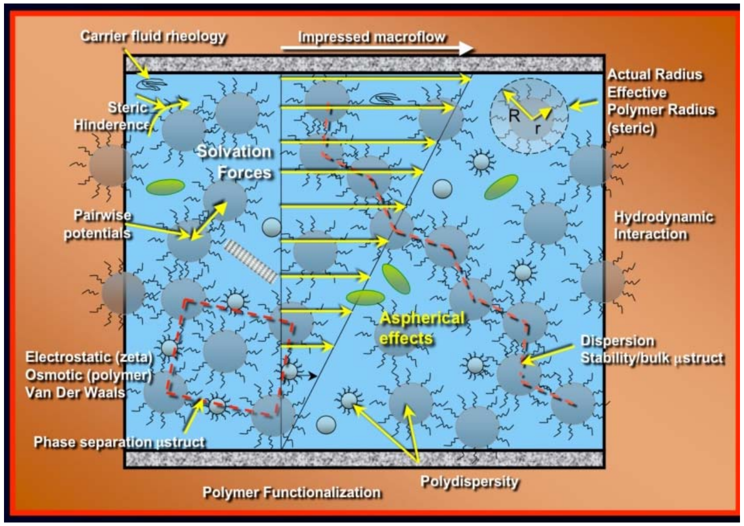
SAND2011-5171C

For presentation at the USNCCM 2010 Conference,25-28 July 2010, Minneapolis, MN.

Application of Conformal Decomposition Finite Elements to Colloidal Suspensions



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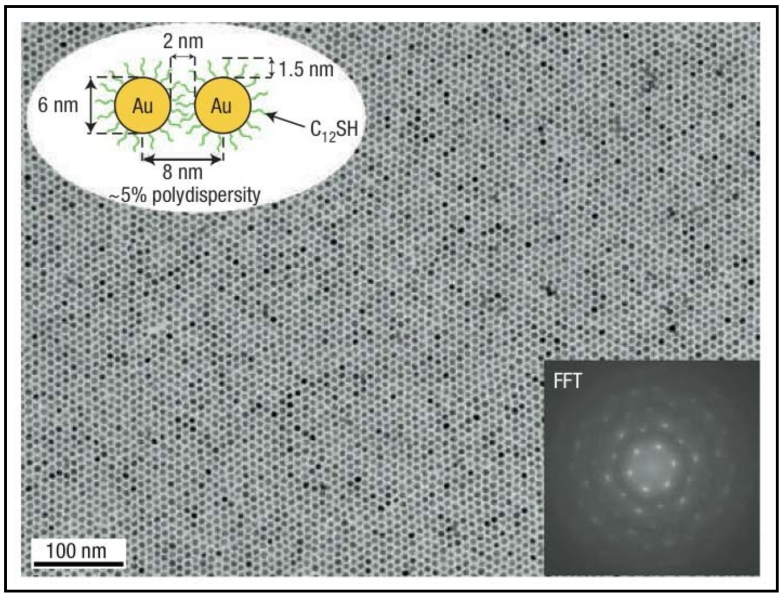
Engineering Sciences Center, SNL

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Nano-Composites

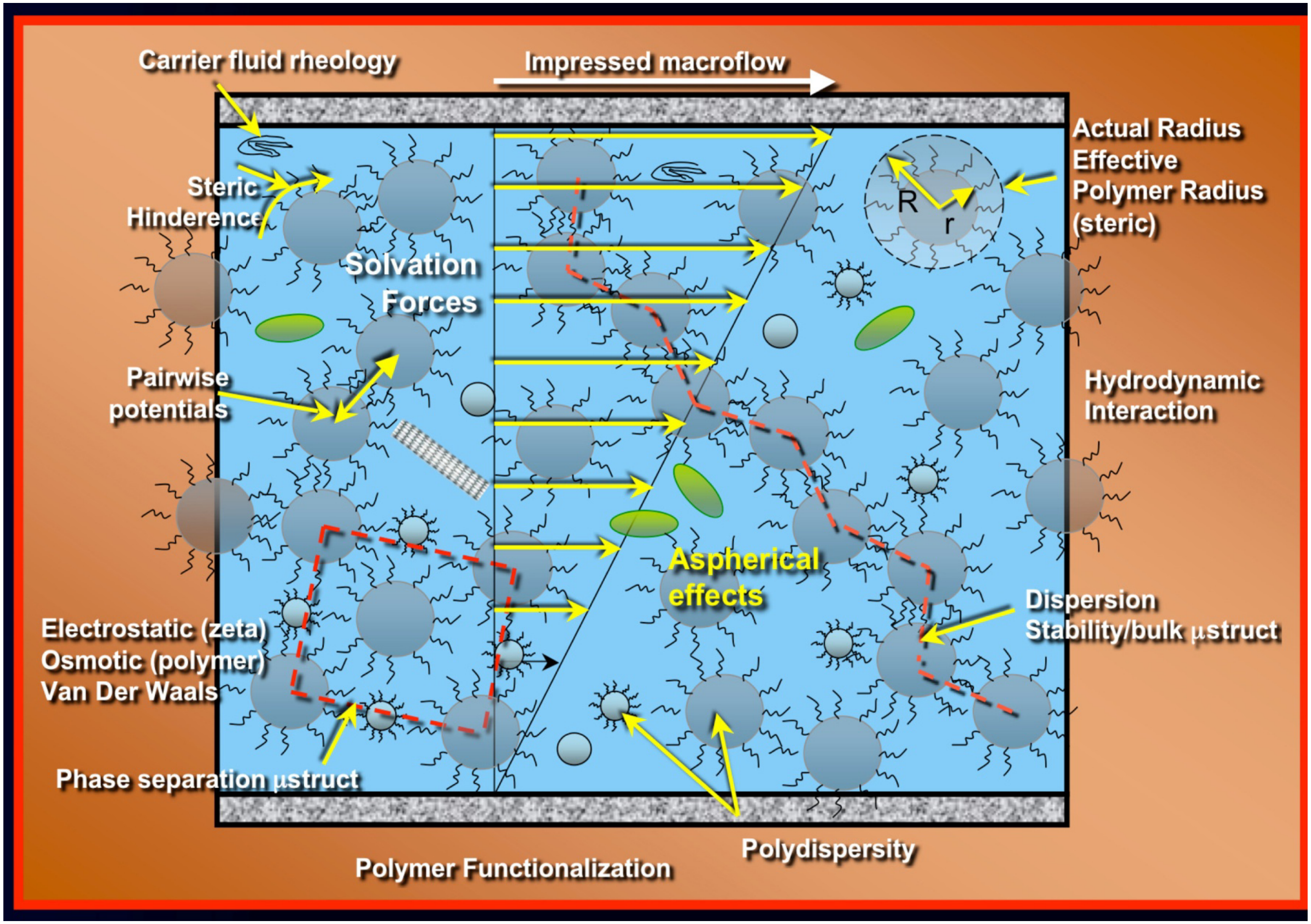
- Nanoparticles can have a profound effect on electrical, thermo-physical, mechanical and optical properties
- Processing particles into “tailored”, functional products through fluidization is a promising approach
- Suspensions exhibit complex rheological and dispersion stability behavior prior to or during processing



– 6*nm* gold particles
– Dodecanethiol capped
– Highly ordered monolayer
– Ordered layers formed on various substrates

Bigioni *et al.*, Nature Mat. ***5***, 265 (2006)

Technical Challenges: rich physical phenomena



Goals

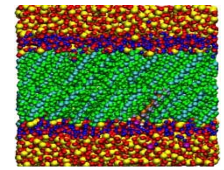
- Achieve a stronger scientific understanding of suspension dispersion stability, rheology, surface modification/interfacial interactions, and solvent effects from numerical simulations
- Develop validated “mod-sim” toolset for nanoparticle suspension and manufacturing process design and control
  - Self-assembly of nanoparticles
    - Evaporation driven self-assembly
  - Rheology of nanoparticle suspensions
    - Dependence on concentration, particle size, shape, shear rate

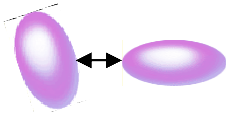
Technical Approach: Integrated Meso-scale Capability

Molecular Dynamics

Molecular scale

nanoseconds
0.1 to 10 nm

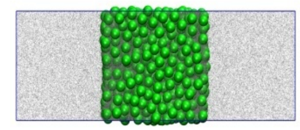


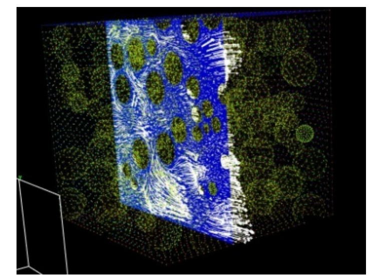


Effective potentials

Particle Scale

microseconds
10 to100 nm





Coupled particle-fluid Interactions:
DEM-NS, SRD, DPD, SD

“Mesoscale”

milliseconds
100 nm-10 μm

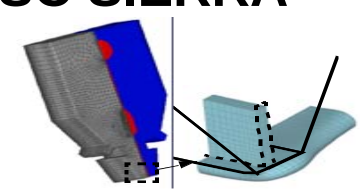
Constitutive Relations **Micro-structure**

Integrated Modeling and Simulation Tool Set
Integrating Tools Across Scales
Experimental Validation
Research (Materials/Numerical methods)

FEM, ASC SIERRA APPS:

ARIA,

GOMA

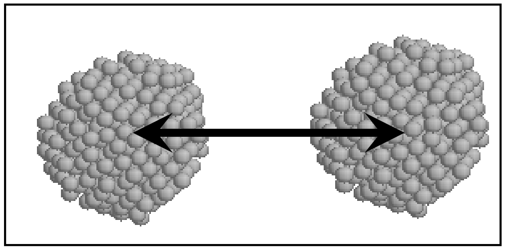


Engineering Scale

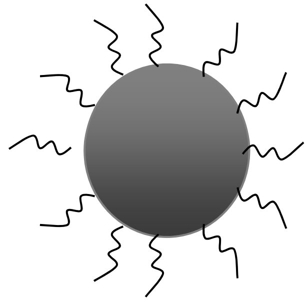
Seconds
mm

Coarse Graining to the Meso-scale

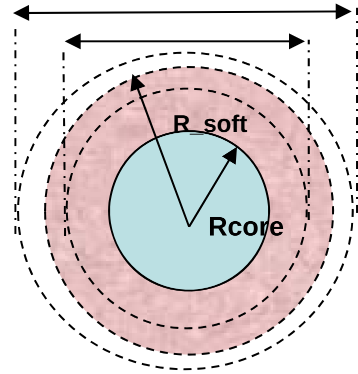
Particle



Integration to Hammaker's Equation and equivalent



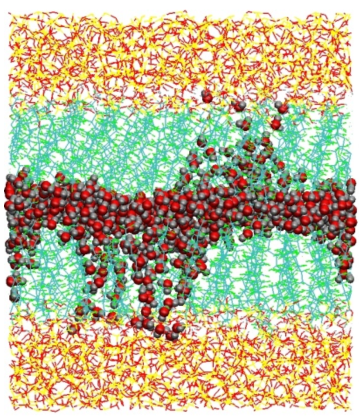
Osmotic and steric/structural representation



- Computational standoffs
- Porous constants
- Polymer layer parameters
- Structural constants (polymer
- And hard sphere)

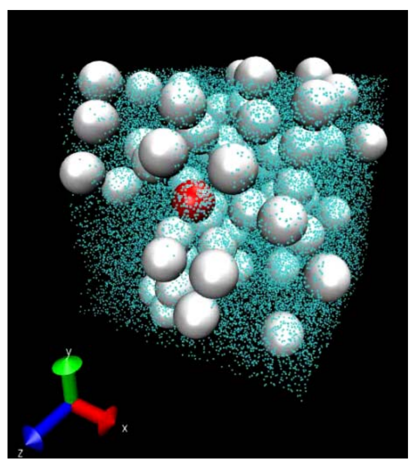
Solvent

Molecular Dynamics

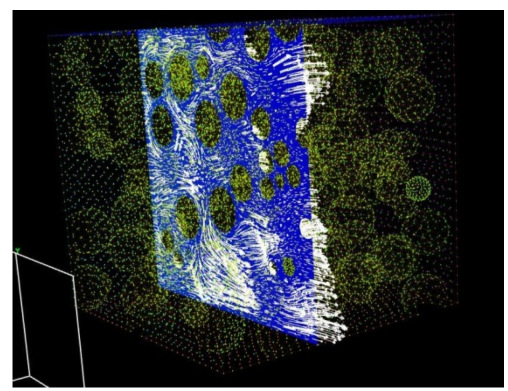


Grest et al.

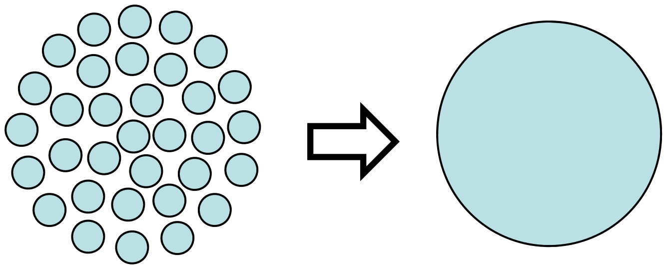
Blobs->SRD/DPD: dual particle approach

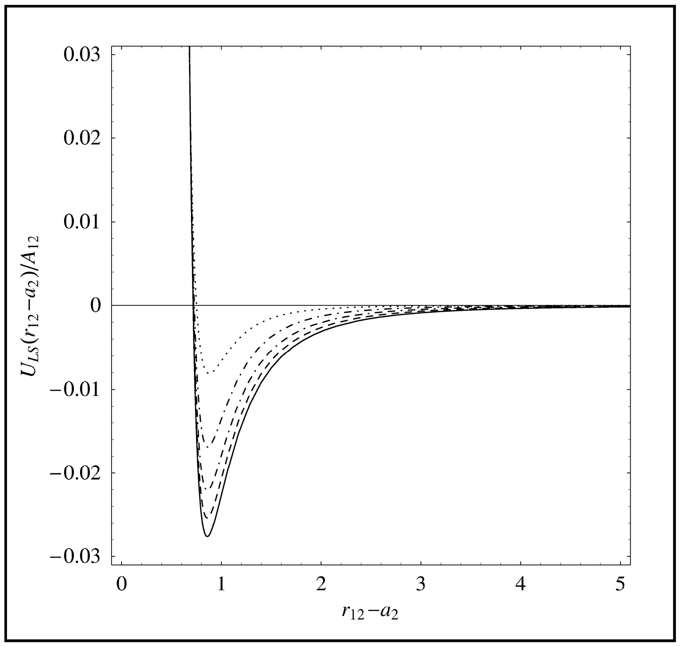


Continuum: FEM, SD

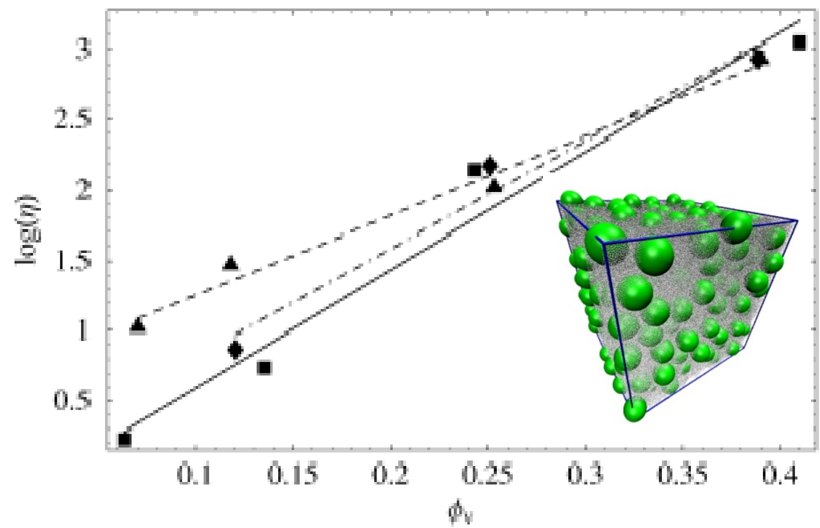


Colloidal Model: First Approach





- Integrated Lennard-Jones potential represents colloidal particle[1]
  - Hard spheres are poor model since they phase separate for disparate sizes
  - Hamaker constant A_{ij} represents pairwise interaction strength

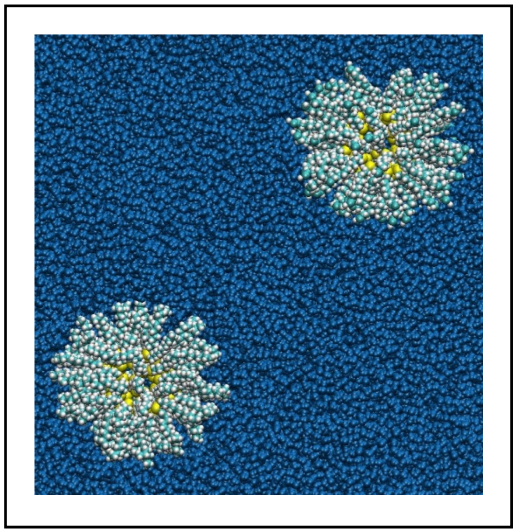


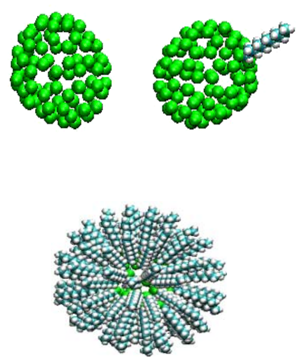
- NEMD Simulations – d = 5.0 (■), 10.0 (▲) , 20.0 (◆) **In't Veld et al. (2008)**

[1]R. Everaers and M.R. Ejtehadi, Phys. Rev. E ***67***, 41710 (2003)

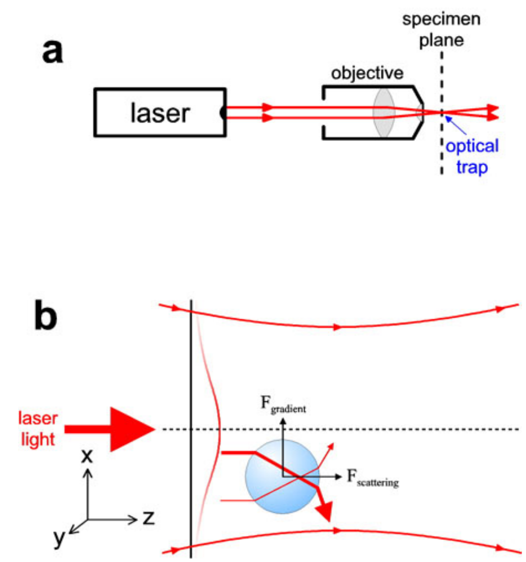
Effective Potential Development

- Molecular dynamics. How small can we go with continuum mechanics principles? Determining interparticle potentials for mesoscale?
  - Velocity dependent and independent parts
  - Various formulations
- Direct force measurement (IFM, Optical Trapping)





Lane et al. (2008)



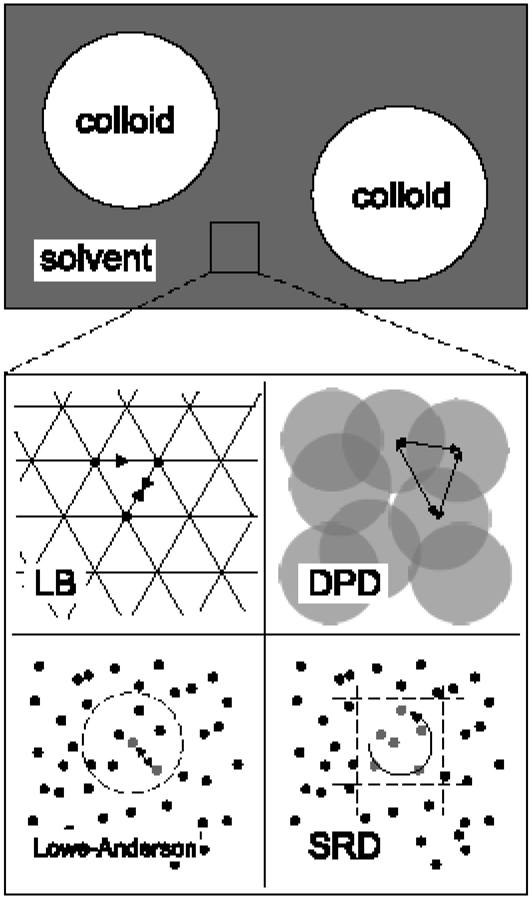
MD of actual Gold/Thiol/Water System (Dynamic and Equilibrium)

Optical Tweezers Measurements

Accurate effective pair potentials required for simulations of nanoparticles in suspension

Simulating the Solvent

- Computational cost
  - **Explicit atomistic solvent requires calculation of all pair-wise solvent-solvent/colloid interactions**
    - **typically *many* orders of magnitude more solvent atoms than solute particles**
    - **light, relatively fast dynamics => short timesteps**
  - **Coarse-grain: Average over fast degrees of freedom**
    - **all => Generalized Langevin dynamics of colloids**
    - **some => coarse-grained solvent with reduced # of solvent dof's, resolving longer length and time scales**
- Multiple "coarse-grained" methods to capture hydrodynamics
  - **Particle-based, coarse-grained, "explicit" solvent**
    - **DPD solvent**
    - **SRD solvent treated as massive, ideal fluid, point particles**
  - **Continuum-based "implicit" solvent**
    - **BD (approximate hydrodynamics – $F_H \sim 6\pi\mu a$, Oseen Tensor, Rotne-Prage Tensor, Fast-Lubrication, etc)**
    - **SD/BEM (creeping Stokes equations)**
    - **LB**
    - **Solve full continuum Navier-Stokes equations numerically (e.g., FEM)**
      - **MOST GENERAL**



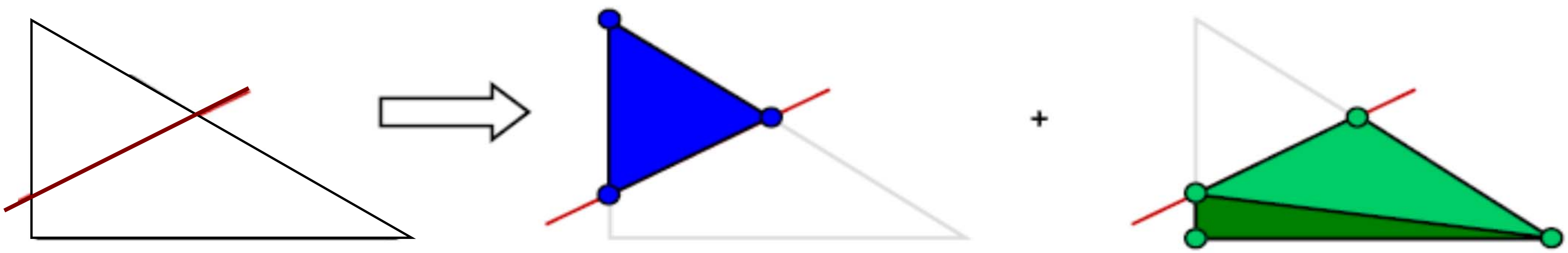
From Padding and Louis (2006) cond-mat/0603391

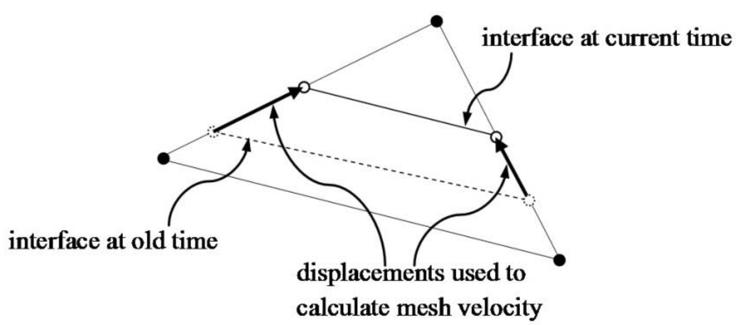
Motivation for FEM Approach

- General toolset parameterized for speed and accuracy
  - Validate computational expedients
    - Telescoping of timescales
    - Artificial compressibility
    - Other approximations to the “physics”
- Numerical infrastructure is general
  - Non-Newtonian rheology
  - “Multi/Coupled physics”
  - Aspherical particles
  - Inertial terms ($\partial u/\partial t$ and $u \cdot grad(u)$)
    - Transient regimes & memory effects
    - High shear rates
    - Larger particles

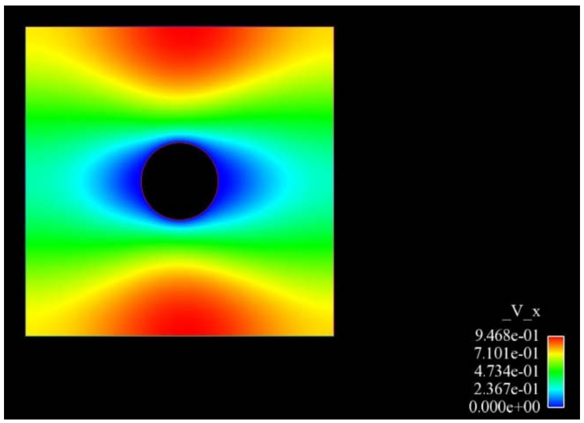
Coupling Colloids to Solvent

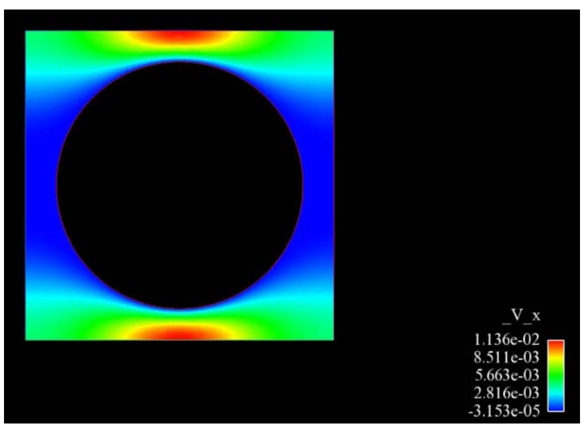
- CDFEM (talk by D. Noble)
  - Sharp interfaces
  - Physical models for boundary conditions
    - No-slip - Dirichlet

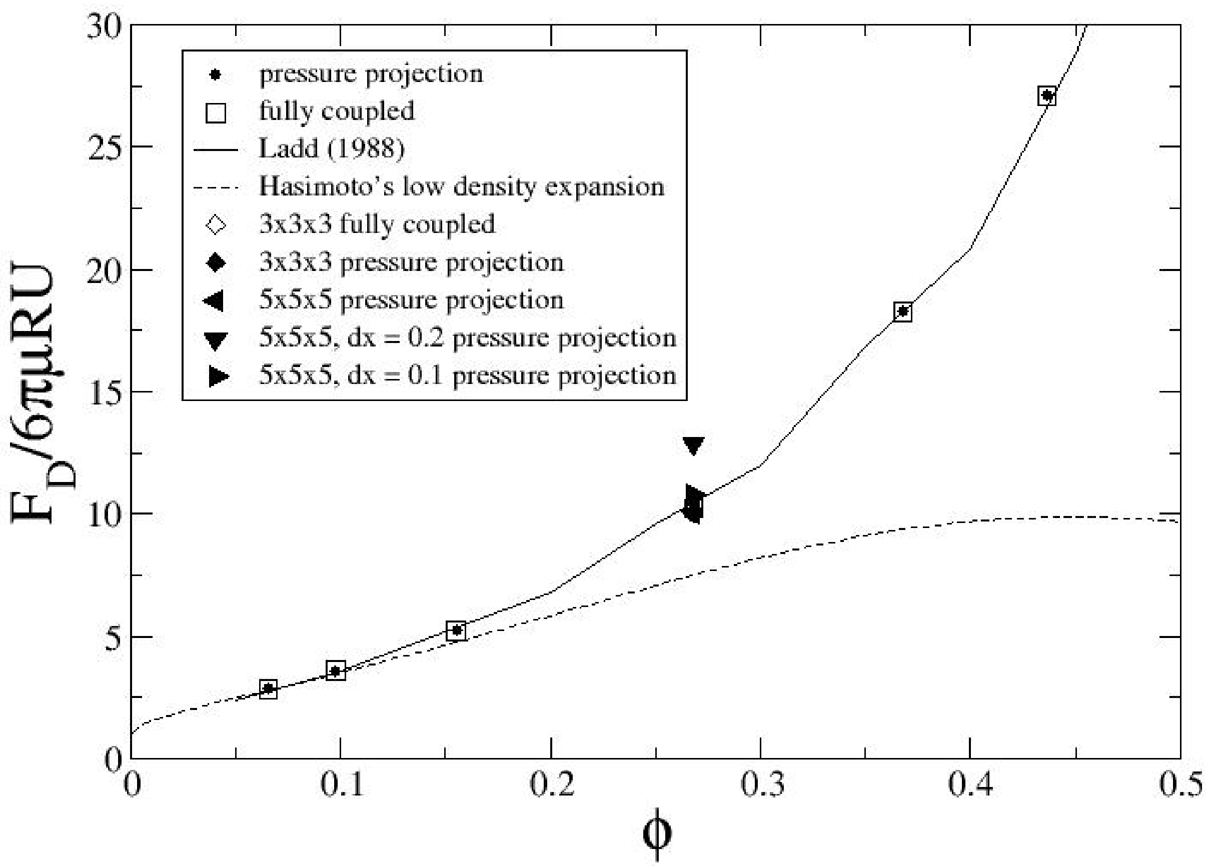




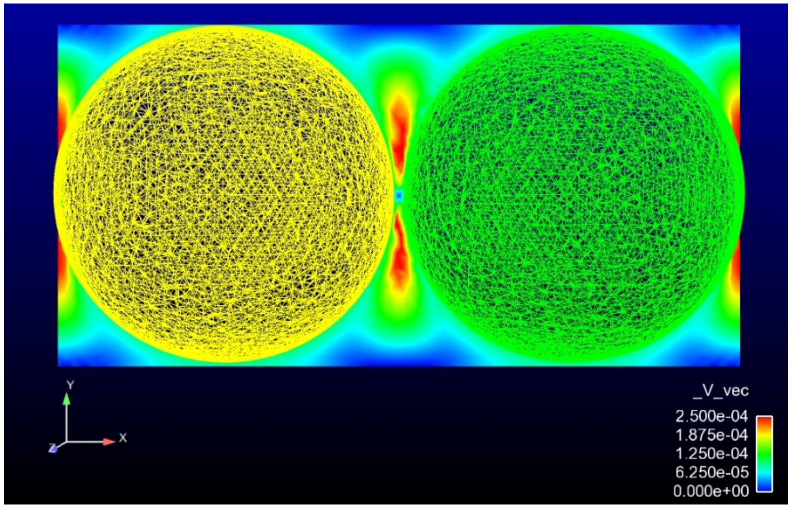
“Static” CDFEM: Translational Friction Coefficient in Stokes Flow

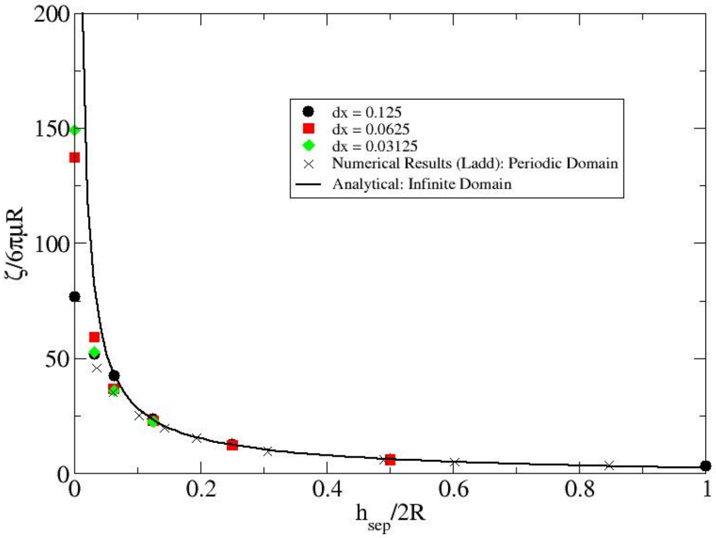


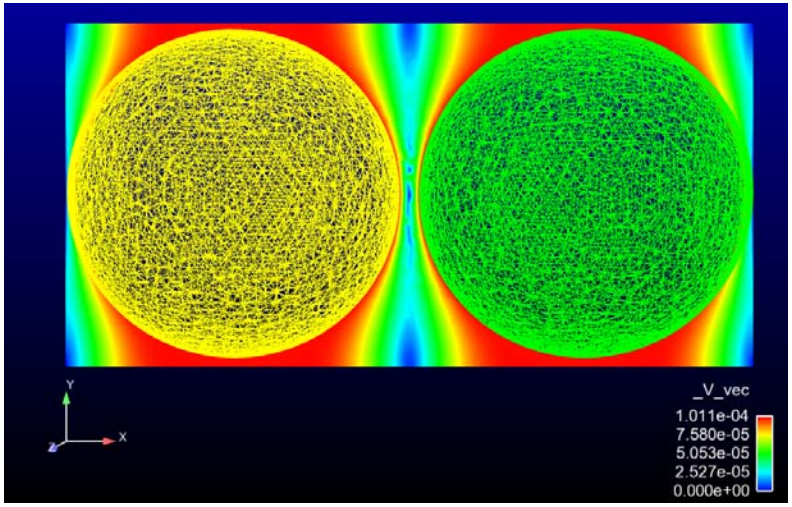


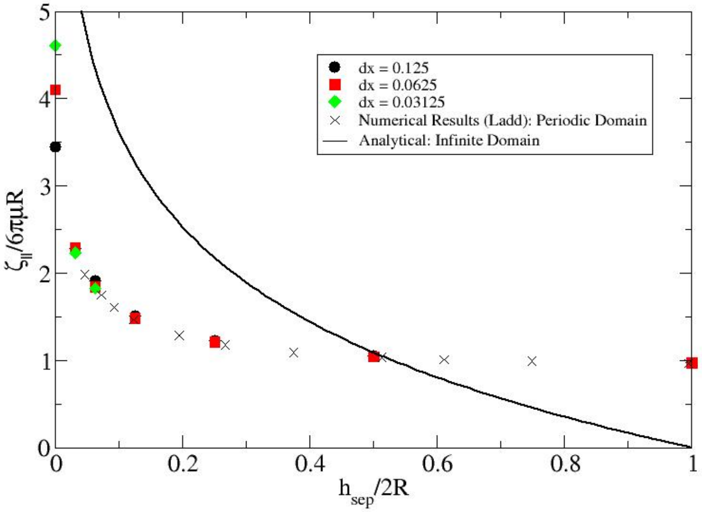


“Static” CDFEM: Lubrication Forces in Stokes Flow



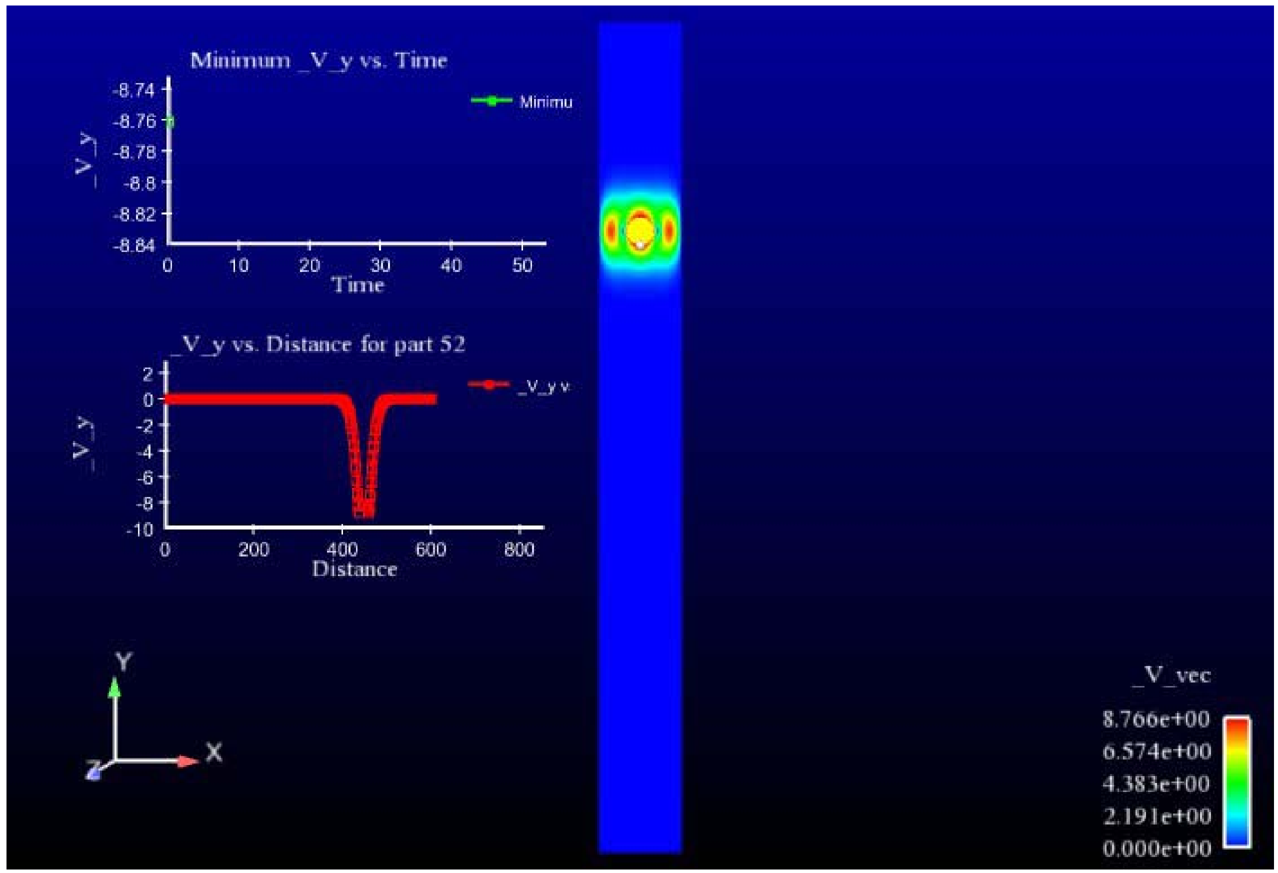






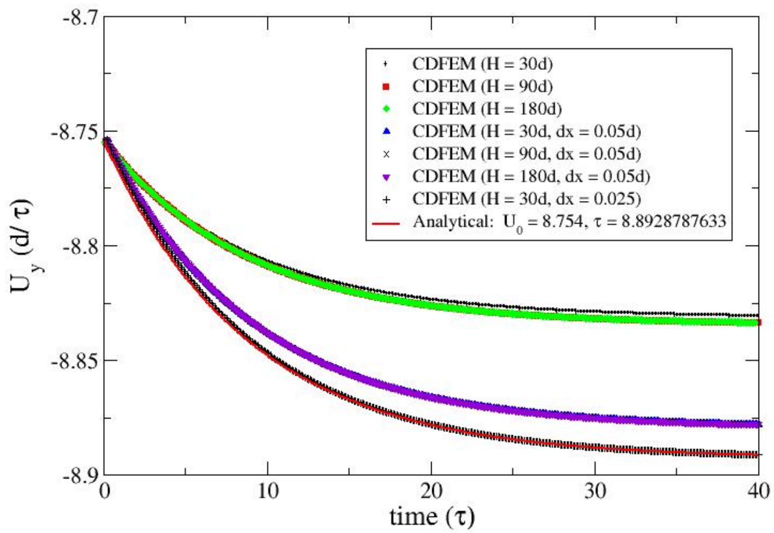
“Quasi-static” CDFEM

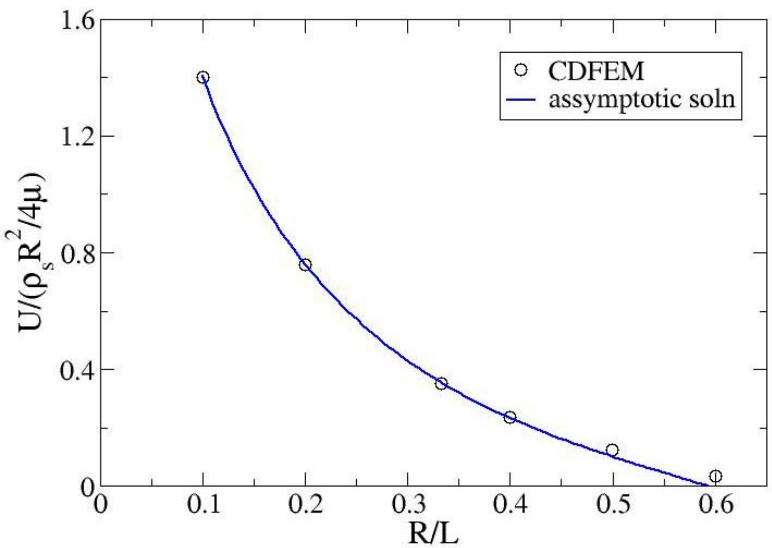
- Couple FEM (Sierra/Aria) and MD (LAMMPS)
- Fluid: Stokes creeping flow
  - Dirichlet BCs
  - Q1Q1
  - PSPP
- Particle
  - Velocity Verlet



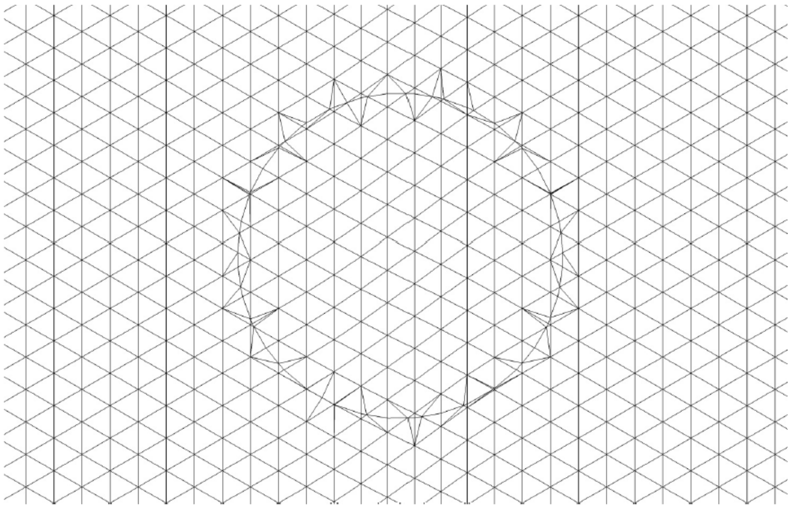
Quantitative Results

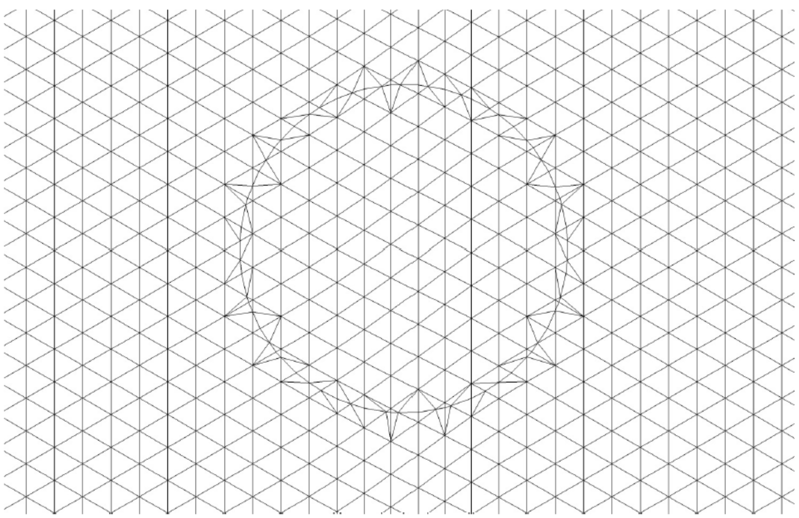
- Fluid:
  - $\rho_f = 0$
  - $\mu = 1.0$
- Solid:
  - $\rho_s = 1.0$
  - $dt_s = dt_f = 0.1$
- 2D asymptotic solution

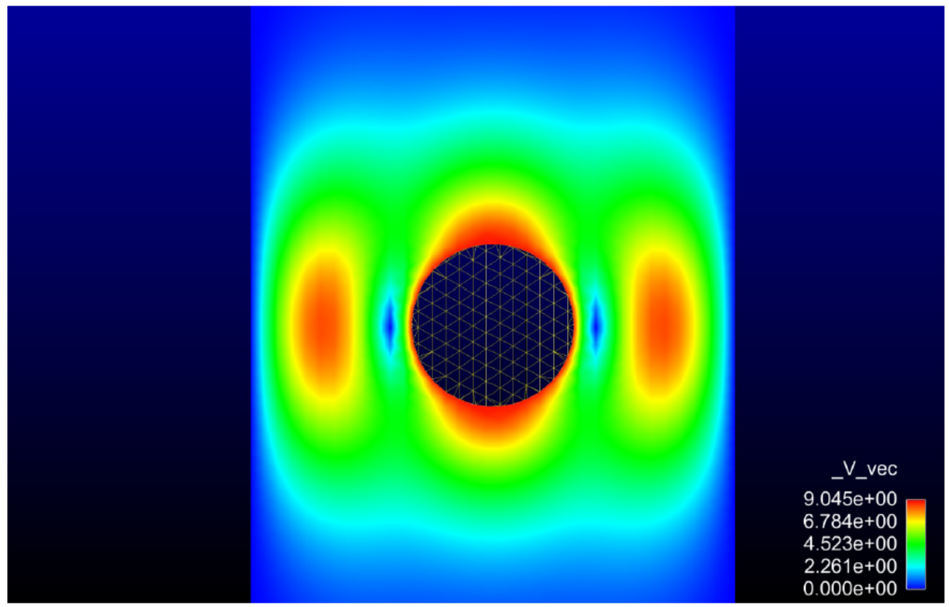




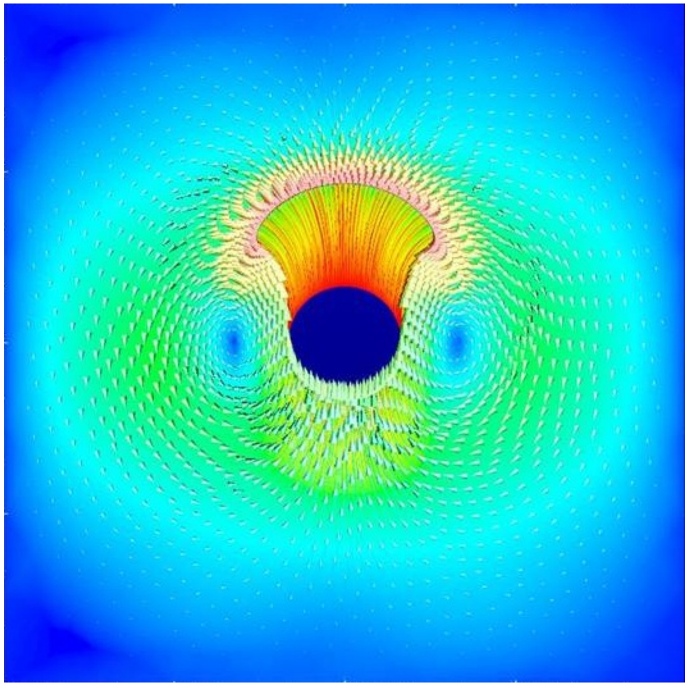
Close-ups: Mesh and flow field







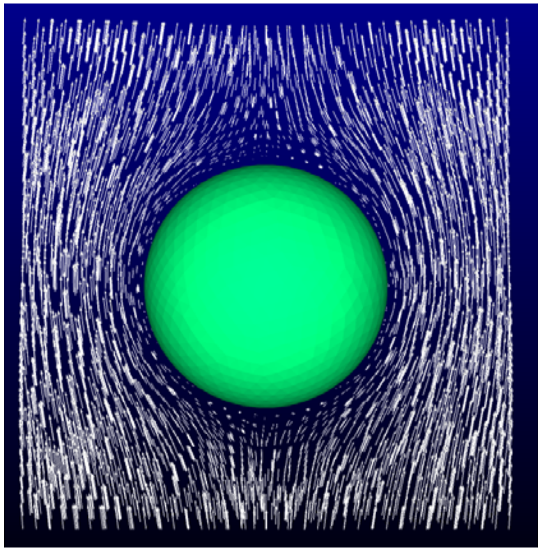
3-D Transient Stokes Flow Verification



Verification Problem

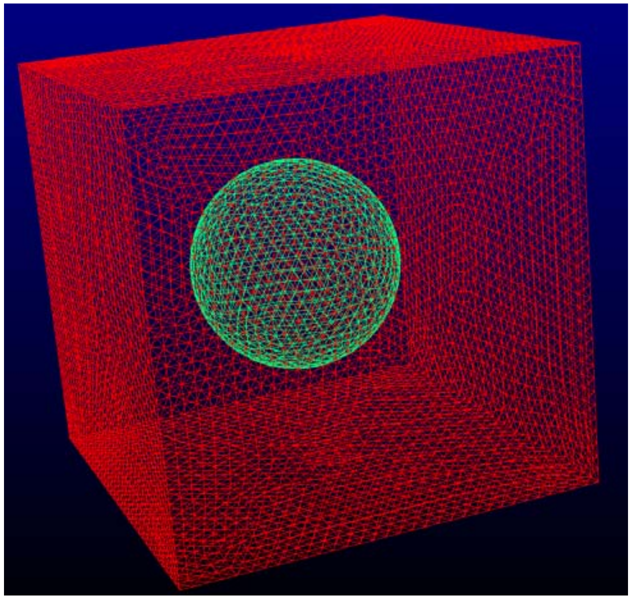
- Transient motion of a 3-D solid sphere under a square-force pulse
  - Analytic solution Felderhof, B. U. (2007) used as velocity boundary condition on all domain boundaries
  - Followed motion of particle in accelerating frame of reference of moving particle

$$\rho_f \frac{Du}{Dt} + \rho_f \frac{du_s}{dt} = \nabla \cdot T$$



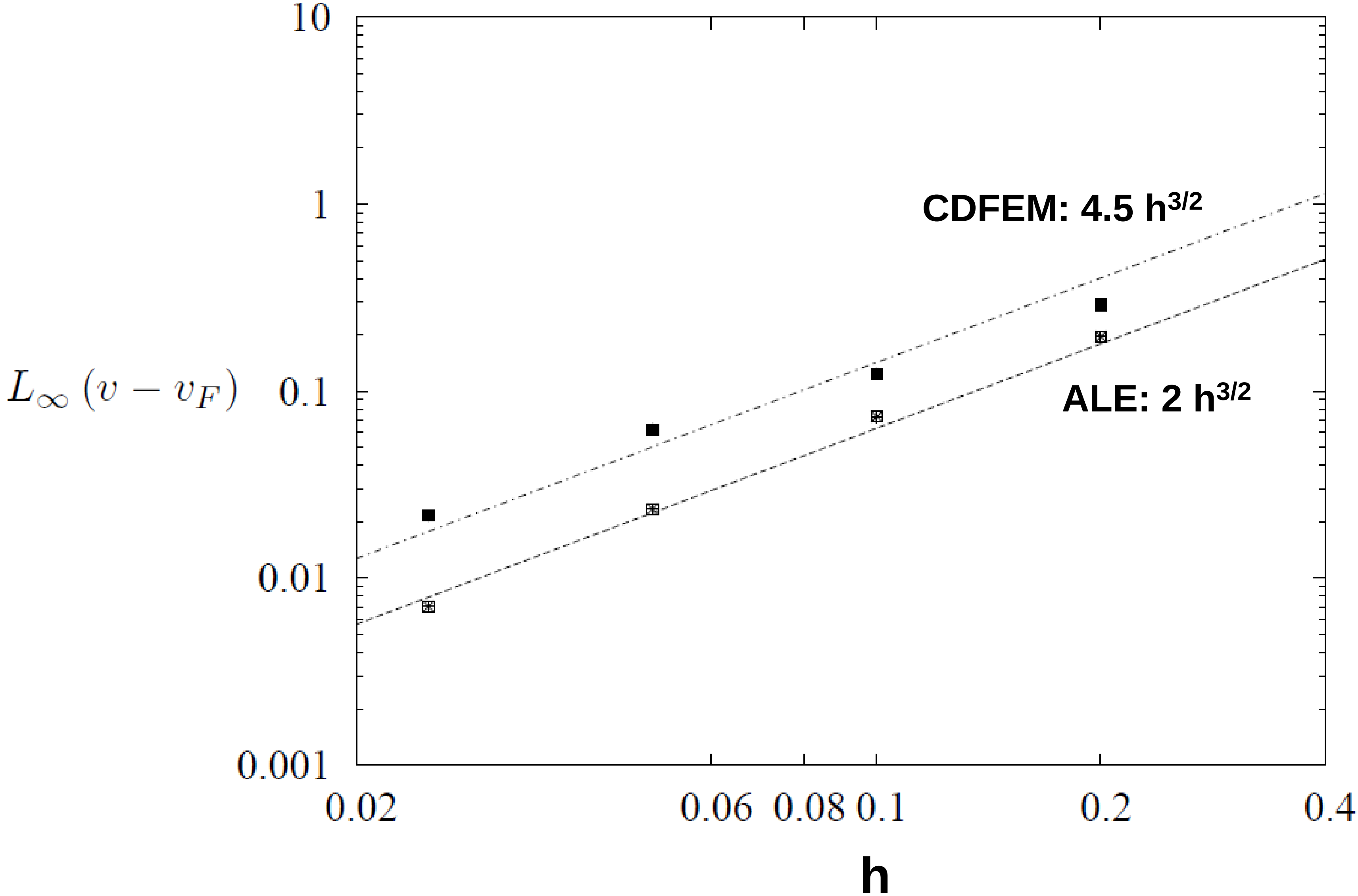
Verification Problem

- $r_s = 1$
- $\rho_s = 1.3$
- $\rho_f = 1.3$
- $\mu = 1$



- Box dimensions: 4 x 4 x 4
- Force = 1 for $0 \le t \le 1$, F = 0 for $t > 1$
- Start simulation at $t = 0.1$, avoid singularity at $t = 0$

Verification results at t = 0.15



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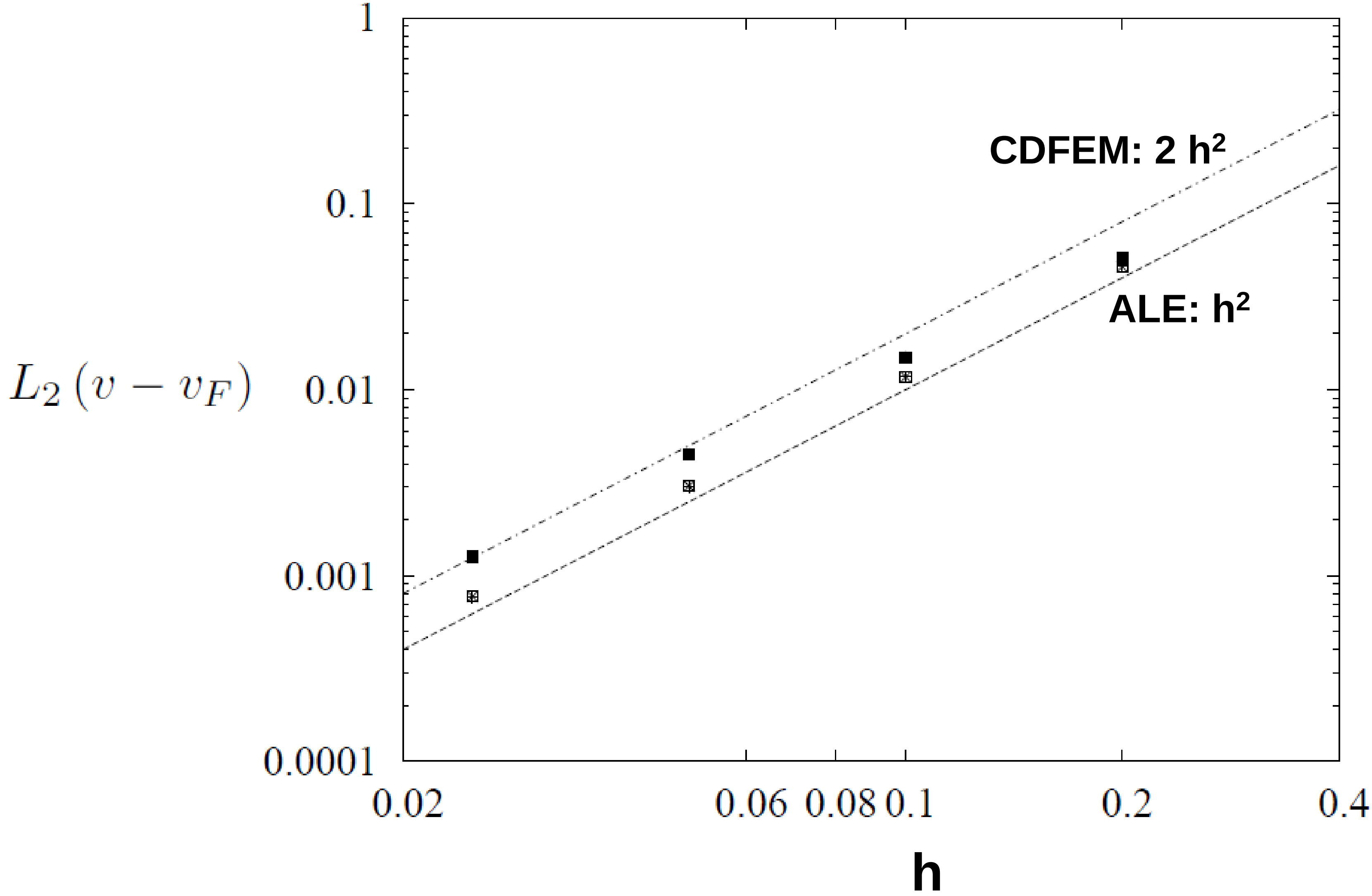

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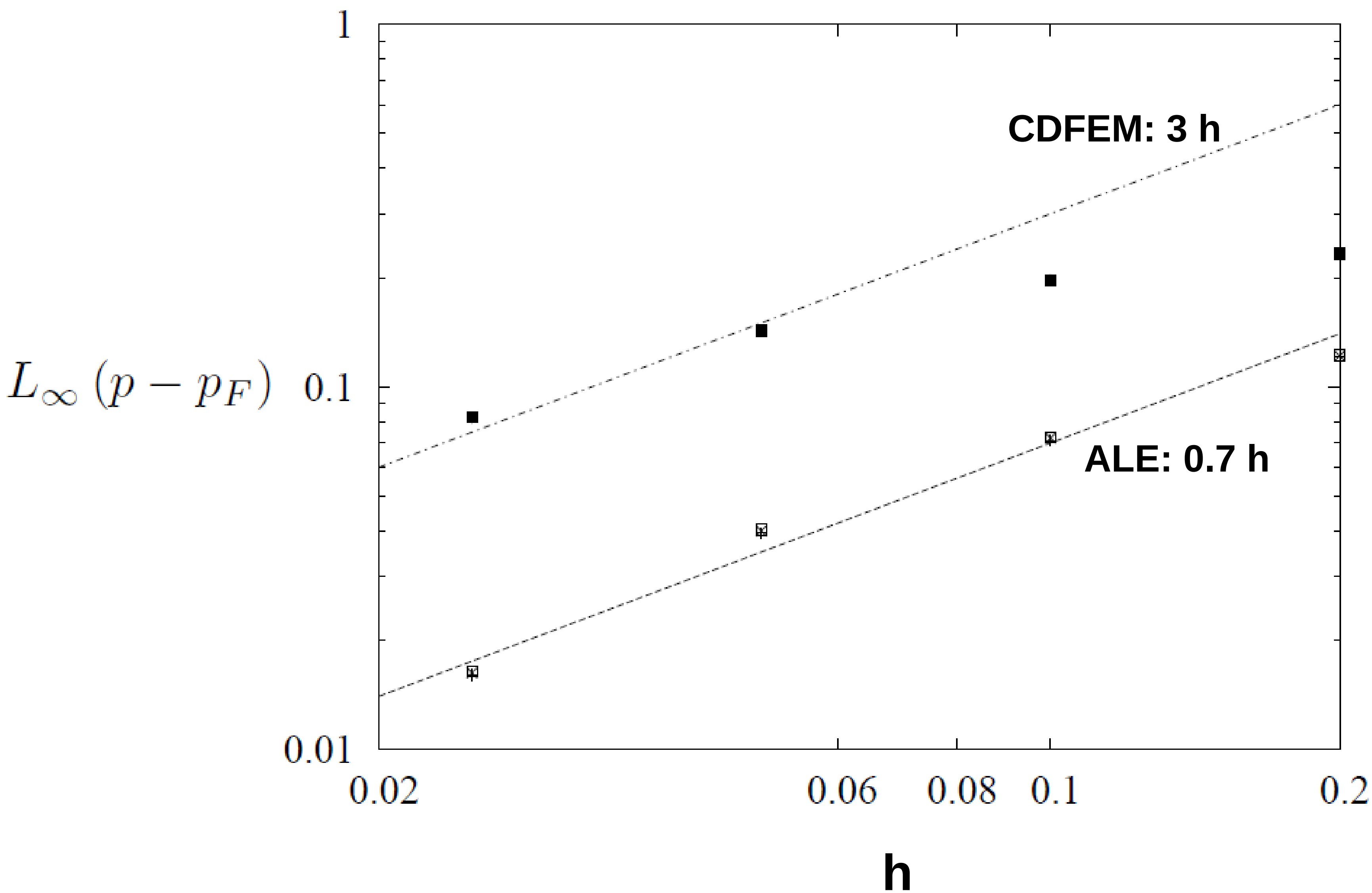

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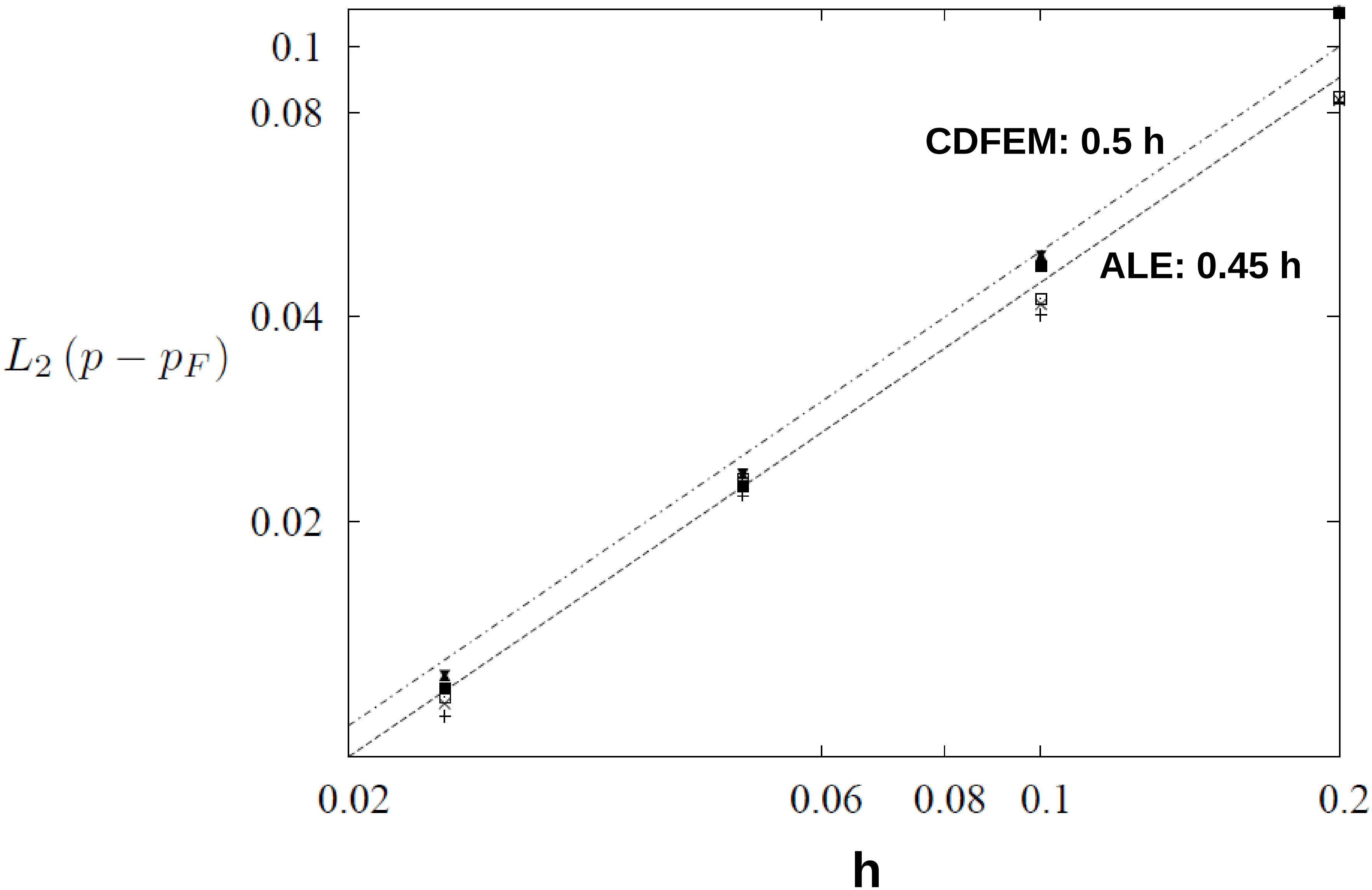
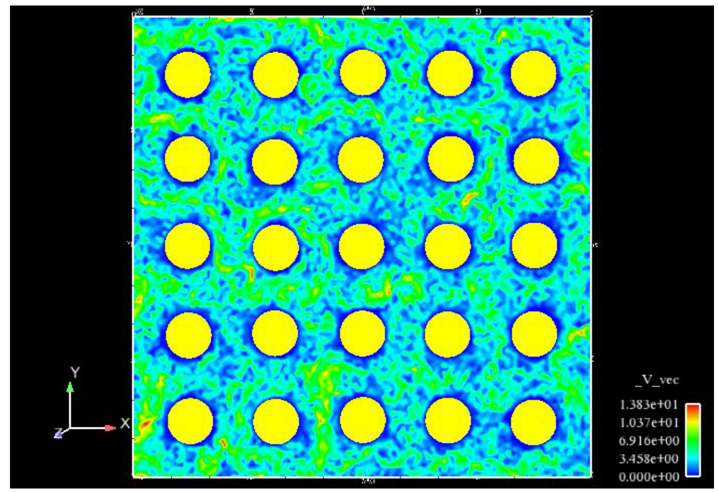
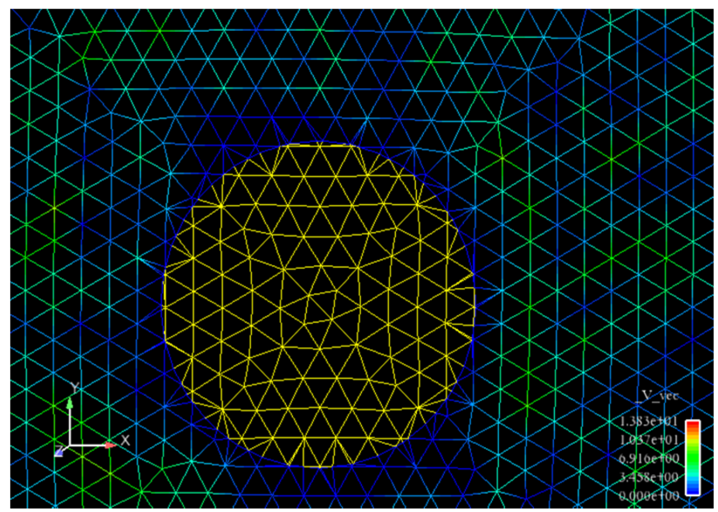


Toward Colloidal Dynamics: CDFEM and Fluctuating Hydro

- Coupled Sierra/Aria and LAMMPS via Mezzo
  - Monodisperse disks
  - $\rho_f = 0.6$
  - $k_B T = 1.0$
  - $\nu = 1.67$
- Can do 3D, no problem
- Parallel, no problem
- Currently
  - quasi-static fluid (creeping Stokes)
  - Explicit time integration of colloid dynamics





Conclusion

- Nanostructured Materials achieved through suspension based processing of nanoparticles requires understanding of
  - dispersion stability
  - bulk rheology
  - induced assembly and structure from volume reduction

- We are advancing a mod/sim platform to meet these needs which targets a scale that bridges between the molecular regime and the engineering regime

- CDFEM is playing a significant role
  - More validation of Quasi-static and transient problems forthcoming

Acknowledgements

- Gary Grest
- Matt Lane
- Matt Petersen
- Ahmed Ismail
- Steve Plimpton
- Mike Brown