi^2 - 4} \ln(R_1 / S)$$

$$f_{Re} = 3.85 + 1.98Re^{0.434}$$

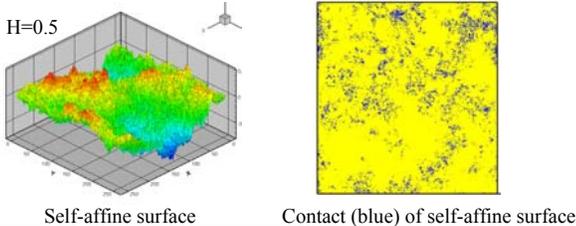
f_S given by assumption that stress saturates at S
 $S = 0.3 + 7U_{tL}$; f_{Re} is phenomenological fit

Summary for Fluid Flow

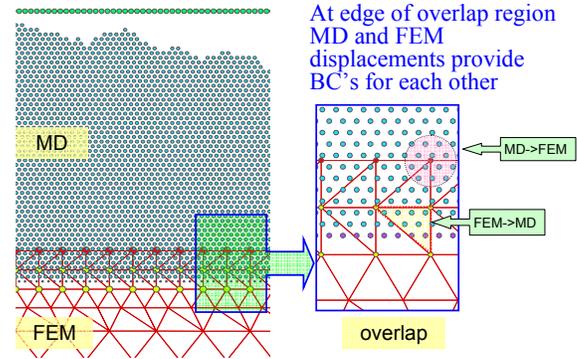
- We have developed a multiscale hybrid method that can simulate a macro-length scale flow while still resolving the atomistic structure in a small region. Treats mass and heat flux
- The ability to resolve the stress on all scales enables the first calculation of the drag force on the moving wall in cavity flow. The force depends on three dimensionless numbers:
 $Re = \frac{\rho UL}{\mu}$, $I = \frac{U\tau}{\sigma}$ and $R_m = \frac{S}{r_1} = \frac{\rho US}{\mu}$.
- Algorithm adapted to dynamic interfaces
Initial results for contact line motion will be extended to study interface shape and stress over wide range of length scales.

Multi-scale modeling of contacts between self-affine surfaces

Contact geometry and stresses central to friction & adhesion
Real surfaces often rough on many scales → self-affine
Surfaces steeper at smaller scales, fractal contact regions, most connected regions of contact at resolution of calculation
⇒ Not clear continuum mechanics applies



Hybrid model for 2d self-affine surfaces Easily treat volumes with ~10⁸ atoms



Continuum Treatment of Solid

- Linear finite elements
- Explicit dynamics for nodes
- Newmark method $\Delta t_{FE} \sim 10 \Delta t_{MD}$, Langevin thermostat
- Staggered time grid as for fluids
- Constitutive law – quadratic in strain
– accurate to 2% in each strain component

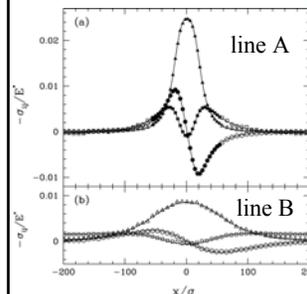
Atomistic Treatment of Solid

- Two dimensional triangular lattice
- Lennard-Jones interactions between neighbors
- Velocity Verlet, Langevin thermostat

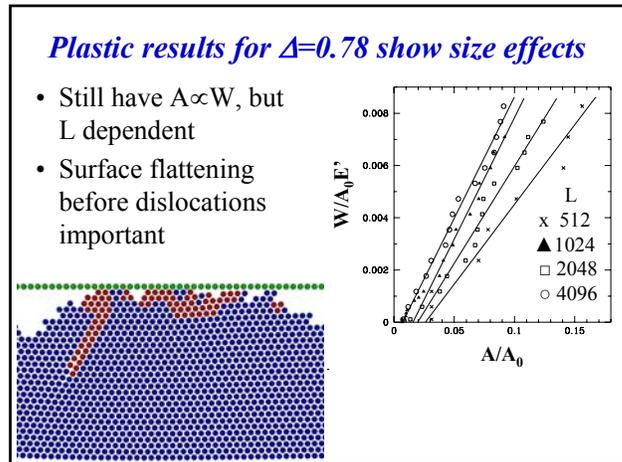
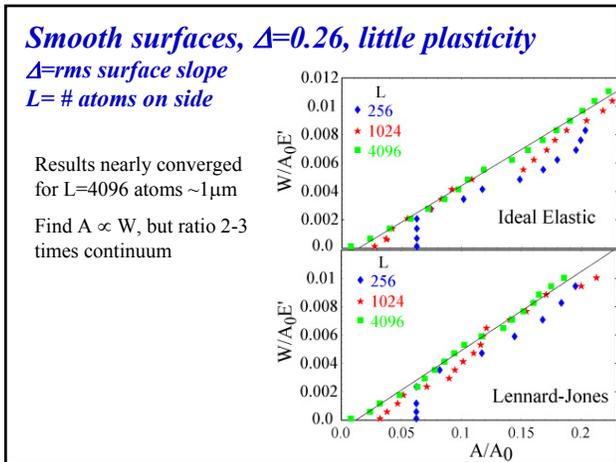
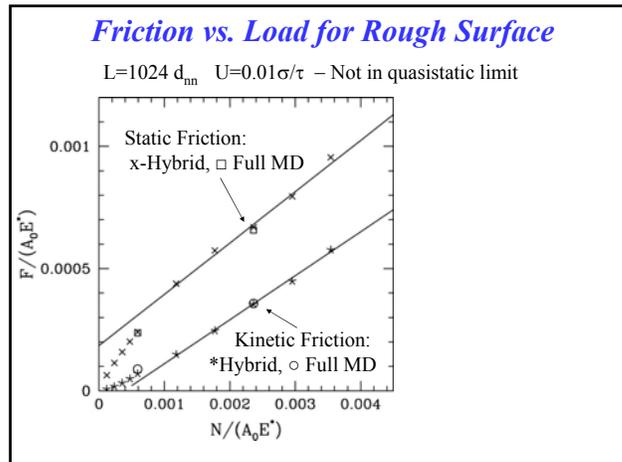
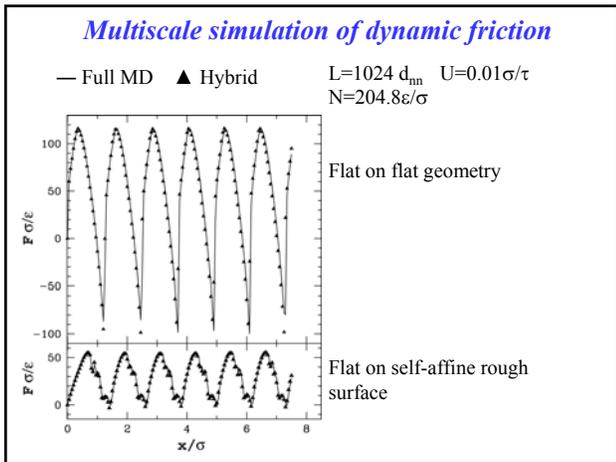
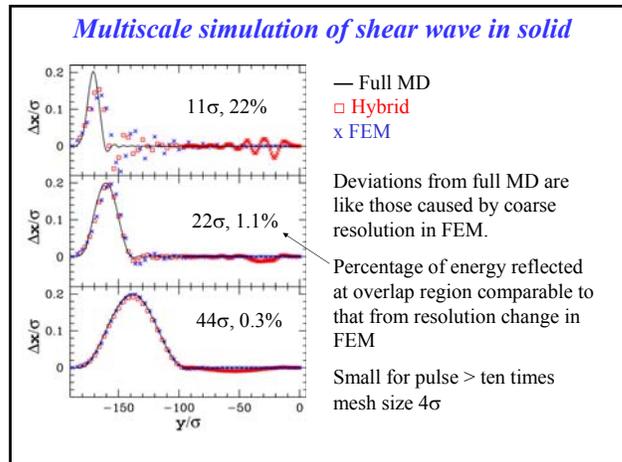
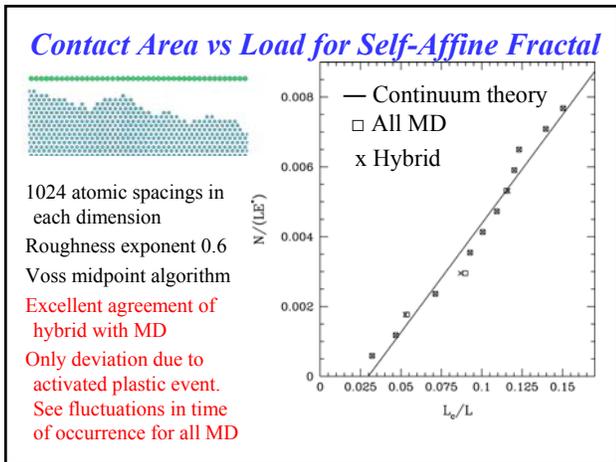
Show low T results to minimize noise, but works at high T

Quasistatic Test Cylindrical Contact

Mesh, atomistic & overlap



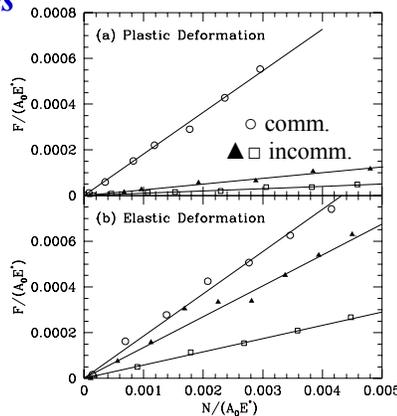
Lines – pure MD
Symbols – hybrid
Filled – MD region
Open – FEM region
▲ - σ_{yy} ● - σ_{xx}
■ - σ_{xy}



Friction forces

commensurate:
plastic \approx elastic

incommensurate:
plastic < elastic
because contacts
bigger, forces
average to zero



Conclusions for Hybrid Method

- Have robust multiscale method for both fluids and solids
 - lengths to $\sim 1 \mu\text{m}$ for dynamic cases, $\sim 1\text{mm}$ for quasistatic
- Implemented for quasi-2D contact between self-affine surfaces
- Incorporated heat flux for sheared fluids
- Comparisons to MD and continuum results show limitations of continuum approximation at interfaces
 - Position and rate dependent slip near solids
 - Sensitivity of contact area and stress to atomic scale structure, unexpected mode of plastic deformation at interface
- First calculation of drag force in singular corner flow
 - integrate stress over 5 orders of magnitude in length
- First calculation of atomistic effects in self-affine contact
 - rough over 4 orders of magnitude in length scale

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