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Nanoparticle Knudsen Layers in Gases

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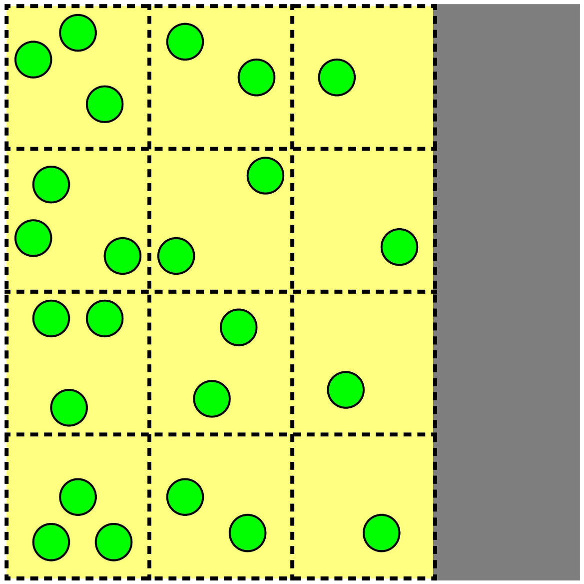
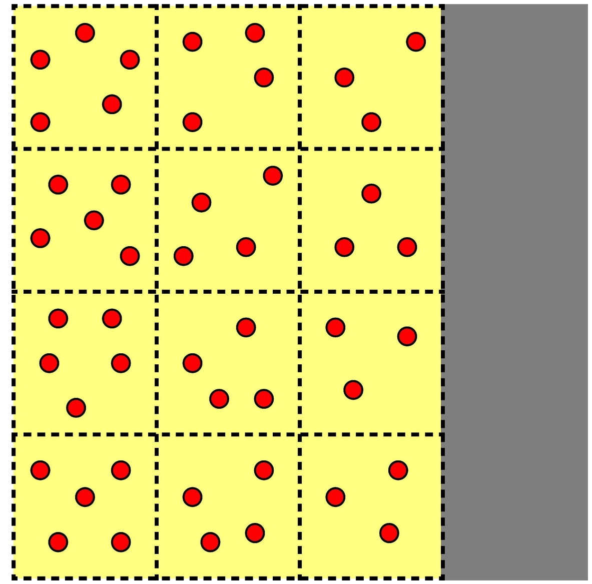
Nanoparticle Knudsen Layer in Gas



Large particles diffusing to wall approach zero concentration



Small particles diffusing to wall approach nonzero concentration

Nanoparticles in gas have a Knudsen layer at wall

- **Concentration doesn't vanish as for larger particles**
- **Concentration at wall proportional to particle flux**

Investigate using theory and simulations

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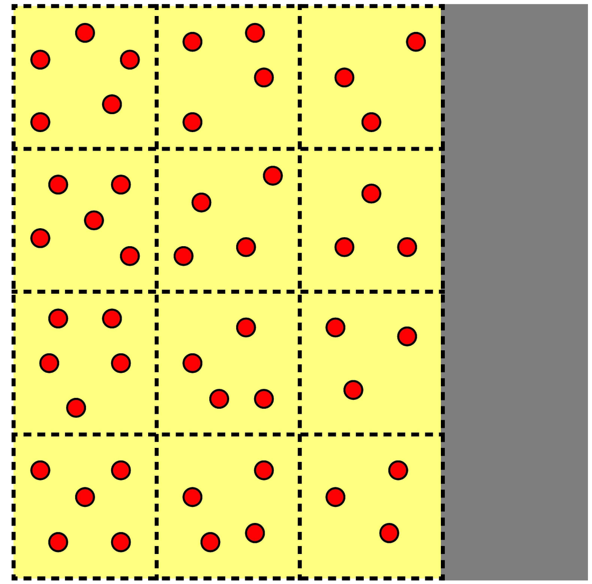
Particle-Flux Boundary Condition

$$\hat{\mathbf{n}} \cdot \left(n\mathbf{U} - D \frac{\partial n}{\partial \mathbf{x}} \right) = \frac{ncf}{\pi^{1/2}}$$

Drift + Diffusion ∝ Jump



Particle-flux boundary condition as wall is approached

- **Can be used in advection-diffusion transport analyses**
  - **Analogous to velocity-slip and temperature-jump BCs**
- **Concentration *n* extrapolated to wall proportional to flux**
  - **Drift velocity *U*, diffusivity *D*, thermal velocity *c***
- **Particle-flux coefficient *f* is dimensionless, order-unity**
  - **Depends on how particles stick to & reflect from wall**

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Approximate Theory

$$\frac{\partial N}{\partial t}+\frac{\partial}{\partial \mathbf{x}}\cdot(\mathbf{u}N)+\frac{1}{\tau}\frac{\partial}{\partial \mathbf{u}}\cdot(-\mathbf{v}N)=\frac{c^2}{2\tau}\frac{\partial}{\partial \mathbf{u}}\cdot\left(\frac{\partial N}{\partial \mathbf{u}}\right),\ \mathbf{v}=\mathbf{u}-\mathbf{U}$$

GFP equation

$$N=\frac{1}{\pi^{3/2}c^3}\exp\left[-\frac{\mathbf{v}\cdot\mathbf{v}}{c^2}\right]\left(n_\infty+n_1\exp\left[\frac{2\mathbf{U}\cdot(\mathbf{x}-\tau\mathbf{v})}{c^2\tau}-\frac{\mathbf{U}\cdot\mathbf{U}}{c^2}\right]\right)$$

exact solution

$$\int_{-\hat{\mathbf{n}}\cdot\mathbf{u}>0} N(-\hat{\mathbf{n}}\cdot\mathbf{u})\,d\mathbf{u}=\int_{\hat{\mathbf{n}}\cdot\mathbf{u}>0} R[\mathbf{u}]N(\hat{\mathbf{n}}\cdot\mathbf{u})\,d\mathbf{u}$$

approximation at wall

$$f=s\left\{2-s\left(1+\operatorname{erf}\left[\hat{U}\right]-\frac{1-\exp\left[-\hat{U}^2\right]}{\pi^{1/2}\hat{U}}\right)\right\}^{-1}$$

sticking-fraction process: s

$$f=\frac{\pi^{1/2}\hat{U}\left(1-\exp\left[-\hat{U}_n^2\right]\right)}{1-\exp\left[-\hat{U}_n^2\right]-\exp\left[-\hat{U}^2\right]+\exp\left[-\left(\hat{U}_n-\hat{U}\right)^2\right]+\pi^{1/2}\hat{U}\left(\operatorname{erfc}\left[\hat{U}_n-\hat{U}\right]+\operatorname{erfc}\left[\hat{U}\right]\right)}$$

cutoff-velocity process: $\sigma=1-\exp\left[-\hat{U}_n^2\right],\ \hat{U}_n=U_n/c,\ \hat{U}=U/c$

- **Generalized Fokker-Planck equation yields approximate particle-flux coefficient *f* for arbitrary wall interactions**
  - **Use exact solution for steady uniform unbounded space**
  - **Approximation: outgoing equals reflected incoming at wall**
- **Apply to two reflection processes *R* with drift velocity *U***
  - **Sticking fraction: sticking probability *s* same for all particles**
  - **Cutoff velocity: stick if normal velocity < U_n (σ is convenient)**
  - **Any reflections are taken to be specular**

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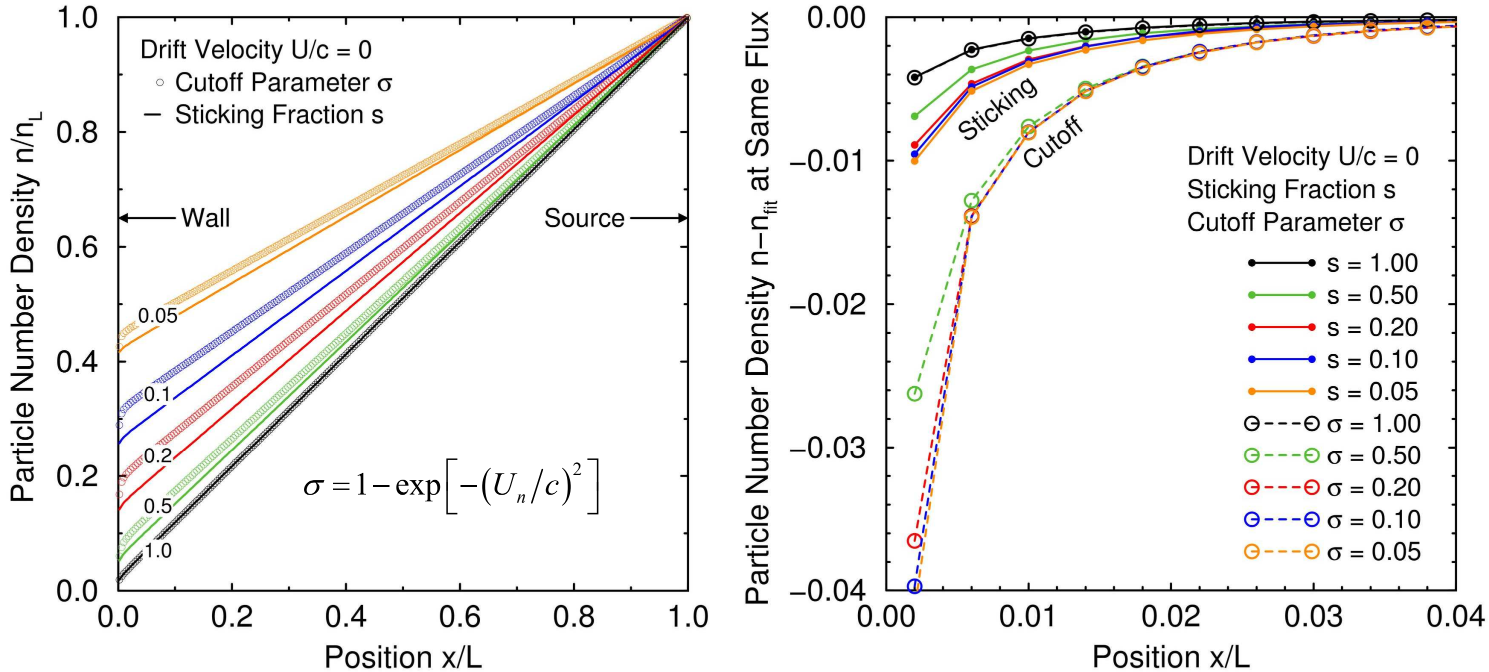
Langevin Simulations



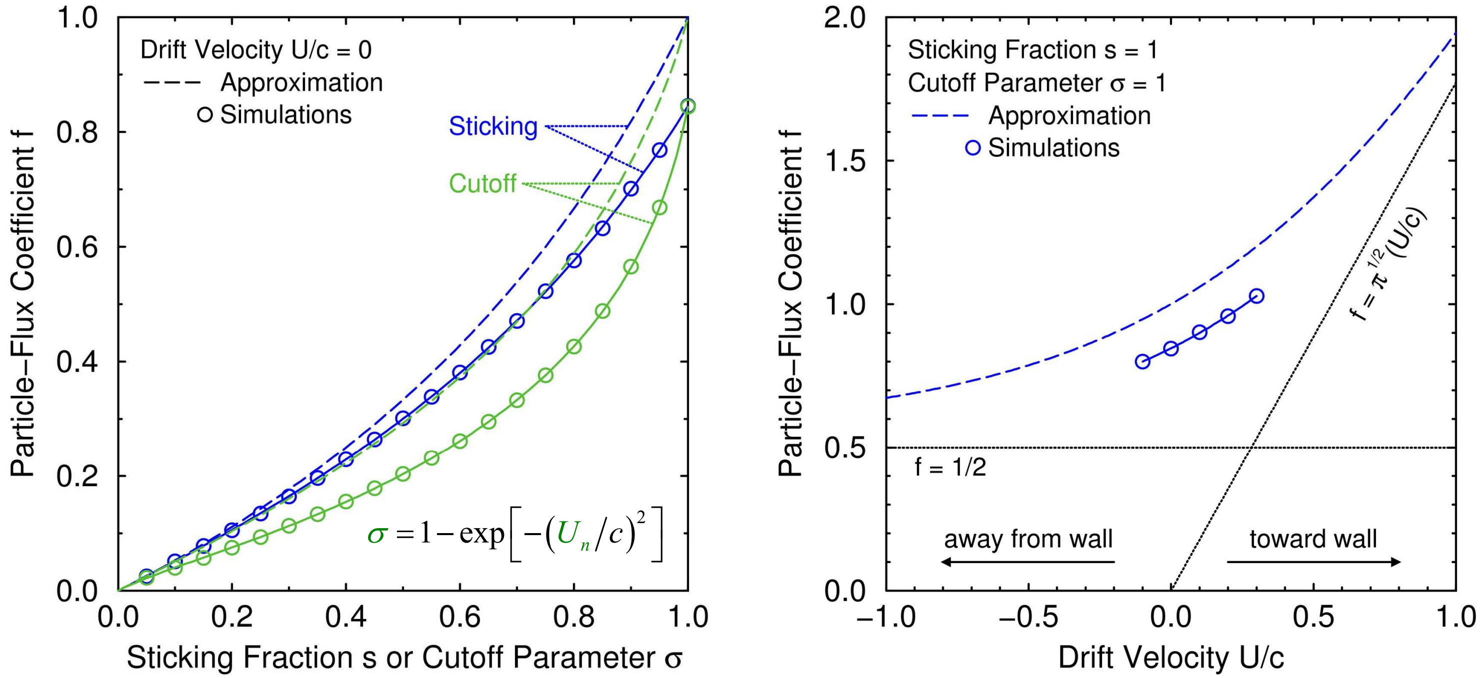
Langevin particle simulations: MP Ermak-Buckholz

- **1D diffusion: wall at x = 0, source at $x = L$ = 1000 nm**
- **20-nm PSL in air, 18-nm stopping distance (NIST, 2005)**

Knudsen layer near wall, linear profile away from wall

- **Particle-flux coefficient *f* by extrapolating linear part to wall**
- **Cutoff σ (U_n) has larger effect than sticking *s***

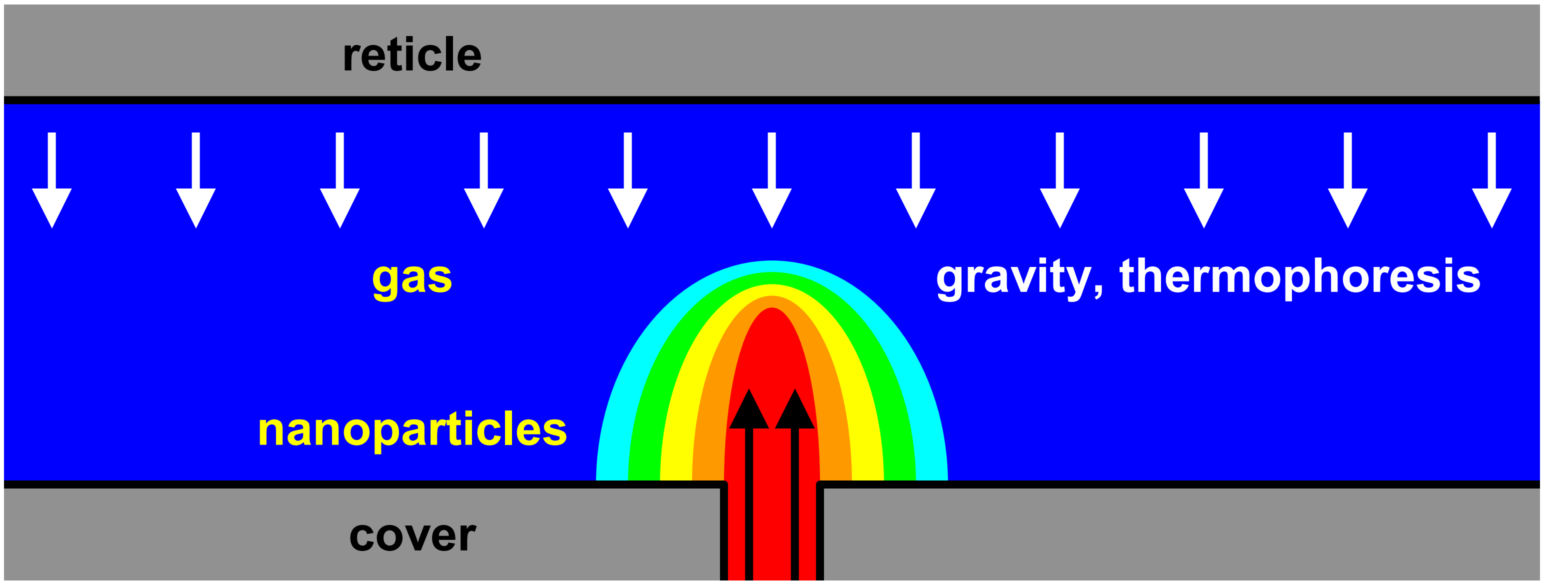
Particle-Flux Coefficient



Particle-flux coefficient *f* from approximation & simulations

- **Left: vs. sticking *s* or cutoff σ with drift velocity *U* = 0**
- **Right: vs. drift velocity *U* with *s* = σ = 1**
- **Approximation and simulations agree reasonably well**
  - **Better for sticking than cutoff: reflected particle distribution**

Semiconductor Application



- Assess contamination of reticle by nanoparticles in gas
  - Nanoparticles injected upward from below
  - Nanoparticles repelled by gravity and thermophoresis
- Find probability that injected nanoparticle sticks to reticle
  - Advection-diffusion particle-transport analysis
    - Particle-flux boundary condition on solid surfaces
  - Massively parallel Langevin particle simulations

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Advection-Diffusion Analysis

$$\frac{\partial n}{\partial t}+\frac{\partial}{\partial \mathbf{x}}\cdot\left(n\mathbf{U}-D\frac{\partial n}{\partial \mathbf{x}}\right)=0$$

advection-diffusion equation

$$\hat{\mathbf{n}}\cdot\left(n\mathbf{U}-D\frac{\partial n}{\partial \mathbf{x}}\right)=\frac{ncf}{\pi^{1/2}}$$

particle-flux boundary condition

$$H=U_I\tau,\ \hat{V}=\pi^{1/2}U/c,\ D=c^2\tau/2$$

$$e_H=\exp\left[HU/D\right],\ e_L=\exp\left[LU/D\right]$$

$$P=\frac{f_2\left(\left(e_H-1\right)f_1+\hat{V}\right)}{\left(e_L-1\right)f_1f_2+\left(e_Lf_1+f_2\right)\hat{V}}$$

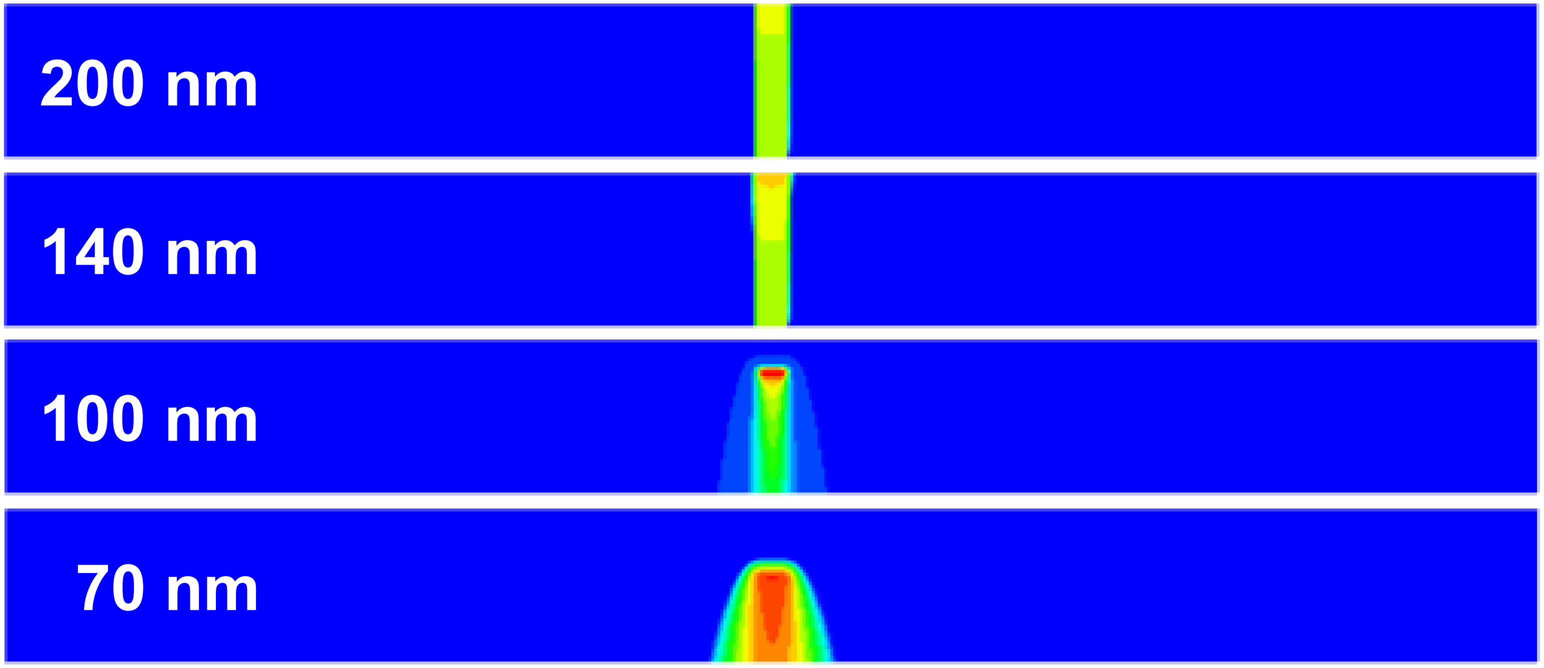
deposition probability

Advection-diffusion analysis for deposition probability *P*

- **Particles injected at speed U_I "stop" at height *H* in gap *L***
- **Particles drift downward at speed *U* and diffuse with *D***
- **Particles stick with probabilities s_1 & s_2 on bottom & top**
- **Particle-flux coefficients f_1 & f_2 on bottom & top**
- **If $H > L$, reflect $(1–s_2)$, so $H \rightarrow 2L–H$, $P \rightarrow s_2+(1–s_2)P$, etc.**

Above analysis does not provide deposition profile

Typical Langevin Simulations



Particle concentration fields for continuous injection

- **Water-like particles in nitrogen at 50 mtorr and 295 K**
- **Gap height 1 cm, upward injection velocity 10 m/s**
- **Gravity 0*g*, temperature gradient 10 K/cm (thermophoresis)**
- **Sticking fraction *s* = 1 on bottom and top**

Diameters 70-200 nm, deposition probabilities 0-1

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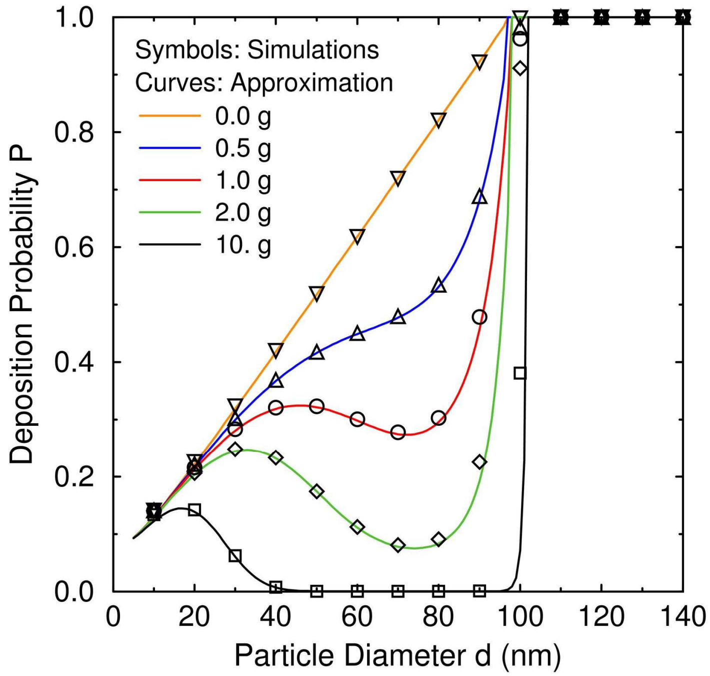
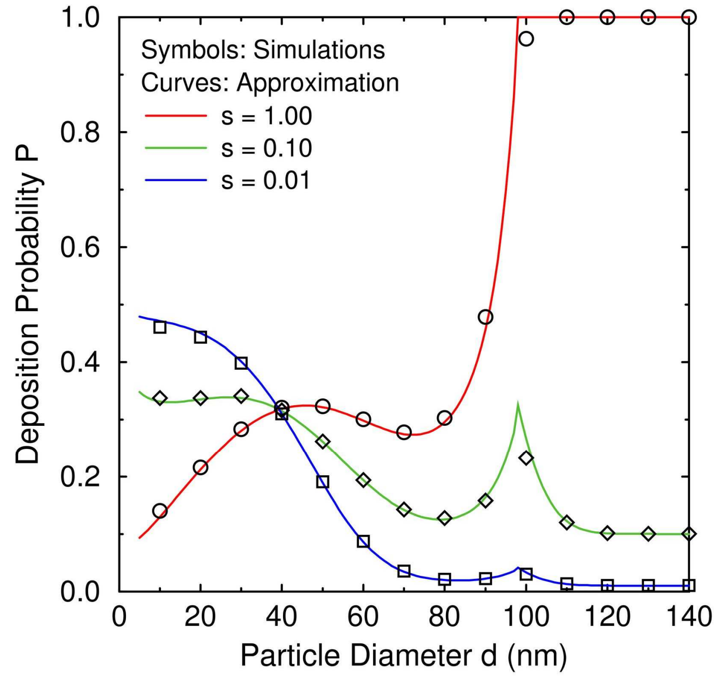
Advection-Diffusion and Langevin





Advection-diffusion and Langevin agree very well

- **Left: gravity 0-10*g* with *s* = 1 (thermophoresis is similar)**
  - **“Bumps” at low diameter, where diffusion exceeds gravity**
- **Right: sticking fraction *s* = 0.01, 0.1, 1 with gravity 1*g***
  - **“Cusps” occur when penetration exceeds gap: $H \geq L$**
- **In both cases, large diameter *d* yields $P \rightarrow s$**

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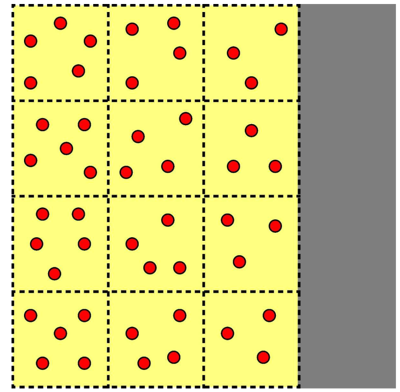
Conclusions

$$\hat{\mathbf{n}} \cdot \left(n\mathbf{U} - D\frac{\partial n}{\partial \mathbf{x}} \right) = \frac{ncf}{\pi^{1/2}}$$

Drift + Diffusion ∝ Jump



- Nanoparticles in gas have Knudsen layer at wall
  - Particle concentration at wall proportional to flux
  - Structure determined by reflection/sticking process
- Nanoparticle Knudsen layer investigated
  - Generalized Fokker-Planck equation
  - MP Langevin particle simulations
- Particle-flux boundary condition developed
  - For advection-diffusion particle-transport analyses
  - Uses particle-flux coefficient ***f*** from above

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