Theoretical derivation of particle collision kernels from a first-time passage approach in the diffusive regime

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Full story:

Motivation

- Is it possible to obtain collision kernels from **first-time passage** approach theoretically?
- Many authors neglect the **transient term** in the Smoluchowski diffusive collision kernel. What is its interpretation and consequences?
- How to theoretically adapt collision kernels for **high concentrations** ($\gg$ 1k ppm)?

Previous works

β: Particle-particle collision kernel ($\mathrm{m^3\,s^{-1}}$)

Smoluchowski approach[1,2],

$$\left.\begin{aligned} F &= 4\pi(R_{p,i}+R_{p,j})^2 J_{r=R_{p,i}+R_{p,j}} \\ J_{r=R_{p,i}+R_{p,j}} &= D(dn/dr)_{r=R_{p,i}+R_{p,j}} \end{aligned}\right\} \Rightarrow \beta = \frac{F}{n_0} = 4\pi(R_{p,i}+R_{p,j})D\left(1+\underbrace{\frac{R_{p,i}+R_{p,j}}{\sqrt{\pi D t}}}_{0}\right)$$

Wiener sausage approach[3],

$$V_{\mathrm{diff}} = \frac{4\pi}{3}(R_{p,i}+R_{p,j})^3 \times\left[1+\frac{6}{\sqrt{\pi}}\frac{\sqrt{(D_i+D_j)t}}{(R_{p,i}+R_{p,j})}+3\frac{(D_i+D_j)t}{(R_{p,i}+R_{p,j})^2}\right]$$

$$\Rightarrow \beta = \frac{dV_{\mathrm{diff}}}{dt} = 4\pi D(R_{p,i}+R_{p,j})\left[\underbrace{\frac{(R_{p,i}+R_{p,j})}{\sqrt{\pi D t}}}_{0}+1\right]$$

New theoretical approach

$$\frac{\partial P(\vec{x}_1,t)}{\partial t} = D_1\vec{\nabla}_1^2 P(\vec{x}_1,t), \quad \frac{\partial P(\vec{x}_2,t)}{\partial t} = D_2\vec{\nabla}_2^2 P(\vec{x}_2,t)$$

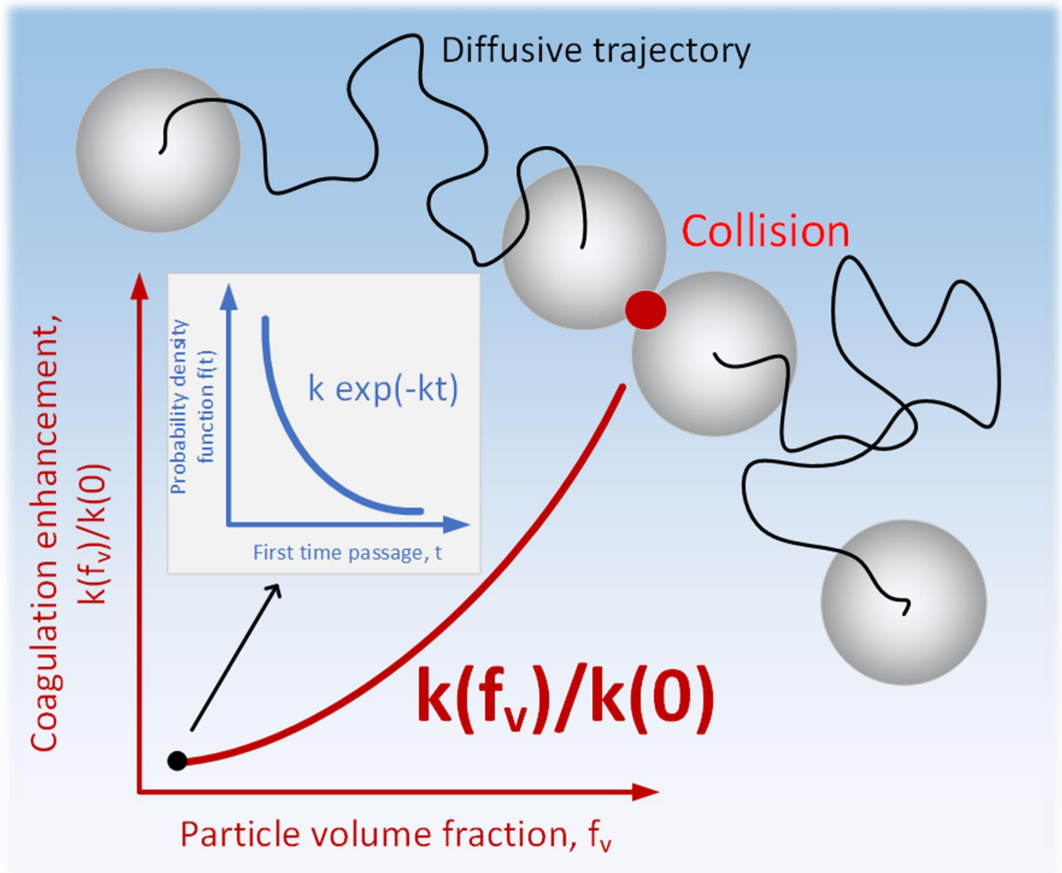
$$\Rightarrow \frac{\partial P(\vec{x}_1,\vec{x}_1-\vec{r},t)}{\partial t} = (D_1\vec{\nabla}_1^2+D_2\vec{\nabla}_2^2)P(\vec{x}_1,\vec{x}_1-\vec{r},t) \approx D\vec{\nabla}_r^2 P(\vec{x}_1,\vec{x}_1-\vec{r},t)$$

$$\Rightarrow Q(\vec{r},t) = \frac{1}{V}\int P(\vec{x}_1,\vec{x}_1-\vec{r},t)\, d\vec{x}_1 \Rightarrow$$

$$S_1(t|r_0) = \int_R^\infty Q(r,t|r_0,t_0)4\pi r^2\, dr \Rightarrow S(t) = \langle S_N\rangle = \lim_{N,V\to\infty}\frac{1}{V^N}\int\prod_{i=1}^N S_1(t|r_{i0})\, 4\pi r_i^2 dr_i \Rightarrow S(t) = \exp(-4\pi RDnt) = \exp(-k_d t) \Rightarrow$$

$$\boxed{f(t) = \frac{dC}{dt} = -\frac{dS}{dt} = k_d\exp(-k_d t)} \Rightarrow \mathrm{E}[t] = \int t' f(t')\, dt' = \frac{1}{k_d} \Rightarrow$$

$$\boxed{k = \frac{1}{\mathrm{E}[t]} = k_d\left(1-\frac{k_d R}{\sqrt{Dk_d}}\exp\left[\frac{R^2 k_d}{\pi D}\right]\mathrm{erfc}\left[R\sqrt{\frac{k_d}{\pi D}}\right]\right)^{-1} \approx k_d\left(1+\sqrt{24 f_v}+24 f_v\left(1-\frac{2}{\pi}\right)\right)}$$



Numerical simulations

Langevin dynamics simulations,

$$m\frac{d\vec{v}}{dt} = -f_t\vec{v} + \vec{F}_B + \underbrace{\vec{F}_e}_{0}$$

Brownian force:

$$\langle F_{\mathrm{B}}(t)\rangle = 0,$$

$$\langle F_{\mathrm{B}}(t)F_{\mathrm{B}}(t')\rangle = 6fk_{\mathrm{B}}T\delta(t-t'),$$

Ermak and Buckholz[4],

$$\vec{v}(t+\Delta t) = \vec{v}(t)e^{-\alpha\Delta t} + \frac{\vec{F}_e}{m\alpha}(1-e^{-\alpha\Delta t}) + \vec{B}_1$$

$$\vec{r}(t+\Delta t) = \vec{r}(t) + \frac{1}{\alpha}\left(\vec{v}(t+\Delta t)+\vec{v}(t)-\frac{2\vec{F}_e}{m\alpha}\right)\frac{1-e^{-\alpha\Delta t}}{1+e^{-\alpha\Delta t}} + \frac{\vec{F}_e\Delta t}{m\alpha} + \vec{B}_2$$

$$\langle B_1\rangle = \langle B_2\rangle = 0,$$

$$\langle B_1^2\rangle = \frac{3k_{\mathrm{B}}T}{m}(1-e^{-2\alpha\Delta t}),$$

$$\langle B_2^2\rangle = \frac{6k_BT}{m\alpha^2}\left(\alpha\Delta t-2\frac{1-e^{-\alpha\Delta t}}{1+e^{-\alpha\Delta t}}\right).$$

$$\Delta t = \tau_{ij}\sqrt{B(B+3\pi/4)}$$

$$B = l_p^2/(6D_{ij}\tau_{ij}), \quad \tau_{ij} = m_{ij}/f_{ij}.$$

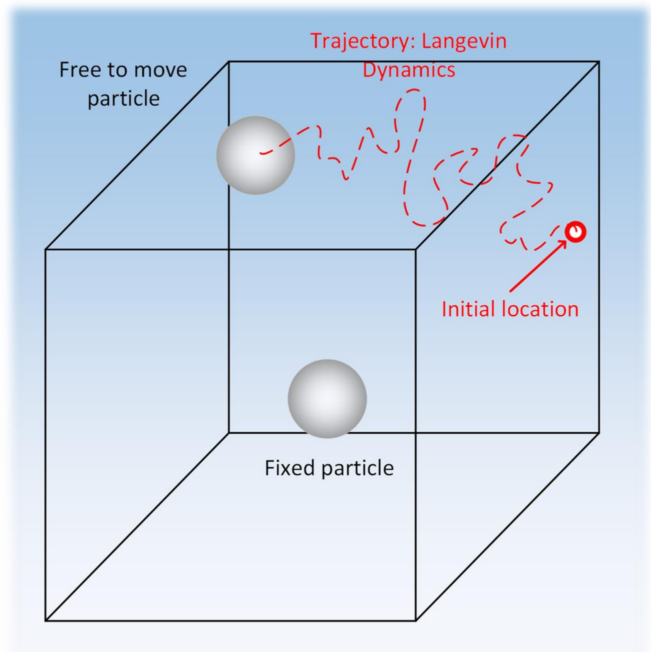
$$l_p = 0.2R_p$$

Collision kernel calculation[5],

$$\beta = \frac{L^3}{\mathrm{E}[t]}$$

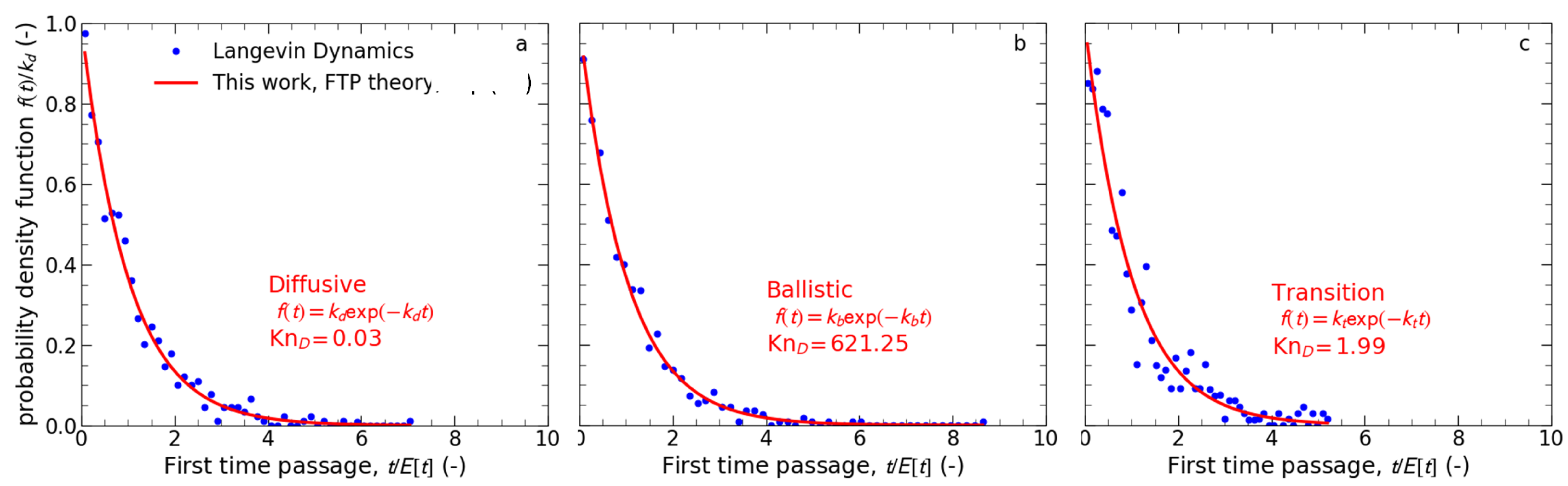
L: Cubic domain length

E[t]: Average first-time passage (time until the first collision)

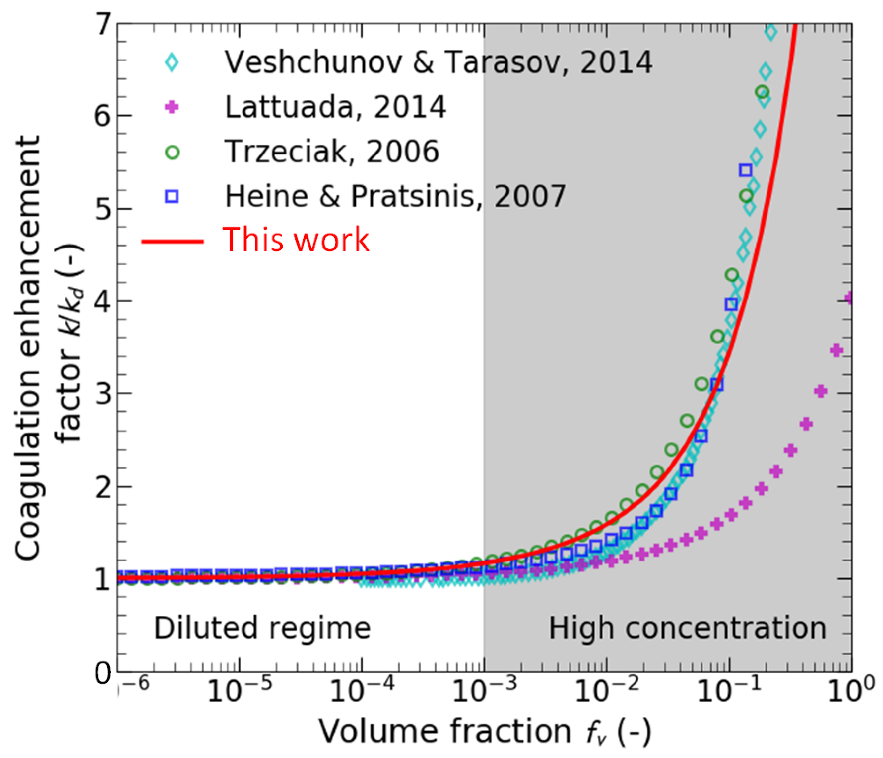


Results

Simulated first-time passage distributions compared to theory



High concentration coagulation enhancement



Conclusions

- The **first-time passage** function of Langevin dynamics simulation follow an **exponential distribution** as predicted by theory (only theoretically probed in the diffusive regime).
- The commonly neglected **transient term** of the diffusive Smoluchowski collision kernel results in a **high-concentration coagulation enhancement**.

References

[1]Friedlander, S. K., & Smoke, D. (2000). Haze: Fundamentals of aerosol dynamics. *Oxford University Press.*

[2]Seinfeld, J. H., & Pandis, S. N. (2016). Atmospheric chemistry and physics. *John Wiley & Sons.*

[3]Berezhkovskii, A., Y. A. Makhnovskii, and R. Suris. 1989. *J. Stat. Phys.* 57 (1–2): 333–46.

[4]Ermak, D. L., and H. Buckholz. (1980). *Comput. Phys.* 35 (2):169–82.

[5]Gopalakrishnan, R., and C. J. Hogan. Jr. (2011). *Aerosol Sci. Technol.* 45 (12):1499–509.

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