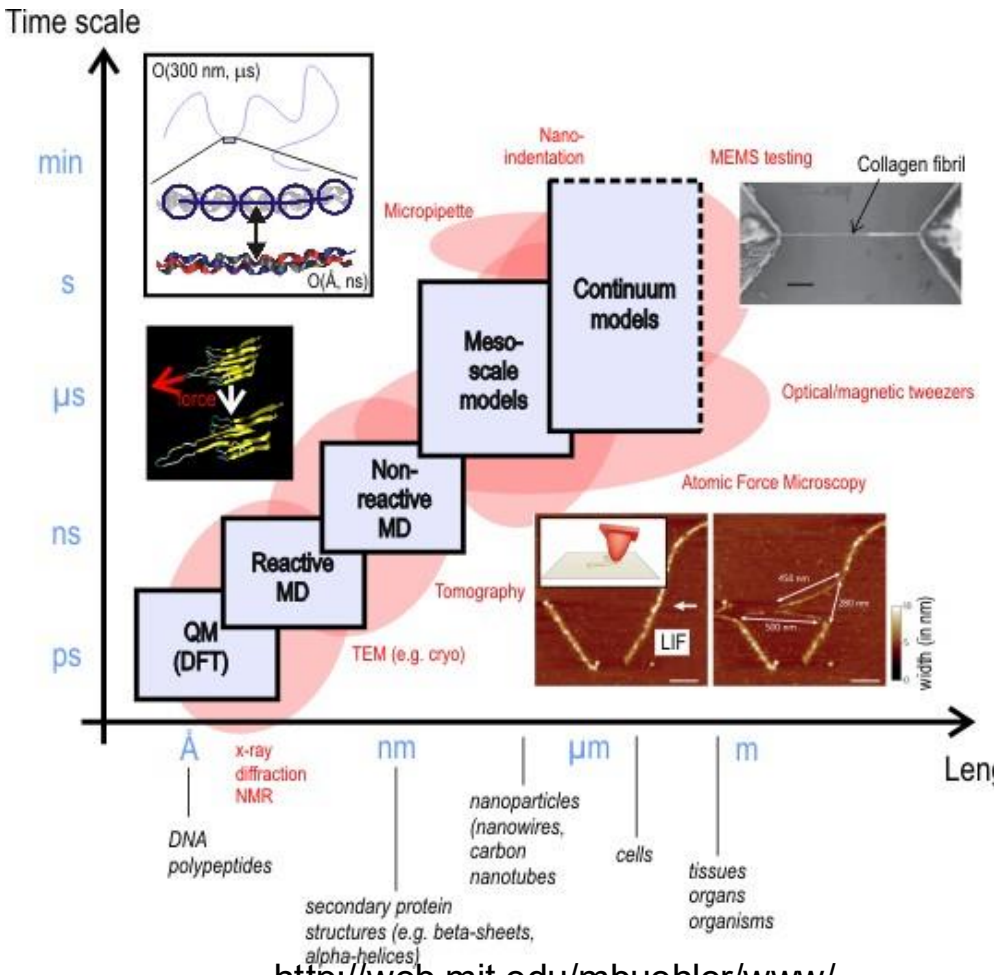
11th i-CoMSE Workshop: Mesoscale Particle-Based Modeling

Mississippi State University
July 21–25, 2025

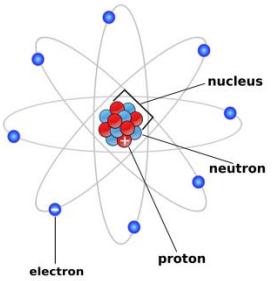
Session 1: Introduction to coarse graining

Multiscale modeling

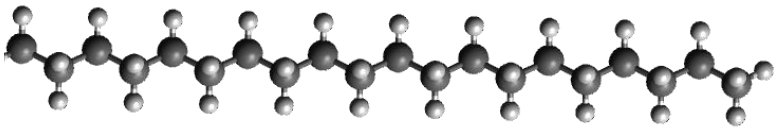
- Length and time scales → level of detail in treatment of matter



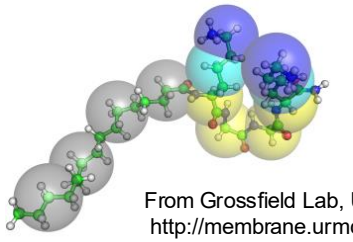
http://web.mit.edu/mbuehler/www/



Quantum (electrons & nuclei)



Atomistic (atoms as soft spheres)



From Grossfield Lab, URMC, http://membrane.urmc.rochester.edu/horn

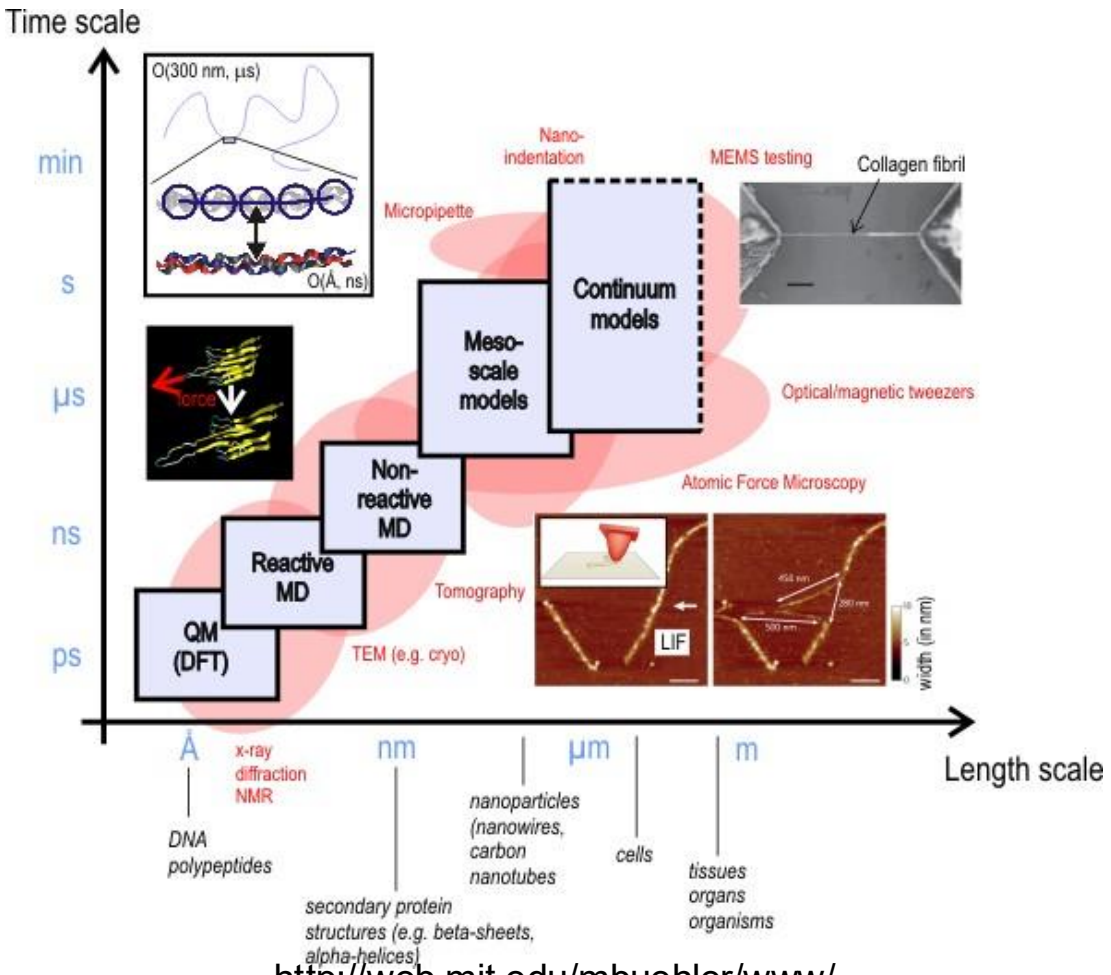
Mesoscale (groups of atoms as soft spheres)

$$\rho\left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v}\right) = -\nabla p + \nabla\cdot\mathbf{T} + \mathbf{f}$$

Continuum (matter as fields)

Multiscale modeling

- What type of phenomena are we trying capture with the model?



- **Quantum:** chemical reactions, charge transfer, etc.
- **Atomistic:** phase diagrams, thermophysical and transport properties, macromolecular structure (protein folding), etc.
- **Mesoscale**: macromolecular self-assembly, complex fluid rheology, hydrodynamic phenomena, etc.
- **Continuum**: device-level modeling of heat, mass, momentum transport.

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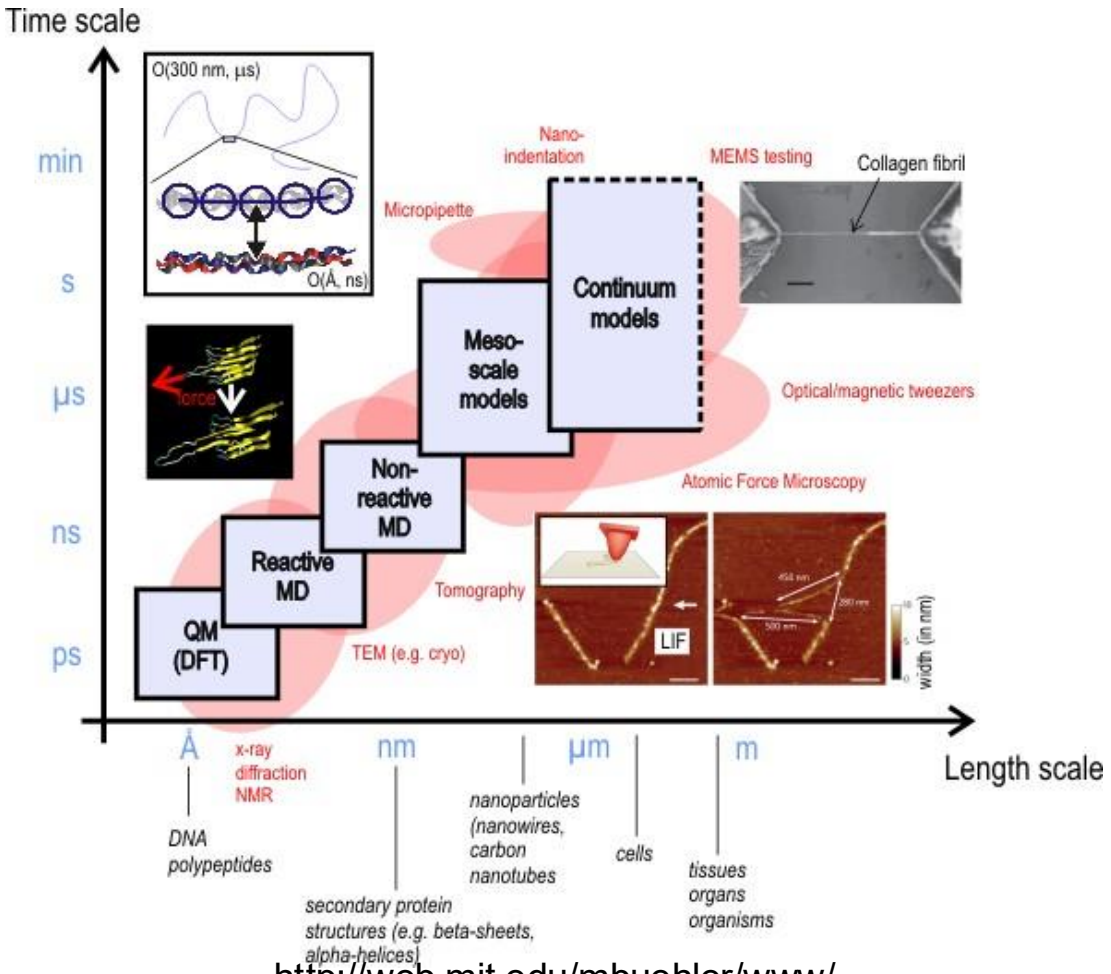
Multiscale modeling

- What inputs do we need to model at different length scales?



- **Quantum:** atomic number*
- **Atomistic:** force fields for describing bonded and non-bonded interactions between atoms
- **Mesoscale**: force fields for describing bonded and non-bonded interactions between mesoparticles
- **Continuum**: bulk material properties (e.g., thermal conductivity, elastic moduli, etc)

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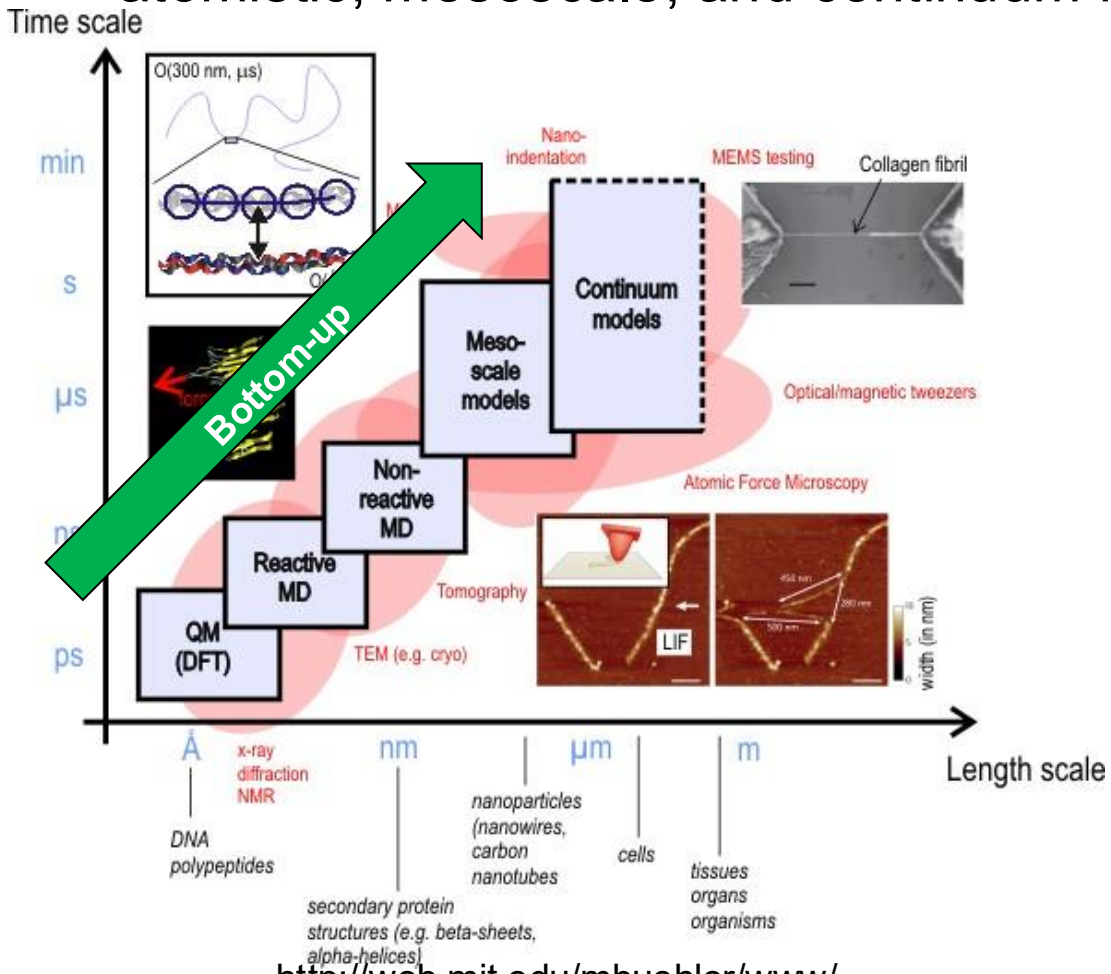
Coarse graining

- Coarse graining approaches allow us to obtain the information (force fields, material properties, etc.) to necessary to develop atomistic, mesoscale, and continuum models



- **Bottom-up:** reproduce specific features of a more detailed (higher-resolution) model

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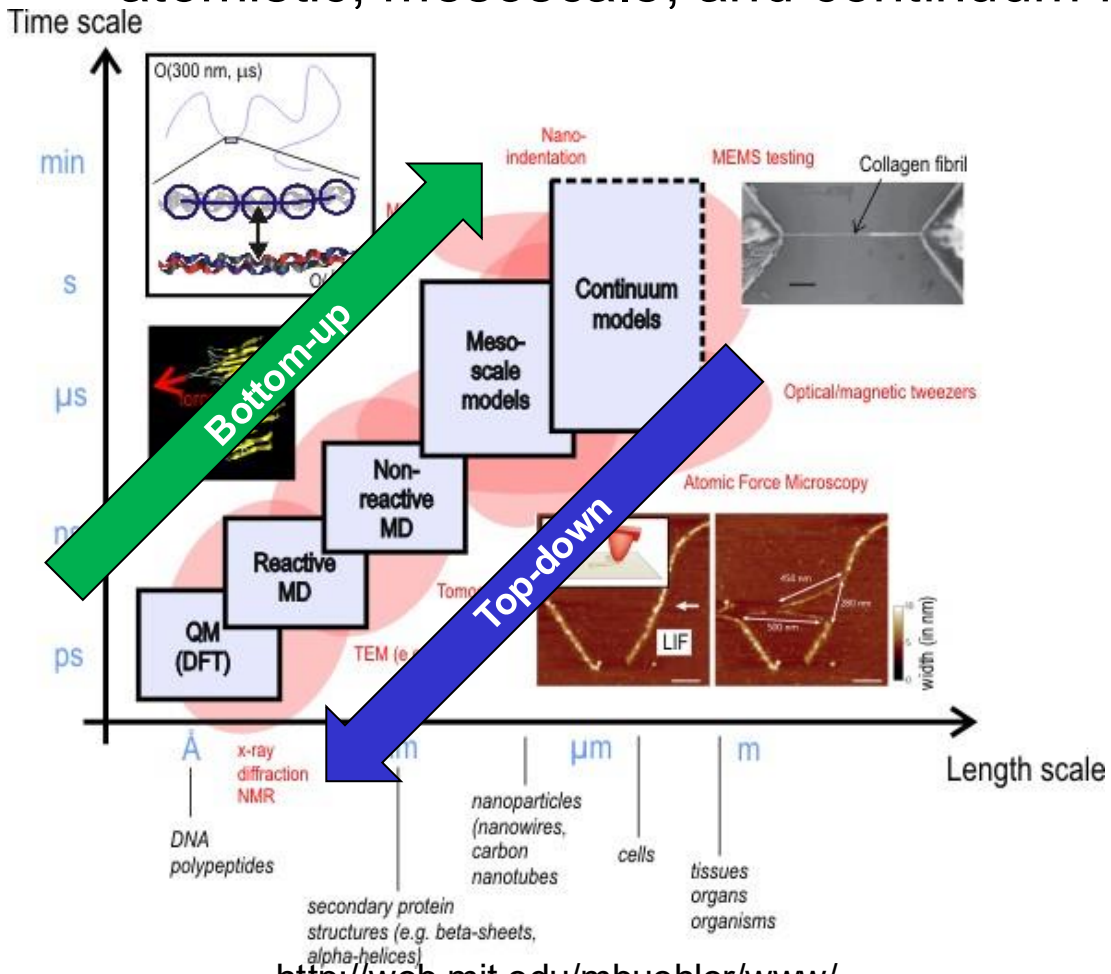

- **Bottom-up:** reproduce specific features of a more detailed (higher-resolution) model
- **Top-down:** reproduce specific phenomenological or emergent properties at macroscale, or theoretical expectations

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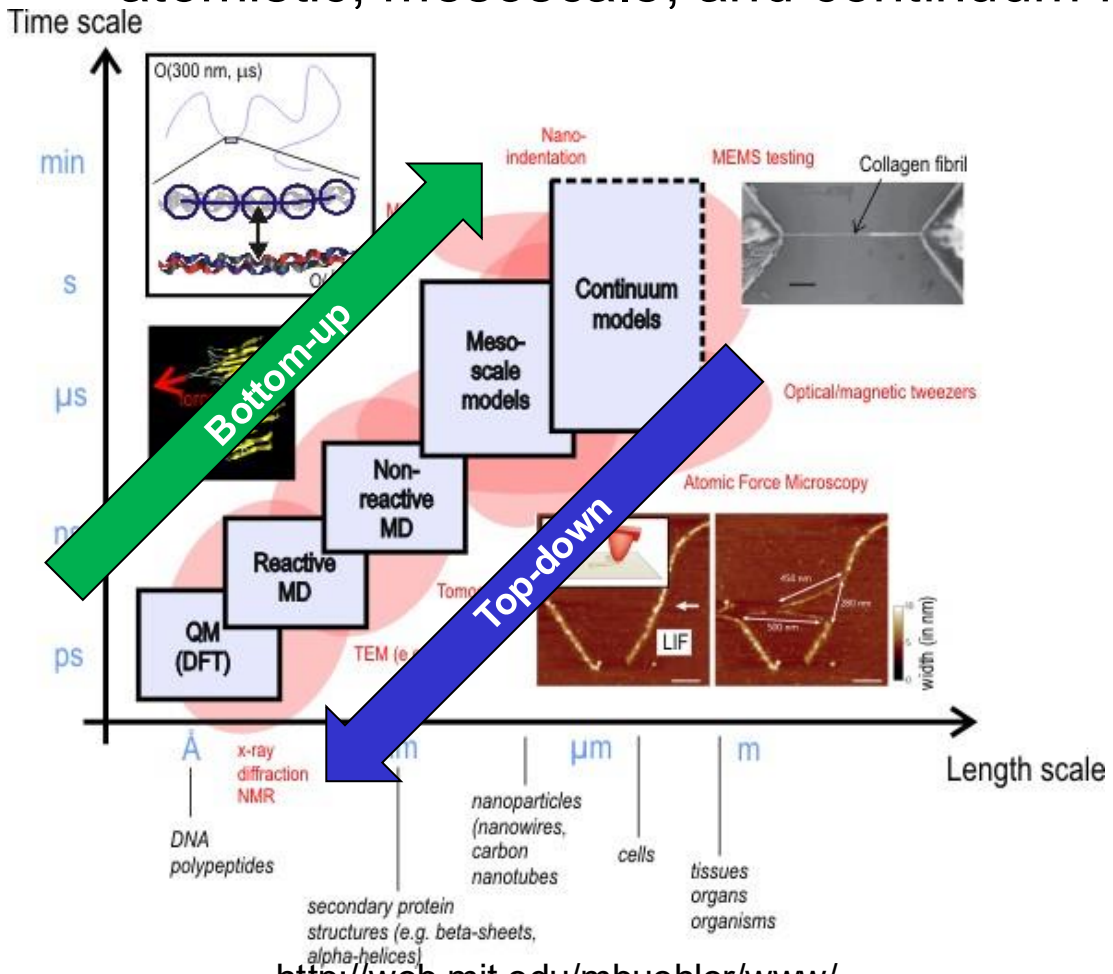
- Coarse graining approaches allow us to obtain the information (force fields, material properties, etc.) to necessary to develop atomistic, mesoscale, and continuum models



- **Bottom-up:** reproduce specific features of a more detailed (higher-resolution) model
- **Top-down:** reproduce specific phenomenological or emergent properties at macroscale, or theoretical expectations
- "**Hypothesis-driven**": use hypotheses regarding essential interactions driving the behavior of interest to propose a CG model

http://web.mit.edu/mbuehler/www/

Bottom-up example I

- Quantum→ Atomistic: parameterization of a class II force field using quantum mechanics data

$$\mathcal{U}_{total} = \mathcal{U}_{nonbonded} + \mathcal{U}_{bonds} + \mathcal{U}_{angles} + \mathcal{U}_{torsions}$$

$$\mathbf{F}_i = -\frac{\partial \mathcal{U}_{total}}{\partial \boldsymbol{r}_i} \text{ (net force on atom } i)$$

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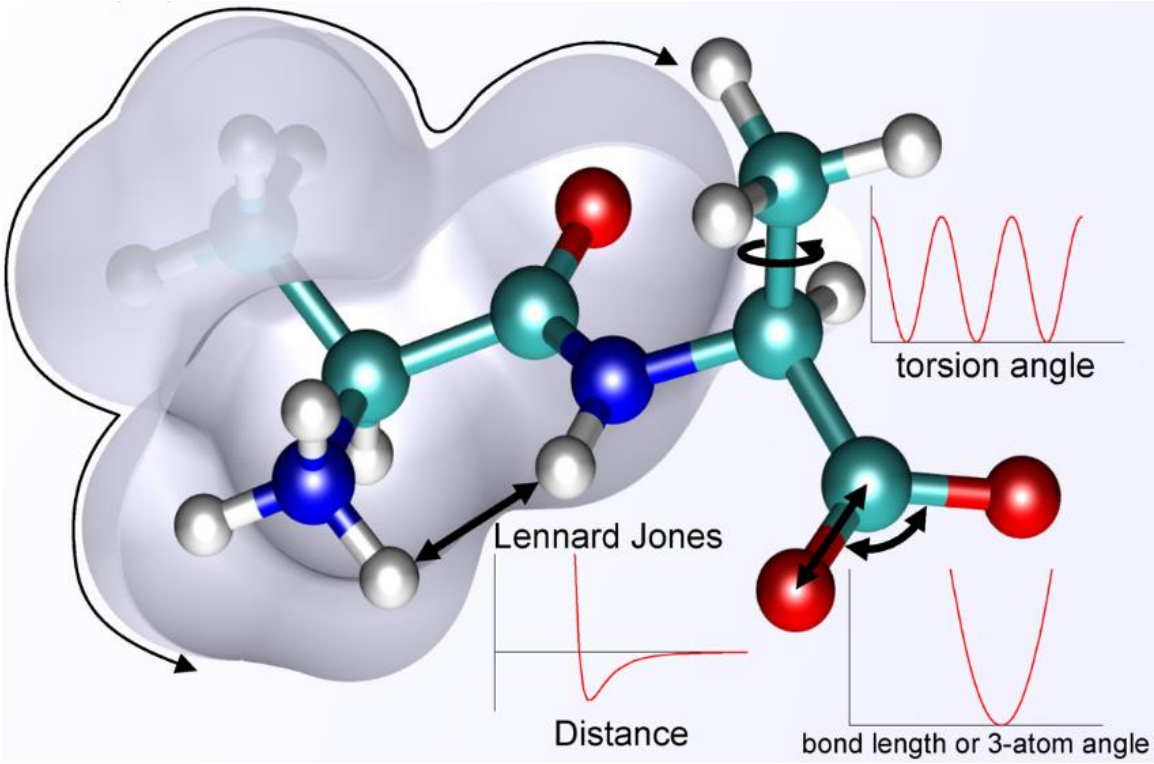

https://encyclopedia.pub/entry/33716

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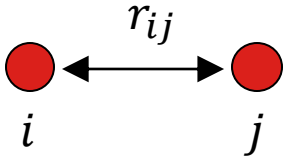

$$\mathcal{U}_{nonbonded} = \sum_i \sum_{j \neq i} 4\varepsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^{6} \right] + \sum_i \sum_{j \neq i} \frac{q_i q_j}{\epsilon r_{ij}}$$

van der Waals — Electrostatic

$$\varepsilon_{ij} = \sqrt{\varepsilon_i \varepsilon_j}. \quad \sigma_{ij} = \frac{1}{2}(\sigma_i + \sigma_j)$$

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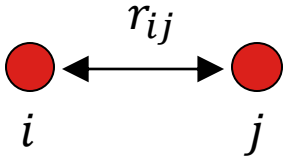

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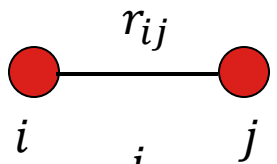
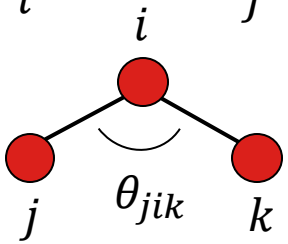
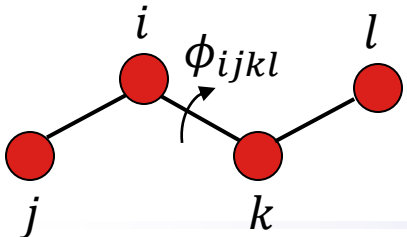


$$\mathcal{U}_{bonds} = \sum_{bonds} \frac{1}{2} k_{bond}\left(r_{ij} - l\right)^2$$



$$\mathcal{U}_{angles} = \sum_{angles} \frac{1}{2} k_{angle}\left(\theta_{jik} - \theta_0\right)^2$$



$$\mathcal{U}_{torsion} = \sum_{torsions} k_{tor}[1 + \cos\left(n_i \phi_{ijkl} + \delta_i\right)]$$

Bottom-up example I

- Quantum→ Atomistic: parameterization of a class II force field using quantum mechanics data

$$\mathcal{U}_{total} = \mathcal{U}_{nonbonded} + \mathcal{U}_{bonds} + \mathcal{U}_{angles} + \mathcal{U}_{torsions}$$

$$\mathbf{F}_i = -\frac{\partial \mathcal{U}_{total}}{\partial \boldsymbol{r}_i} \text{ (net force on atom } i)$$

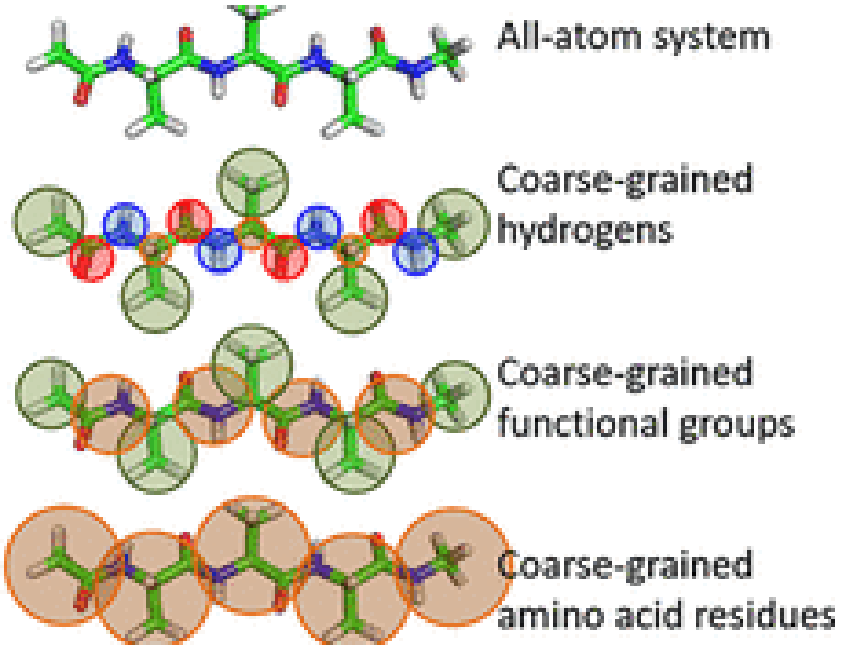
- Perform quantum calculations on different configurations of the system to generate reference data

- "Force matching": use numerical optimization / machine-learning methods to find model parameters for the atomistic force field that best reproduce energies and forces from quantum calculations (i.e., minimize L)

$$L = w_{energy} \sum_{configs} \left(\mathcal{U}^A_{total} - \mathcal{U}^Q_{total}\right)^2 + w_{force} \sum_{configs} \sum_{atoms} \sum_{x,y,z} \left(\mathrm{F}^A_i - \mathrm{F}^Q_i\right)^2$$

Bottom-up example II

- Atomistic → Mesoscale: parameterization of a mesoscale force field using atomistic simulation data
- Determine the desired resolution of the mesoscale model
  - How are atoms grouped to define each mesoscale beads?



Ye et al., ACS Omega 6, 1758–1772, 2021

Bottom-up example II

- Atomistic → Mesoscale: parameterization of a mesoscale force field using atomistic simulation data
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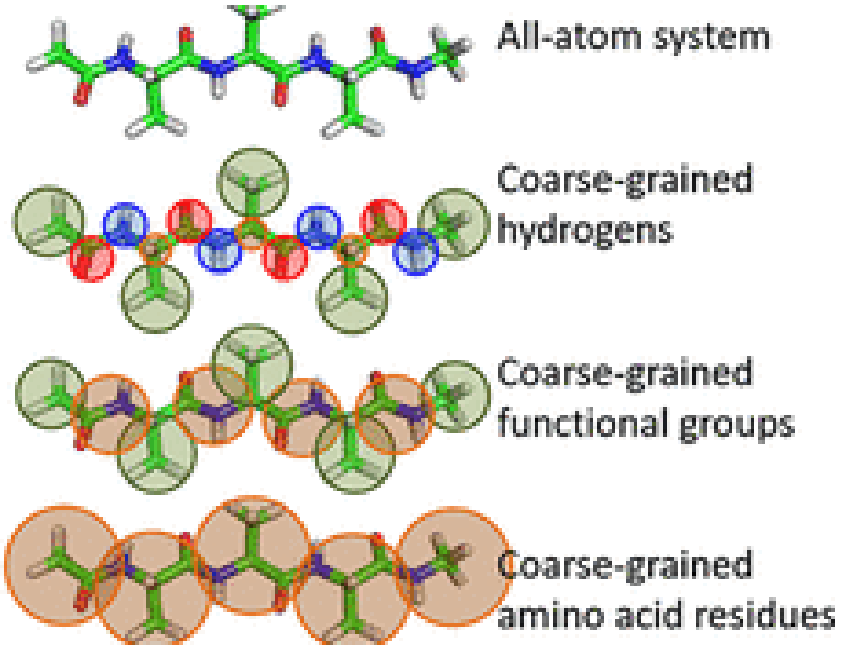
  - How are atoms grouped to define each mesoscale beads?
  - What type of potential terms are required?



$$U_{total}^{A} = U_{nonbonded} + U_{bonds} + U_{angles} + U_{torsions}$$

$$U_{total}^{CG1} = U_{nonbonded}^{CG} + U_{bonds}^{CG} + U_{angles}^{CG} + U_{torsions}^{CG}$$

$$U_{total}^{CG2} = U_{nonbonded}^{CG} + U_{bonds}^{CG} + U_{angles}^{CG}$$

$$U_{total}^{CG3} = U_{nonbonded}^{CG} + U_{bonds}^{CG}$$

Ye et al., ACS Omega 6, 1758–1772, 2021

Bottom-up example II

- Atomistic → Mesoscale: parameterization of a mesoscale force field using atomistic simulation data
- Determine the desired resolution of the mesoscale model

- Perform atomistic simulations of the systems to generate reference energy and force data

- Use numerical optimization / machine-learning methods to find model parameters for the mesoscale (CG) force field that best reproduce energies and forces from atomistic simulations (i.e., minimize L)

$$L = w_{energy} \sum_{configs} \left(\mathcal{U}_{total}^{CG} - \mathcal{U}_{total}^{A}\right)^2 + w_{force} \sum_{configs} \sum_{beads} \sum_{x,y,z} \left(\mathrm{F}_i^{CG} - \mathrm{F}_i^{A}\right)^2$$

Atomistic vs. mesoscale force fields

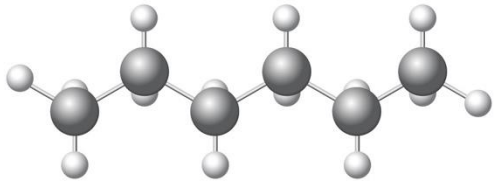
- For atomistic systems
  - $\mathcal{U}_{total}$ typically has a pre-defined functional form (e.g., class II force fields)
  - $\mathcal{U}_{total}$ can also be represented using neural networks; these can provide better accuracy but are ~10-100 times more computationally intensive

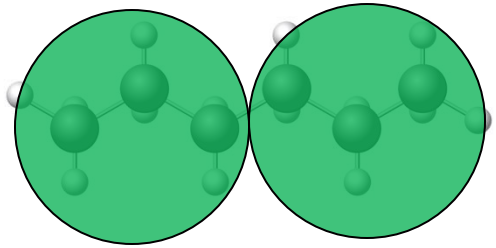
- For mesoscale systems
  - Interactions between mesoscale beads are typically “softer” than those for atomistic systems
  - Pre-defined functional forms are common, but it is often challenging to develop simple analytical functions to describe these interactions
  - Tabulated potentials with interpolation schemes (e.g., splines) offer greater flexibility and accuracy and do not appreciably change the computational cost

Top-down example

- Develop mesoscale force field by using theory to find parameters that reproduce experimental phase diagram
- Determine the desired resolution of the mesoscale model
  - How are atoms grouped to define each mesoscale beads?
  - What type of potential terms are required?

e.g., Hexane



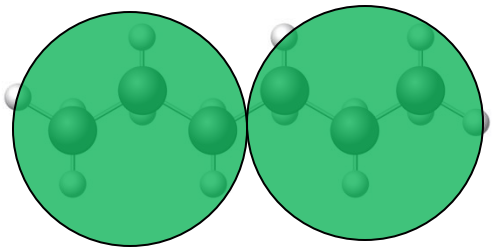


$$\mathcal{U}^{CG}_{total} = \mathcal{U}^{CG}_{nonbonded} + \mathcal{U}^{CG}_{bonds}$$

Muller and Jackson., Annu. Rev. Chem. Biomol. Eng. 5:405–27, 2014

Top-down example

- Develop mesoscale force field by using theory to find parameters that reproduce experimental phase diagram
- Determine the desired resolution of the mesoscale model
- Use theory (e.g., statistical associating fluid theory) + numerical optimization methods minimize deviation from experimental vapor-liquid equilibrium curve



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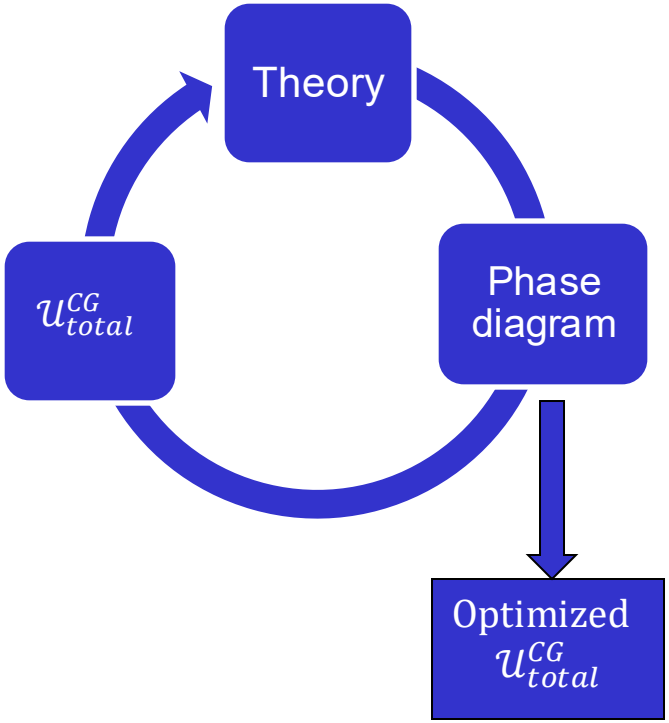

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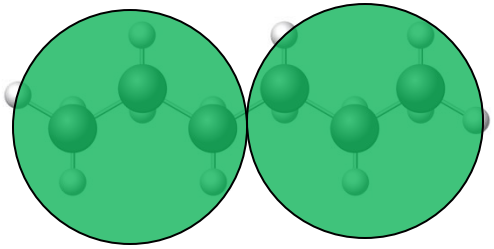

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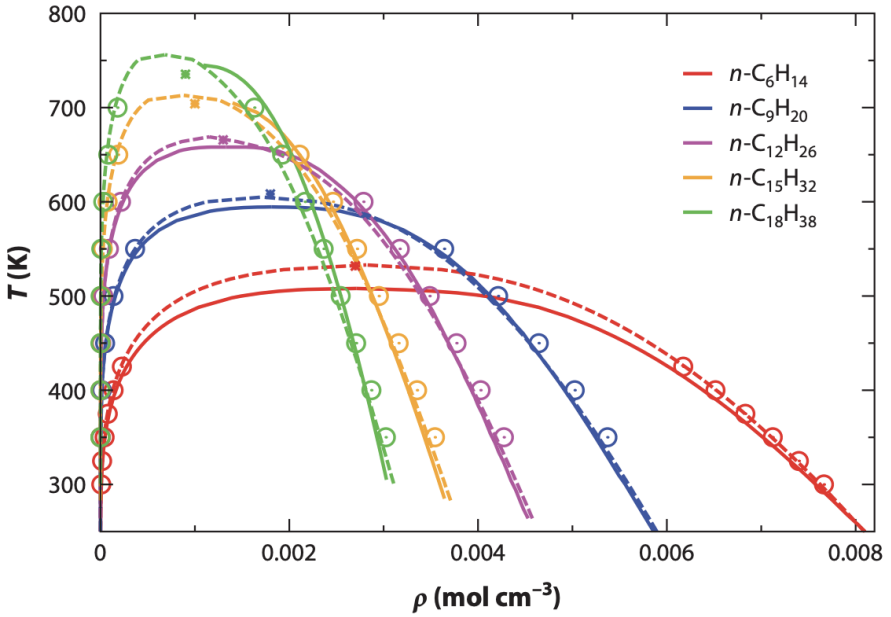

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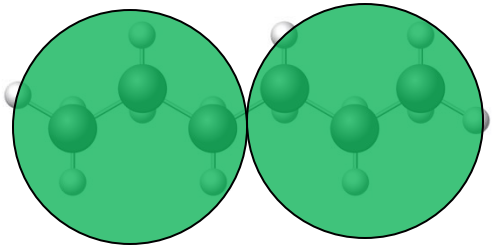

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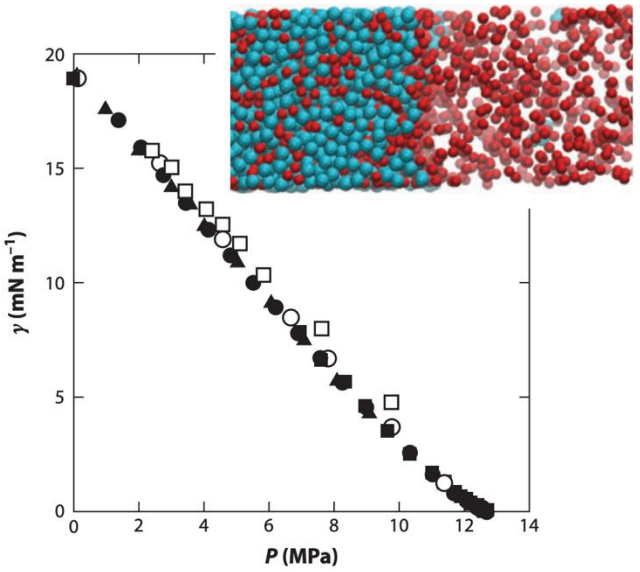


CO_2 + n-$C_{10}H_{22}$: exp (closed), sim (open)

Mesoscale models of soft materials

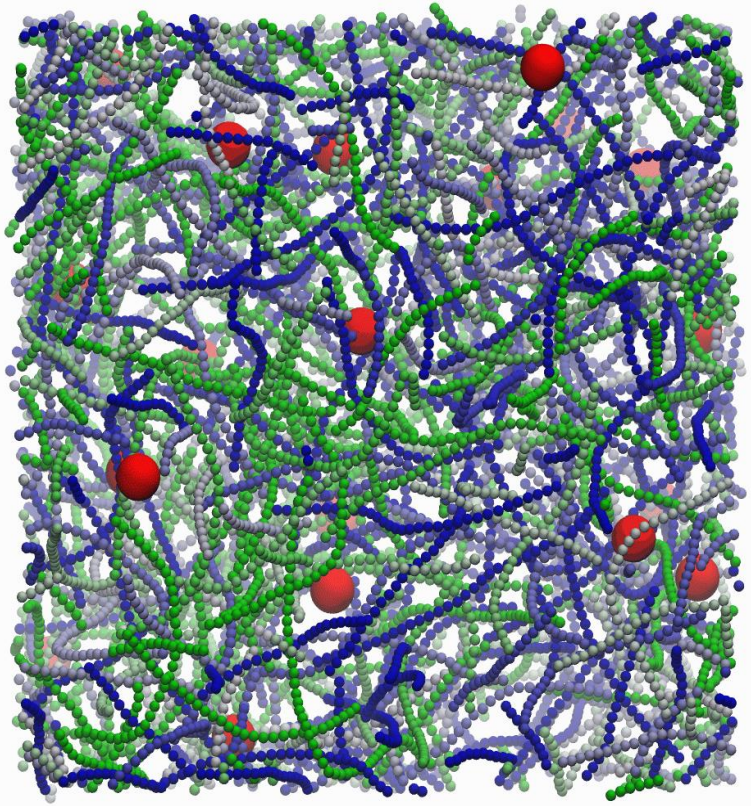
- “Soft materials”: typical interactions strengths on order of thermal energy (~kT)

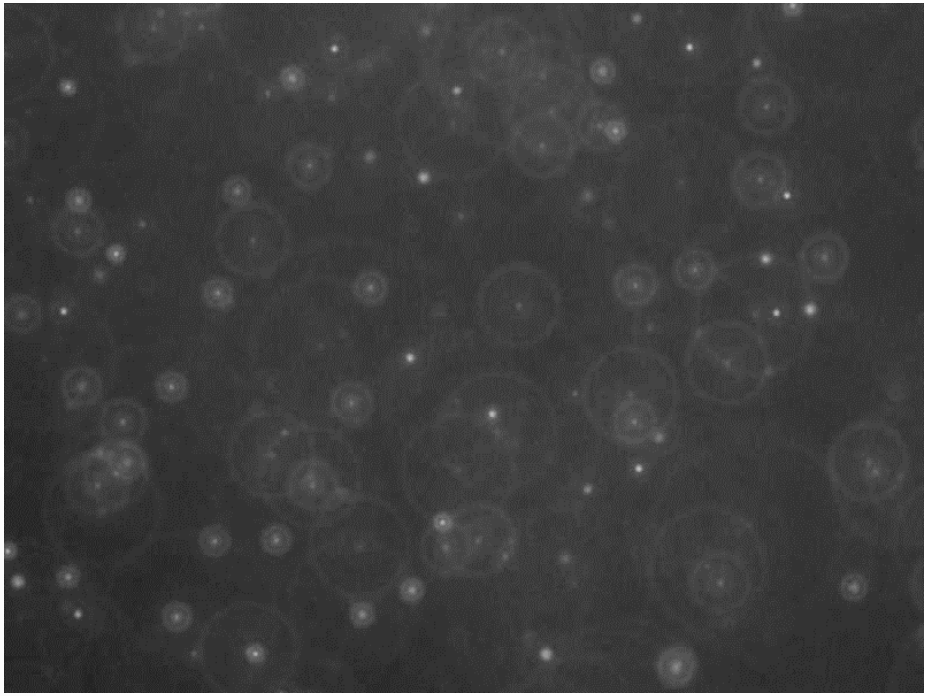
- Examples:
  - Pure fluids (gasses, liquids, supercritical)
  - Macromolecules (polymers, proteins)
  - Suspensions of colloids and nanoparticles
  - Complex fluid (mixtures of the above)

- Phenomena
  - Equilibrium structure (e.g., radius of gyration) and self-assembly behavior (e.g., aggregation)
  - Equilibrium dynamics (diffusion coefficients, viscosity, etc.)
  - Nonequilibrium processes (transport under flow, shearing, etc.)

Examples from Palmer Group

- Modeling nanoparticle and polymer solutions for the design of nanocomposites
- Use mesoscale simulations to test theories and provide improved understanding of experimental observations





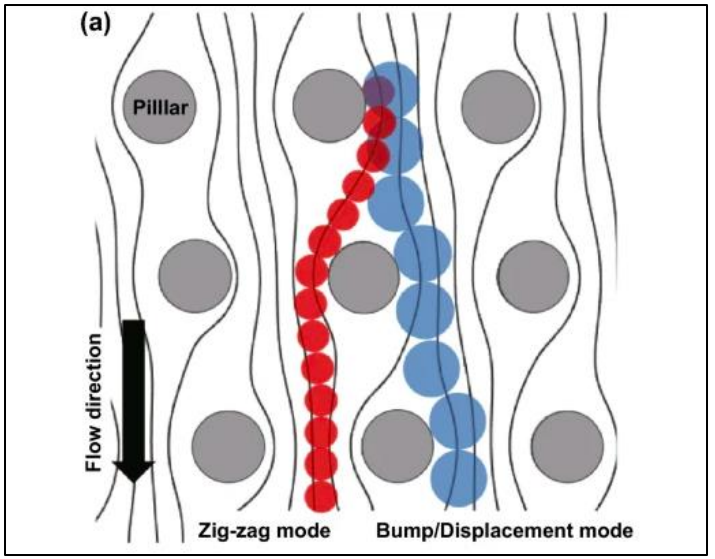
Examples from Palmer Group

- Nanoparticle particle transport through porous media for separations and diagnostic applications

Lateral flow assays

Image credit: NPR

Microfluidic separation



Salafi *et. al*., *Nano-Micro Lett.*, **2019**, 11, 77

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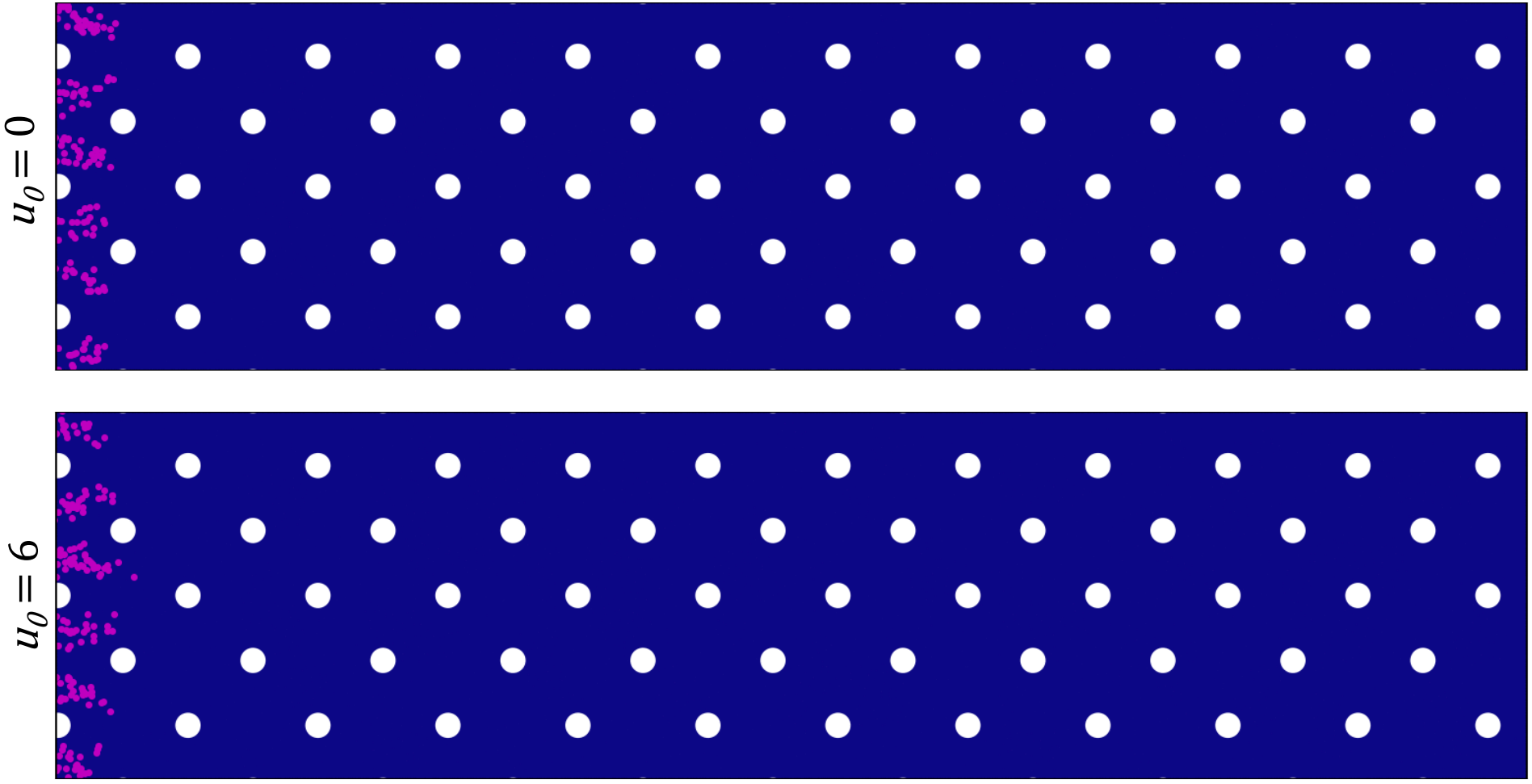
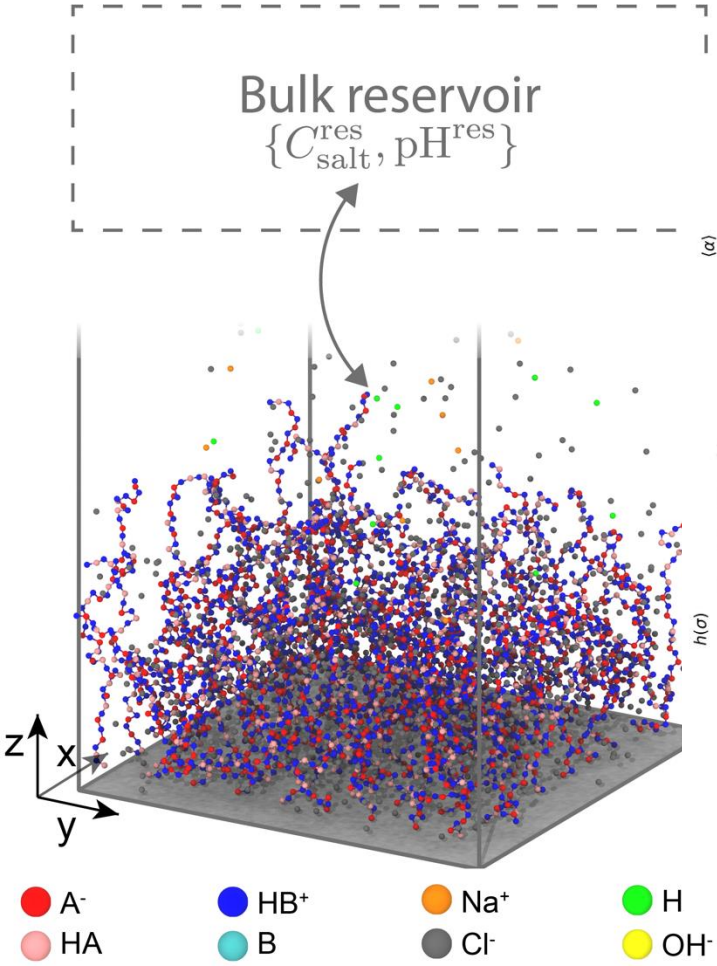
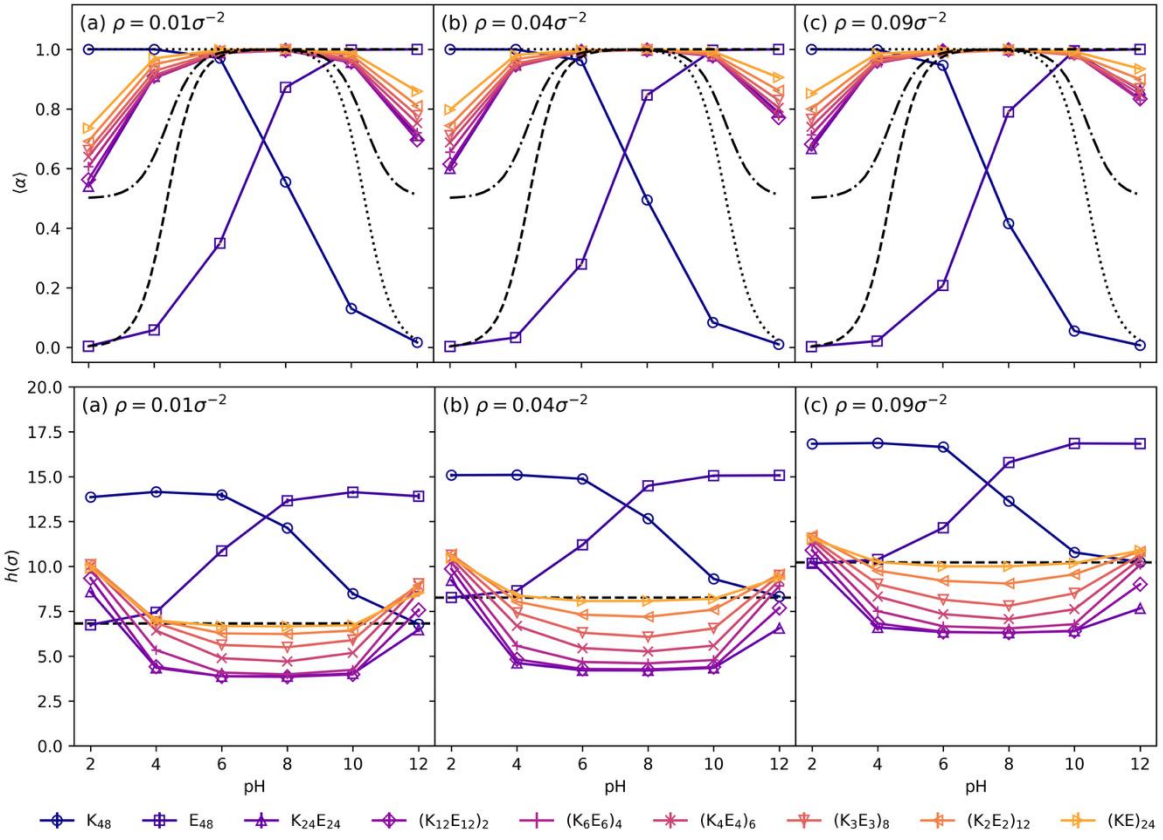


Examples from Palmer Group

- Stimuli-response polymer brushes for sensing, actuation, and self-cleaning surfaces





Workshop overview

- Day 1: Coarse-grained models
  - Session 1: Introduction to coarse graining (this lecture)
  - Session 2: Top-down coarse graining
  - Session 3: Bottom-up coarse graining
  - Session 4: Visualization

- Learning objectives
  - Define the concepts of coarse graining and mesoscale models.
  - Explain basic approaches for top-down and bottom-up coarse-graining.
  - Explain the concepts of potential of mean force (PMF) and implicit-solvent modeling for effective interactions between mesoparticles.
  - Be able to visualize output from molecular simulations.

Workshop overview

- Day 2: Molecular dynamics and Langevin dynamics
  - Session 5: Molecular dynamics (MD)
  - Session 6: Langevin dynamics (LD)
  - Session 7: Periodic boundary conditions I
  - Session 8: Periodic boundary conditions II
  - Session 9: Walls

- Learning objectives
  - Explain the classical mechanics underlying MD, its numerical integration algorithm, and how to choose the integration time step
  - Define the equations of motion for LD and explain how key parameters can be chosen to develop implicit-solvent models.
  - Explain the concept of periodic boundary conditions and how they affect the calculation of structural and dynamics quantities.
  - Be able to model quasi-2D, 1D, and 0D systems using solid surfaces (walls).

Workshop overview

- Day 3: Multiparticle collision dynamics I
  - Session 10: Algorithm, pure solvent
  - Session 11: Coupling to polymers
  - Session 12: Coupling to colloids
  - Poster session

- Learning objectives
  - Explain the basic MPCD algorithm and the importance of including an explicit coarse-grained solvent to capture hydrodynamic phenomenon in mesoscale simulations.
  - Be able to use the MPCD solvent to model hydrodynamic interactions between mesoparticles (polymers & colloids).

Workshop overview

- Day 4: Multiparticle collision dynamics II
  - Session 13: Coupling to boundaries I (wall-driven flow)
  - Session 14: Coupling to boundaries II (pressure-driven flow)
  - Session 15: Reproducibility / open-source software roundtable
  - Session 16: Applied project

- Learning objectives
  - Explain how an MPCD solvent can be coupled to solid boundaries to simulate wall- and pressure-driven flows.
  - Be able to simulate wall- and pressure-driven flows, then analyze simulation results to extract properties of interest.
  - Define reproducibility and explain its importance in computational science. Identify strategies that computational scientists can use to promote reproducibility and accelerate scientific advances.
  - Explain what open-source software is, and identify best practices for using and contributing to it.
  - Apply mesoscale particle-based models to a scientific research problem.

Workshop overview

- Day 5: Wrap up
  - Session 17: Project presentations and discussions.
  - Session 18: What we didn't cover / ask us anything!

- Learning objectives
  - Develop communication skills for computational research.
  - Identify topics for future learning.