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Sandia National Laboratories

NNSA National Nuclear Security Administration

Particle Interaction Measurements

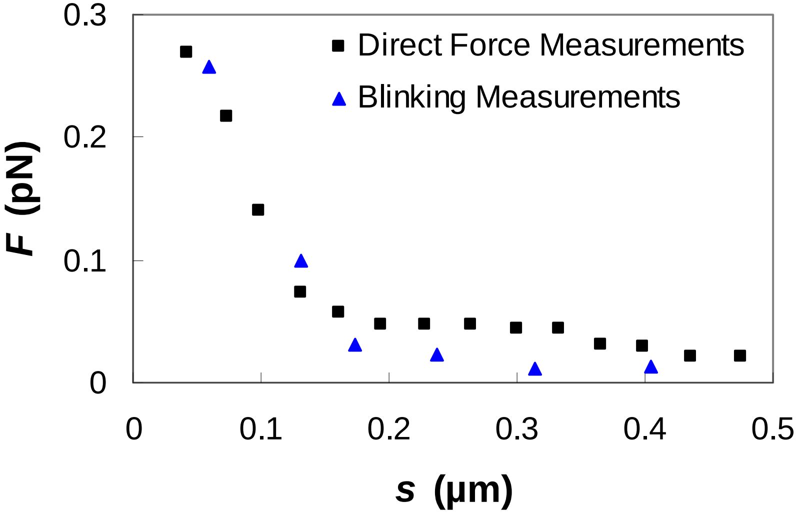
T. Koehler, A. Grillet, E. Dufresne, C. Brotherton *et al.*

- Outline
  - Quick comparison of our capability and Gutsche et al.
  - Detailed summaries of our & Gutsche et al.
  - PDMS system
  - Why (Anne thinks) you need an experimental component

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.

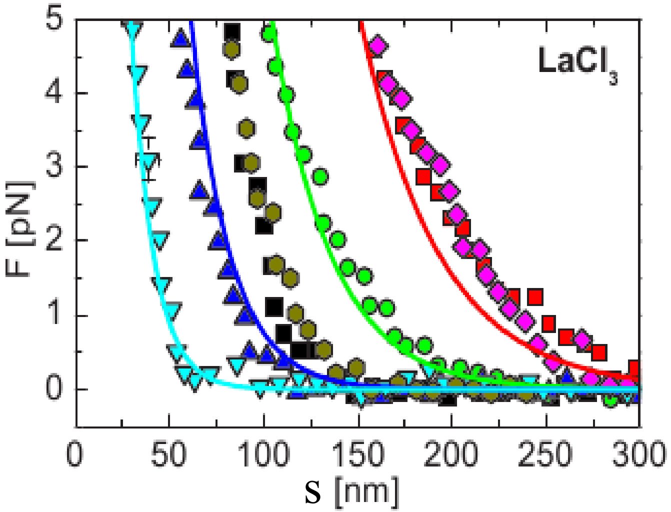
Comparison of Gutsche & Grillet

Koehler, Grillet et al.



- **Experimental System:**
  - 2.47 ± 0.02 µm Silica in DI H_2O
- **Operating Parameters:**
  - Camera: 800 × 600, 500fps
  - Blinking: ~2,000 trajectories, 30ms
  - Direct: Trap Stiffness= 0.026 pN/nm
  - Refined intensity peak-finding algorithm (~10 nm resolution)
- **Errors:** Spatial: ±20 nm; Force: ±10 fN (0.01 pN)

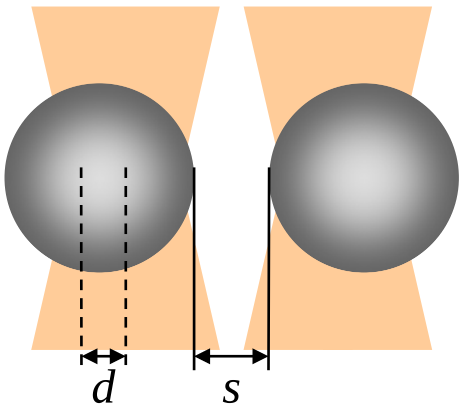
Gutsche et al. (2007)



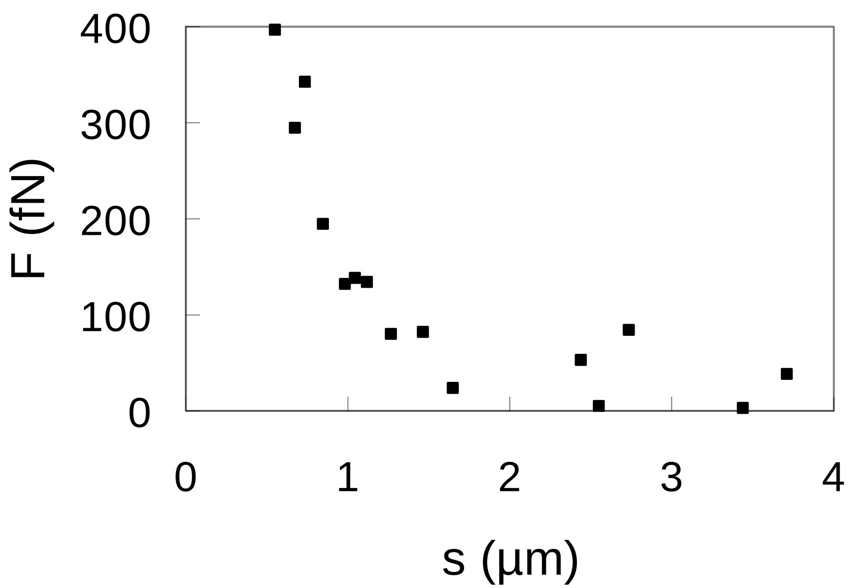
- **Experimental System:**
  - 2.26 ± 0.02 µm dia. Polystyrene
- **Imaging and Processing:**
  - Camera: 1392 × 1040, 30 fps
  - Edge detection center-finding algorithm (~2 nm resolution)
- **Trap Stiffness:** 0.085 pN/nm
- **Errors:** Spatial: ±10 nm; Force: ±0.3 pN

Sandia's Direct Force Capability

$$F = k_{trap} d$$

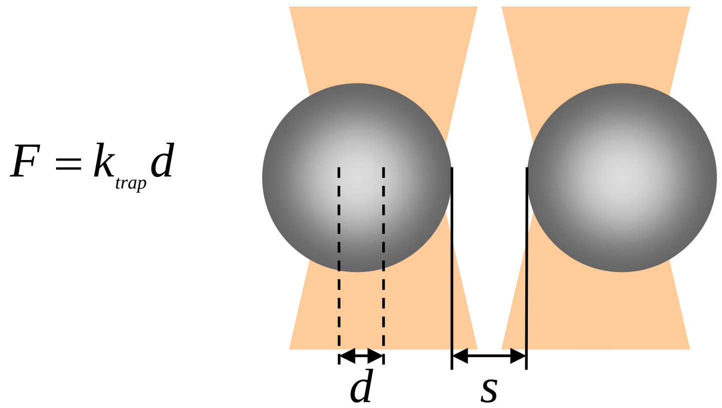


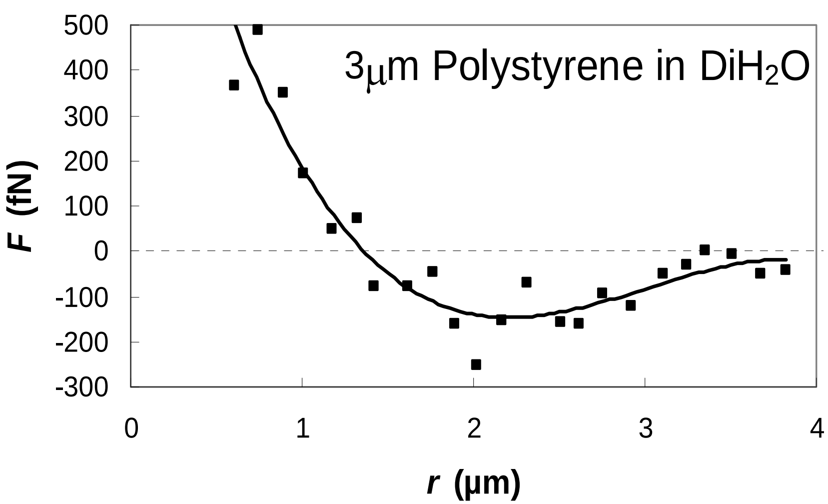
15nm



- **Experimental System:**
  - 1 ± 0.02 µm dia. Polystyrene beads (n = 1.6) w/ surface carboxyl groups
  - 1mM AOT / Hexadecane solution (n = 1.43)
  - Laser Power: ~100mW (max 1W)
- **Imaging and Processing:**
  - Camera: **600 × 800**, 500 fps
  - Objective: 100X, 1.3 N/A Oil
  - Refined intensity peak-finding algorithm (~20 nm resolution)
- **Trap Stiffness: 0.026 pN/nm**
- **Errors:** Spatial: ±20 nm; Force: ± 0.1pN
- Force range can be adjusted by changing laser power and image magnification

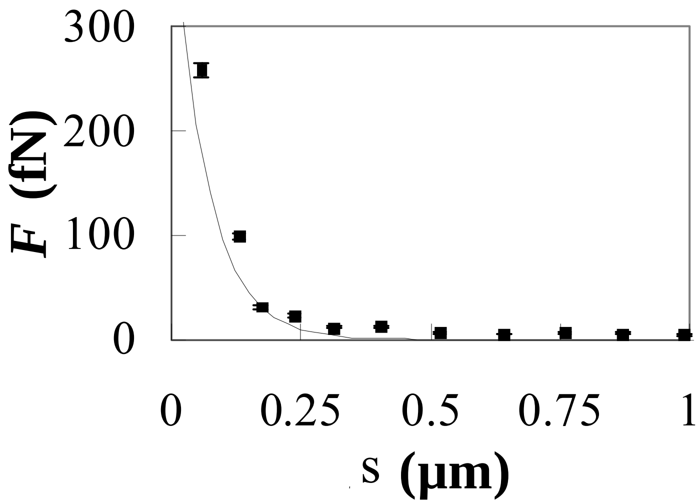
Sandia's Direct Force Capability

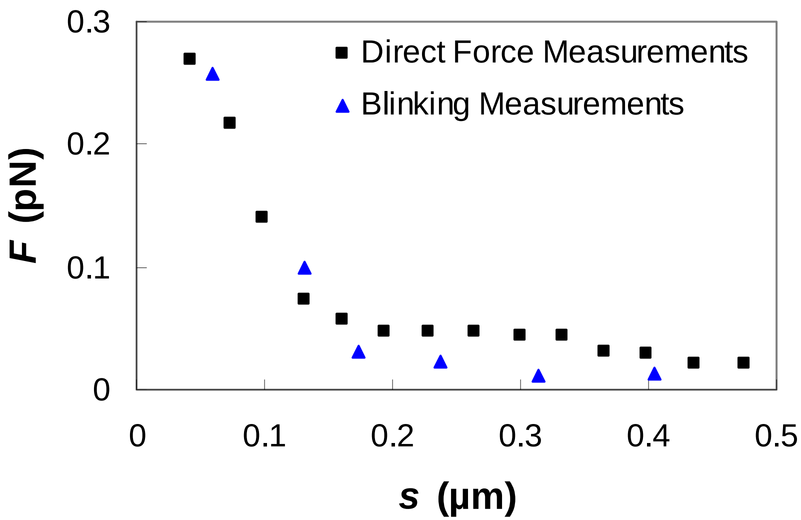




- **Experimental System:**
  - 3 ± 0.02 μm dia. Polystyrene beads (n = 1.6) w/ surface carboxyl groups
  - Aqueous (n = 1.33)
  - Laser Power: ~70mW (max 1W)
- **Imaging and Processing:**
  - Camera: **600 × 800**, 500 fps
  - Objective: 100X, 1.3 N/A Oil
  - Refined intensity peak-finding algorithm (~20 nm resolution)
- **Trap Stiffness: 0.026 pN/nm**
- **Errors:** Spatial: ±10 nm; Force: ± 0.1pN
- Force range can be adjusted by changing laser power and image magnification

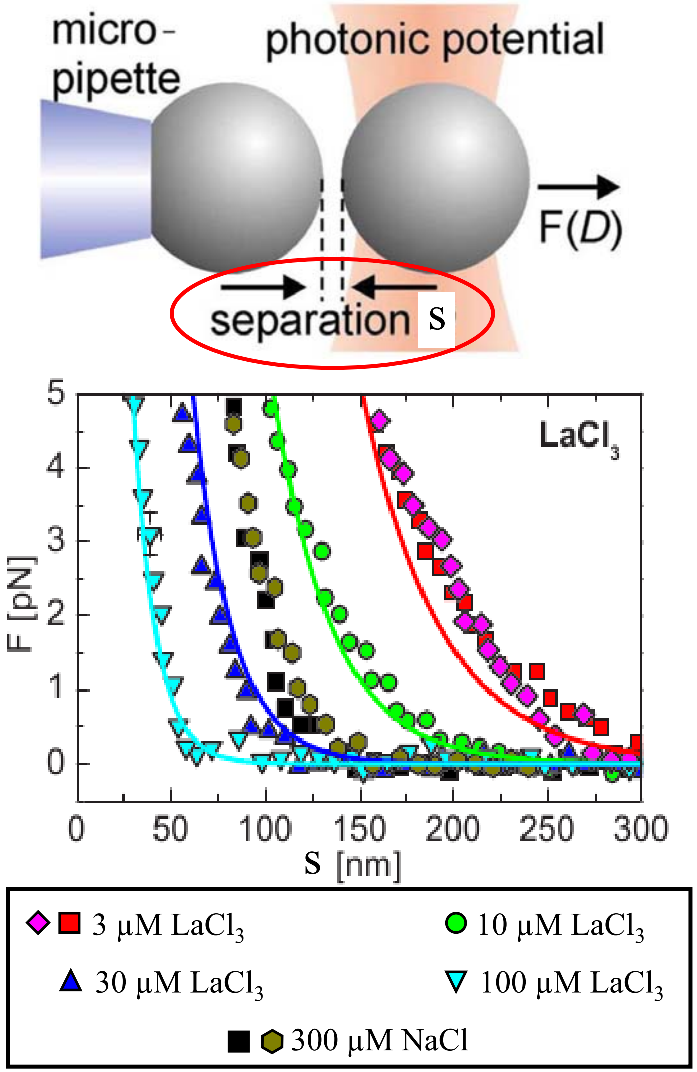
Sandia's Blinking Capability





- **Experimental System:**
  - 2.47 ± 0.02 µm Silica beads (n = 1.37)
  - D.I. H_2O (n = 1.33)
- **Operating Parameters:**
  - Frame Rate: 500 fps
  - Diffusion time: 30 ms
  - 100,000 Images @ each *D* → ~2,000 trajectories
  - Max. Resolution: 800 × 600 pix
  - Objective: 100X, 1.3 N/A Oil
  - Refined intensity peak-finding algorithm (~20 nm resolution)
- **Trap Stiffness:** Measurements are independent of trap stiffness
- **Errors:** Spatial: 20 nm; Force: *O*(1 fN)

Summary of Gutsche et al. (2007)

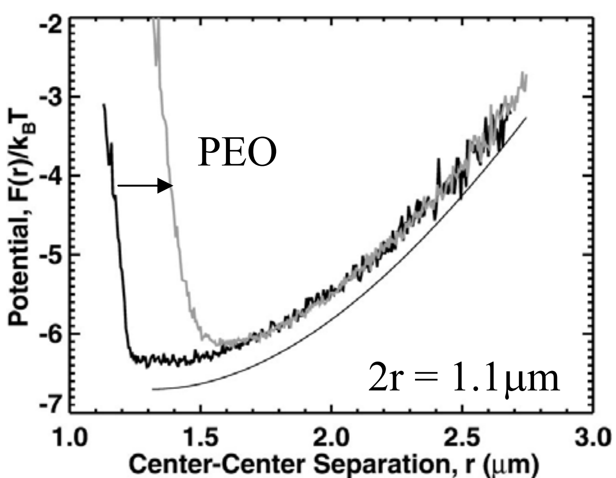


- **Experimental System:**
  - 2.26 ± 0.02 µm dia. Polystyrene (n = 1.6) beads w/ sulfate surface groups
  - Aqueous salt solutions (n = 1.33) of varying molarities of NaCl, $CaCl_2$, $LaCl_3$
- **Imaging and Processing:**
  - Frame Rate: 30 fps
  - Max. Resolution: 1392 × 1040 pix
  - Objective: 100X, 1.3 N/A Oil
  - Edge detection center-finding algorithm (~2 nm resolution)
- **Trap Stiffness:** 0.085 pN/nm
- **Errors:** Spatial: ±10 nm; Force: ±0.3 pN

PDMS system challenges

- Difficulties with PDMS-silica system:
  - Particles are sticky ⇒ flow cell
  - Particles too small ⇒ diameter $\geq 2\lambda$
  - Due to porosity of microparticle silica, particles are ~ same refractive index as PDMS solvent $n_{PDMS} = 1.400$ @589nm
    - $n_{silica} = 1.39\text{-}1.45$ ⇒ borosilicate particles n~1.5
    - Titania shell absorbed/scattered laser

- Silica – PEO – water system would be MUCH easier (Owen et al.)
  - DOI: 10.1103/PhysRevE.64.011401
  - Effect of adsorbed polymer on interactions
  - Line scanning optical trapping



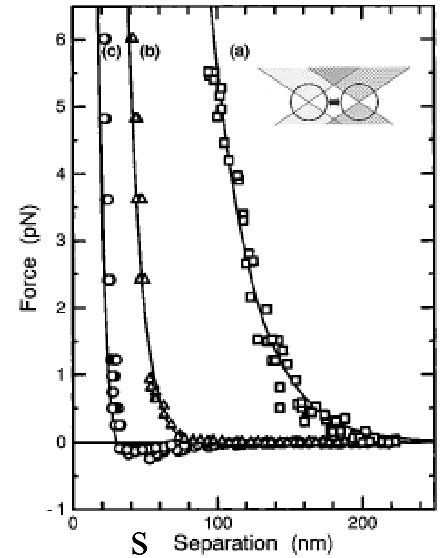
Measurement Summary

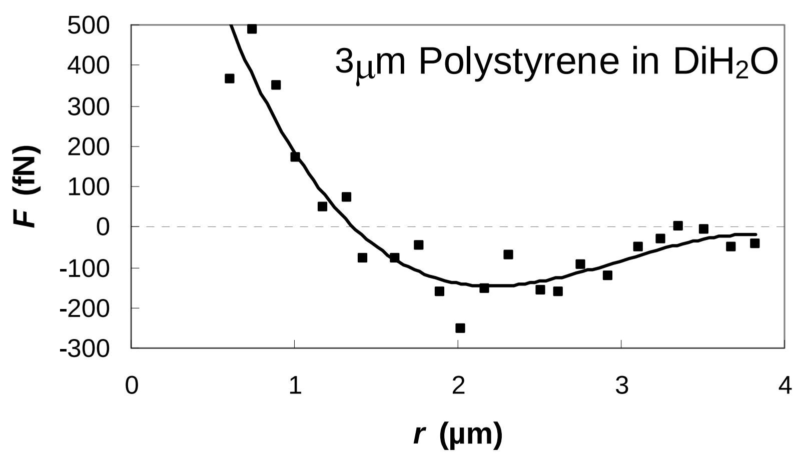
- Operating parameters must be tuned specifically to the experimental system
  - **– Define force/separation range of interest**
- **Measurement uncertainties** result from calibration of trap stiffness, image resolution, etc.
- **Need large particles:** diameter $\geq 2\lambda$ so traps don't overlap at contact
- **Particle Separation, *s*:**
  - Higher laser power attains smaller *s*, less out of plane motion
  - Measurement of *s* depends on spatial resolution of images, center-finding, and exposure time

- Blinking Force Measurement
  - small forces (<kT)
  - independent of lasers or induced dipole
- Direct Force Measurement
  - larger and attractive forces
  - calibration required

Sugimoto, *et al.*, *Langmuir* (1997)

- 2.13 ± 0.06 μm Silica particles in varying ionic strength aqueous solutions



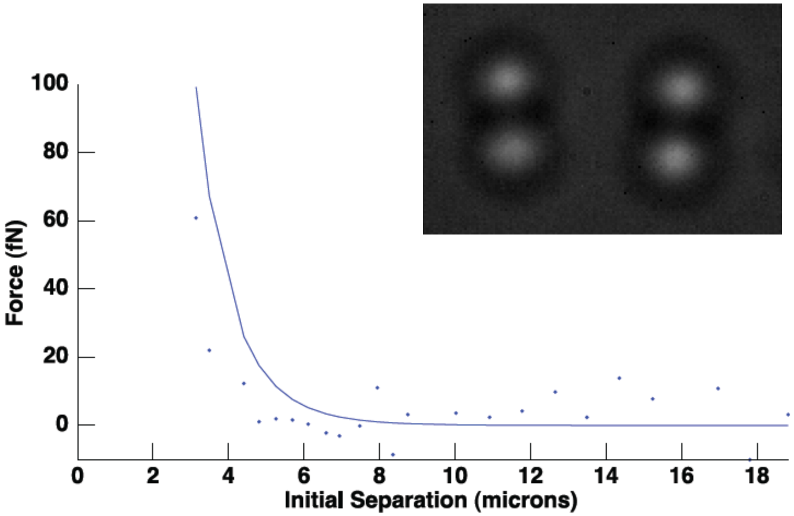


Experimental Impact

- Anisotropic Particle Interactions
  - Can measure the diffusion tensor of dumbell particles $\tilde{D}\vec{F} = k_B T \vec{v}.$
  - Assuming pairwise-additivity using the Debye-Huckel form overestimates the actual repulsion between the spheres by about a factor of two

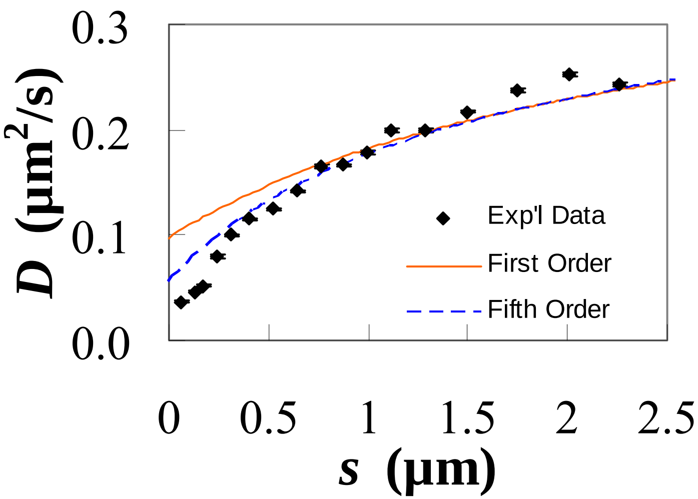
$$F(r) = k_B T \tilde{\zeta}^2 \frac{a^2}{\lambda_B} \frac{e^{-\kappa(r-2a)}}{r} \left(\frac{1}{r} + \kappa \right)$$

E. Dufresne (Yale Univ.)



2.47 µm Silica in DI H_2O

- Multiparticle Effects
  - Pair-wise additivity likewise can not predict multiparticle effects

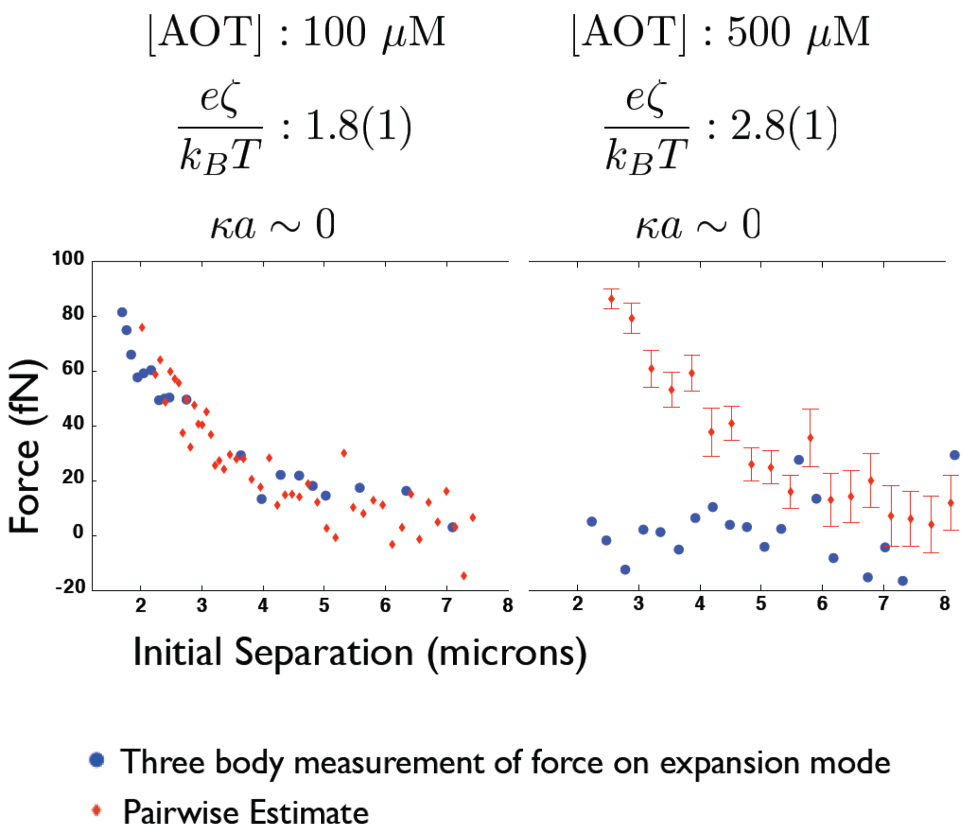


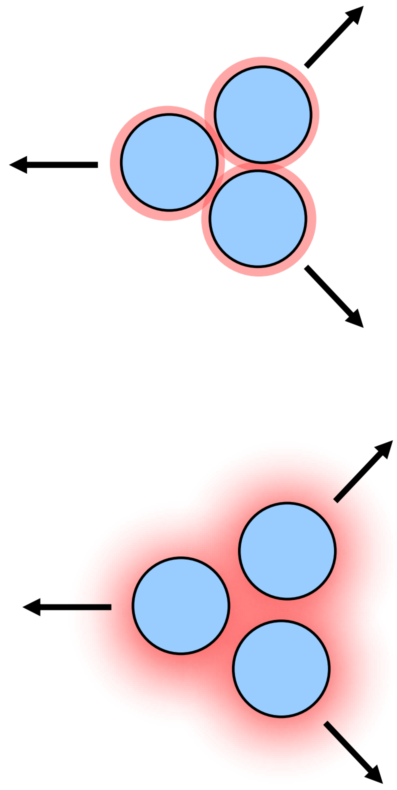
- First order hindered diffusion model breaks down at close separations

$$D = 2D_o \left(1 - \frac{3a_h}{2r} + \left(\frac{a_h}{r} \right)^3 - \frac{15}{4} \left(\frac{a_h}{r} \right)^4 + O\left(\frac{a_h}{r} \right)^6 \right)$$

Multiparticle effects

- Breakdown of DLVO theory is especially important in nanoparticle systems.

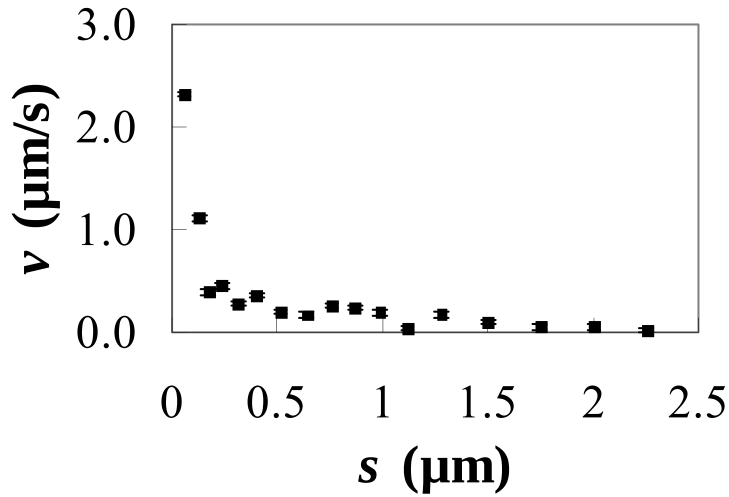


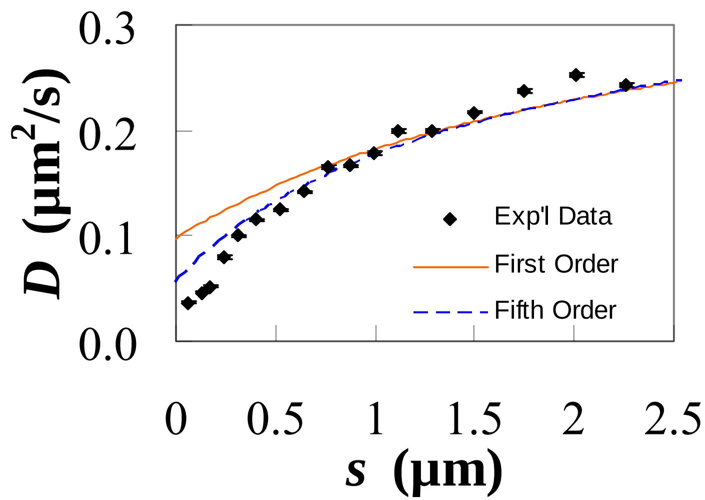


E. Dufresne (Yale Univ.)

Velocity and Diffusivity

2.47 μm Silica in de-ionized H_2O





- Diffusivity: $D = 2D_o\left(1 - \frac{3a_h}{2r}\right)$
- Force:

$$F = k_b T\left(\frac{e\zeta}{k_b T}\right)^2 \frac{a^2}{\lambda_B}\frac{e^{-\kappa(r-2a)}}{r}\left[\frac{1}{r} + \kappa\right]$$

- Fit for Debye screening length, κ, and surface charge, $\left|e\zeta / k_b T\right|$