

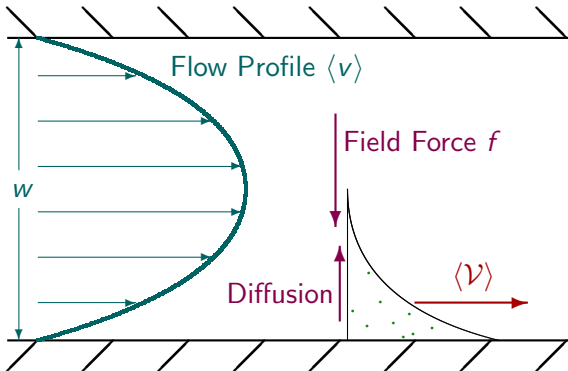
Ideal Retention Theory for μ FFF

Tyler Shendruk
Gary Slater

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Normal Mode



$$\lambda \equiv \frac{k_B T}{fw}$$

Figure: Schematic of normal-mode FFF.

Normal Mode

Ideal Retention Ratio

When we have point particles in a channel of height w feeling a force f then we can define a retention parameter

$$\lambda = k_B T / fw \neq \langle \tilde{y} \rangle$$

$$\begin{aligned} R &= \frac{t_0}{t_R} = \frac{\langle \mathcal{V} \rangle}{\langle v \rangle} = \frac{\langle cv \rangle}{\langle c \rangle \langle v \rangle} \\ &= 6\lambda \left[\coth \left(\frac{1}{2\lambda} \right) - 2\lambda \right] = 6\lambda \mathcal{L} \left(\frac{1}{2\lambda} \right) \end{aligned}$$

i.e. constant for given force.

Steric Mode

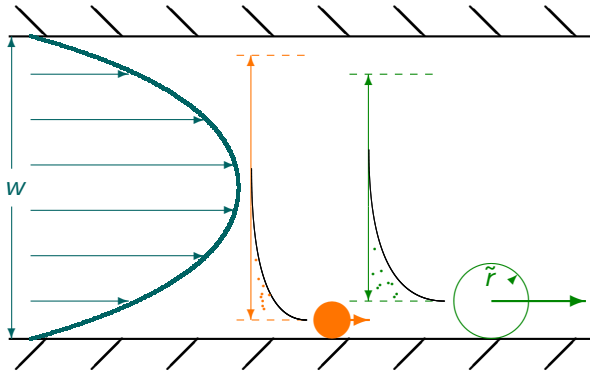


Figure: Schematic of steric-mode FFF.

Steric Mode

Retention Ratio

The new boundaries on the integrals give

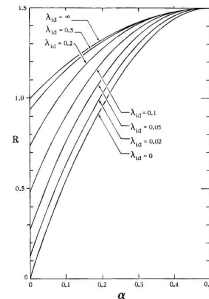
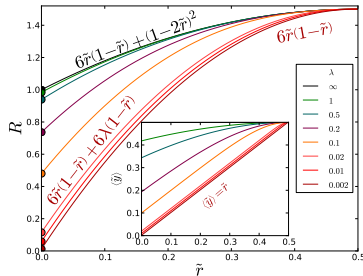
$$R = \frac{\langle cv \rangle}{\langle c \rangle \langle v \rangle}$$

$$= 6\lambda (1 - 2\tilde{r}) \mathcal{L} \left(\frac{1 - 2\tilde{r}}{2\lambda} \right) + \mathcal{F}_p$$

where $\mathcal{F}_p(r, w) = 6\tilde{r}(1 - \tilde{r})$.

Steric Mode

Even if the force is held constant for all particles sizes there is still separation because steric effects move larger particles closer to the centre than smaller particles.



Scaling

Before

Had variation of R as function of $\tilde{r}$ for fixed λ i.e. fixed force f .

Closer to Reality

Force will change with particle size. For instance, in sedimentation the weight goes as the volume of the particle or $\lambda \propto r^{-3}$. To be general, let force scale as α such that the retention parameter is

$$\lambda = \frac{k_B T}{\mu w} \tilde{r}^{-\alpha} = \Lambda \tilde{r}^{-\alpha}$$

where μ is the “strength”/coefficient of the force.

Scaling

FFF-Method	f	α	Λ
Sedimentation	$\frac{4\pi}{3} g \Delta \rho r^3$	3	$\frac{3}{4\pi} \frac{k_B T}{g \Delta \rho w^4}$
Flow	$6\pi \eta \frac{w \dot{V}_c}{V_0} r$	1	$\frac{k_B T}{6\pi \eta w^3} \frac{V_0}{\dot{V}_c}$
Thermal	$6\pi \eta \frac{D_T \Delta T}{w} r$	1	$\frac{k_B T}{6\pi w \eta D_T \Delta T}$
Magnetic	$\frac{\Delta \mu H}{6} \frac{dH}{dy} r^3$	3	$\frac{6 k_B T}{w^4 \Delta \mu H} \left(\frac{dH}{dy} \right)^{-1}$
Electrical	$6\pi \eta \mu_e E r$	1	$\frac{6\pi \eta}{\mu_e E w}$

Retention Ratio

Retention Ratio

$$R(\tilde{r}, \Lambda) = \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \mathcal{L} \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) + \mathcal{F}_p(\tilde{r})$$

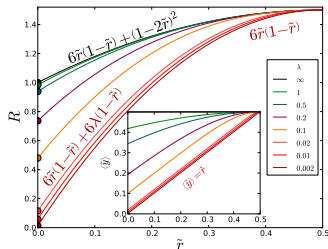


Figure: Fixed λ (equivalent to $\alpha = 0$).

Retention Ratio

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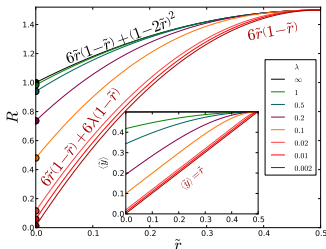


Figure: Fixed λ (equivalent to $\alpha = 0$).

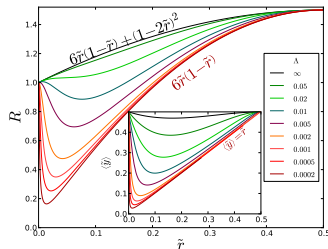


Figure: $\alpha = 1$.

Nonmonotonic

Moral of the Story:

Non-monotonic is bad

- One elution time,
two particle sizes.

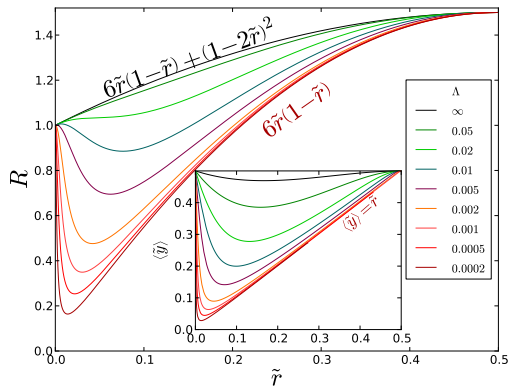


Figure: $\Lambda_c = 1.90 \times 10^{-2}$ for $\alpha = 1$.

Elution Order

Statements:

Recall $R = \langle \mathcal{V} \rangle / \langle v \rangle = t_0/t_R$

- In normal-mode, smaller particles elute before larger particles.

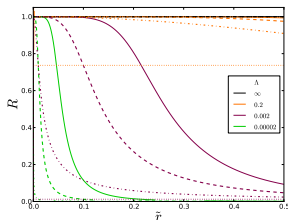


Figure: Normal Mode
 $\alpha = 1, 2, 3$

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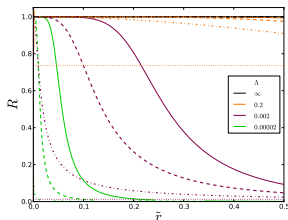


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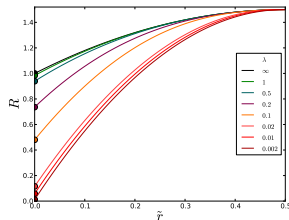


Figure: Steric Mode, fixed λ

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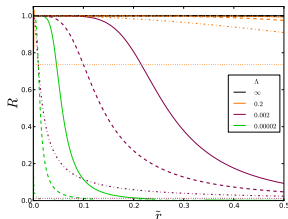


Figure: Normal Mode
 $\alpha = 1, 2, 3$

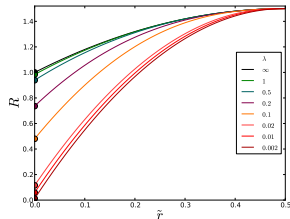


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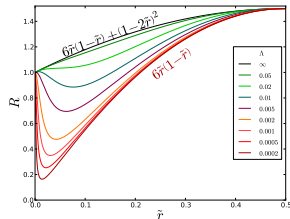


Figure: Steric Mode, $\alpha = 1$

Normal-Steric Transition

Order

Normal Mode when $R' < 0$ smaller particles elute before larger.

Steric Mode when $R' > 0$ larger particles elute earlier than smaller.

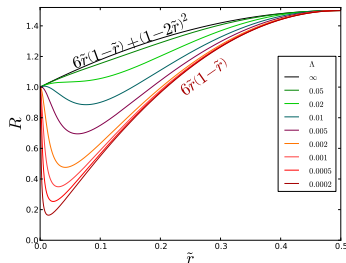


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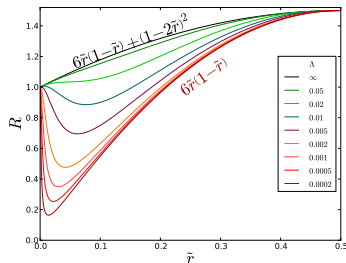


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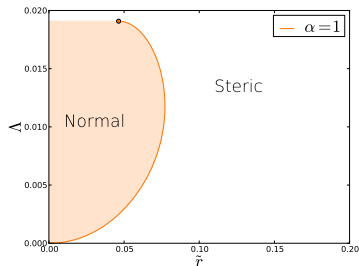


Figure: Trace the minimum of the retention ratio.

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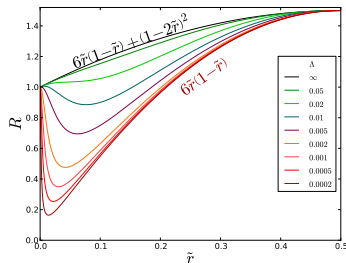


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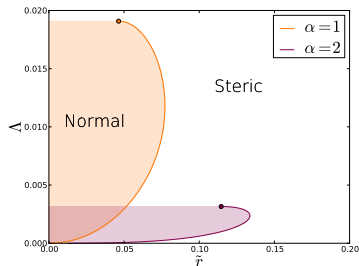


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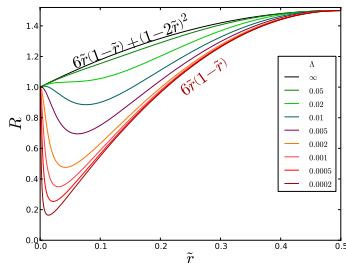


Figure: $\alpha = 1$

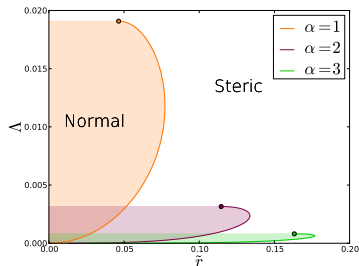


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Hydrodynamic Chromatography-mode

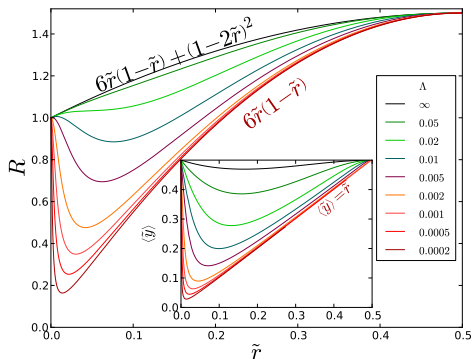


Figure: For $\alpha = 1$ and mid-range Λ , R shows a steric-mode-like elution order at tiny $\tilde{r}$.

Hydrodynamic Chromatography-mode

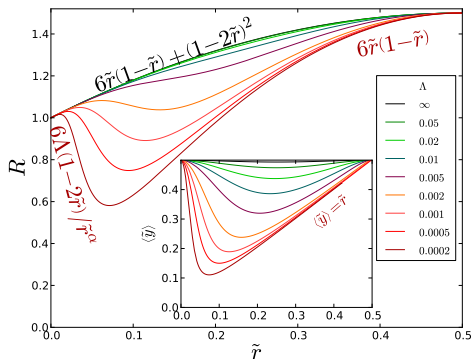
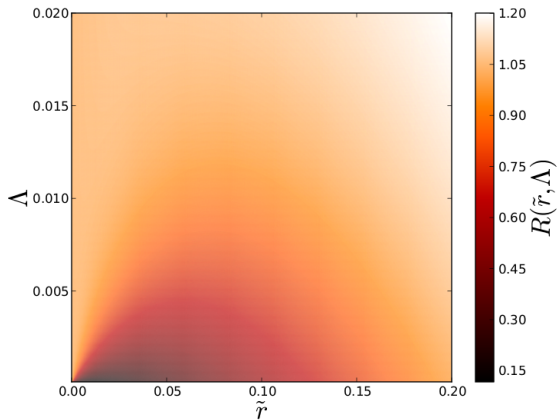
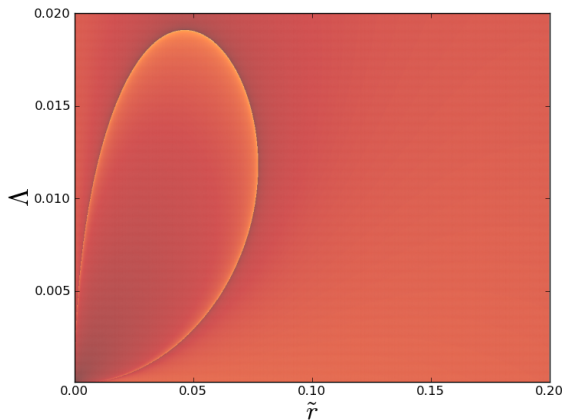


Figure: $\alpha = 2$ illustrates the HC mode better.

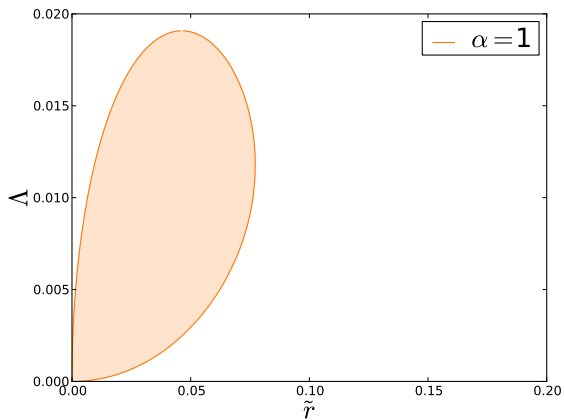
Mode-Space



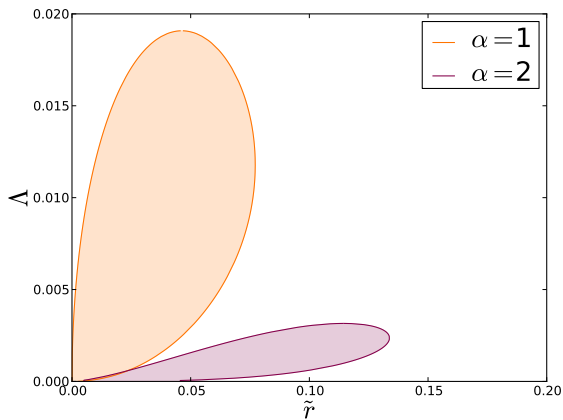
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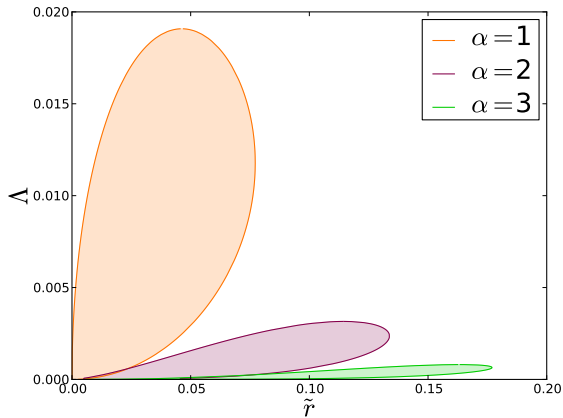
Mode-Space



Mode-Space



Mode-Space



Normal-mode operation enclosed by steric-mode. For $\alpha = 3$, normal-mode can extend as far as 18% the channel height.

Stress over Surface Area

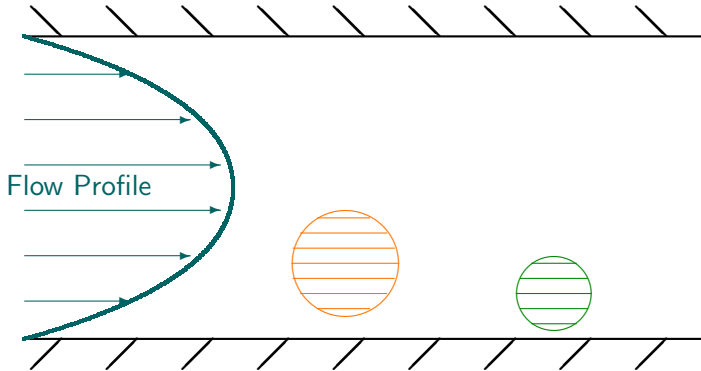


Figure: Schematic of surface area integration of stress on particle.

Finite-Size Effects

$$R = \frac{\langle \mathcal{V} \rangle}{\langle v \rangle} = \frac{\langle c \mathcal{V} \rangle}{\langle c \rangle \langle v \rangle} = \frac{1}{\langle v \rangle} \frac{\int_0^w c(y) \mathcal{V}(y) dy}{\int_0^w c(y) dy}$$

but $\mathcal{V}(\tilde{y}) \neq v(\tilde{y})$

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but $\mathcal{V}(\tilde{y}) \neq v(\tilde{y})$

Faxén's Law

$$\begin{aligned} \mathcal{V}(\tilde{y}) &= \left(1 + \frac{\tilde{r}^2}{6} \nabla^2\right) v(\tilde{y}) \\ &= 6 \langle v \rangle \left[\tilde{y} - \tilde{y}^2 - \frac{\tilde{r}^2}{3} \right] \end{aligned}$$

Retention Ratio

$$R = \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \mathcal{L} \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) + \mathcal{F}_f(\tilde{r})$$

where $\mathcal{F}_f(r, w) = 6\tilde{r} \left(1 - \frac{4}{3}\tilde{r}\right)$

Comparision

Steric Mode

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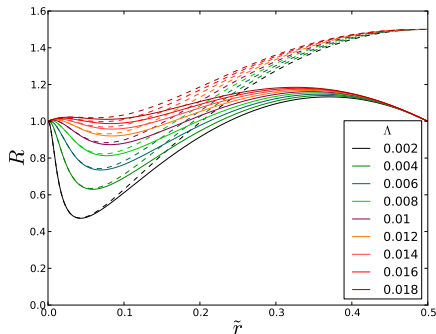
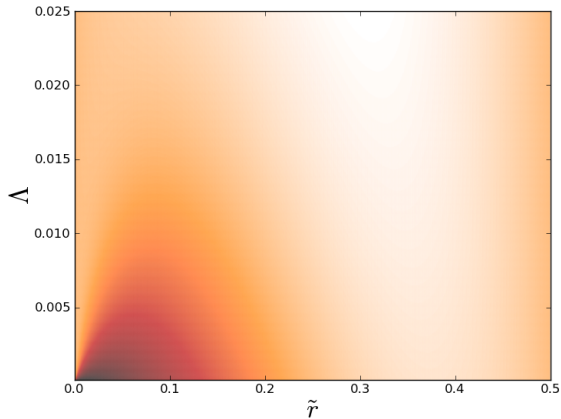
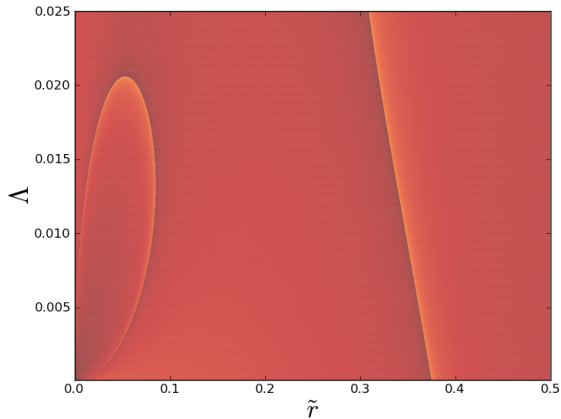


Figure: Retention ratios for $\alpha = 1$.

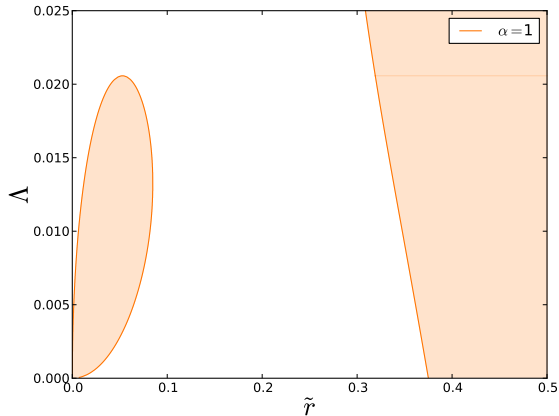
Mode-Map



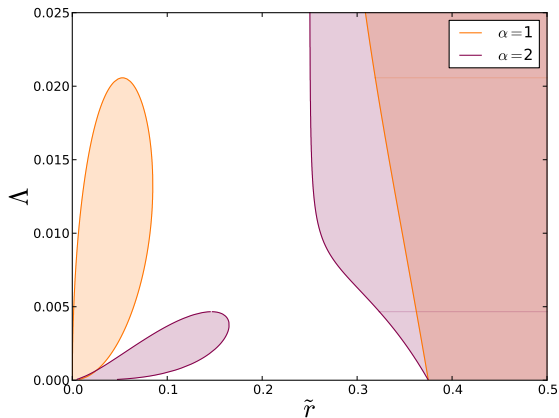
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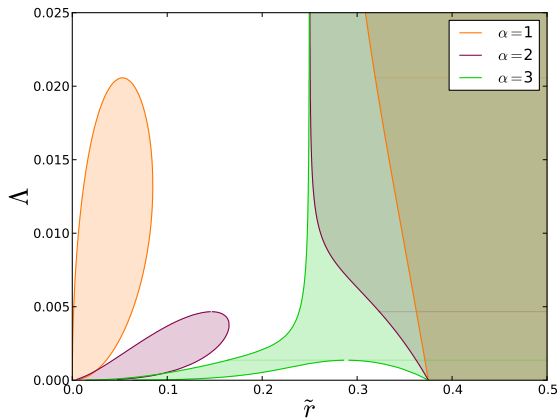
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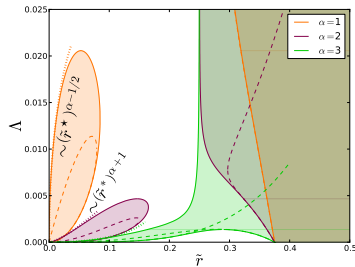
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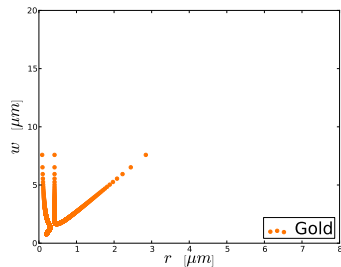
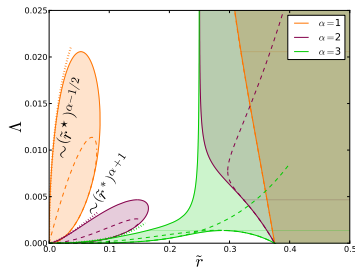
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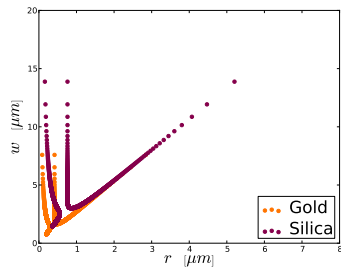
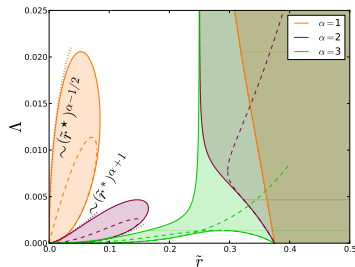
Map (experimentally relevant)



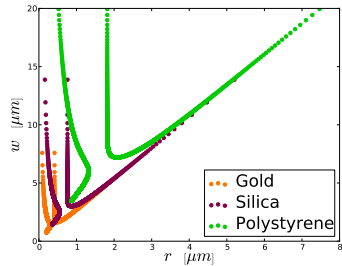
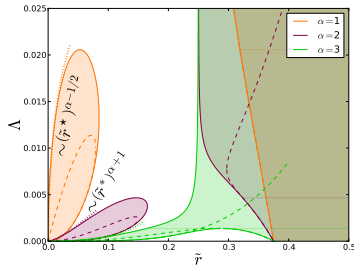
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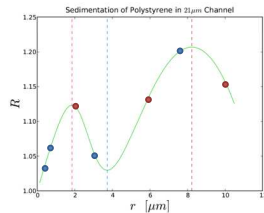
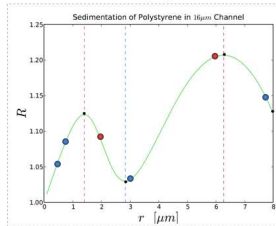


Map (experimentally relevant)



Our Choices

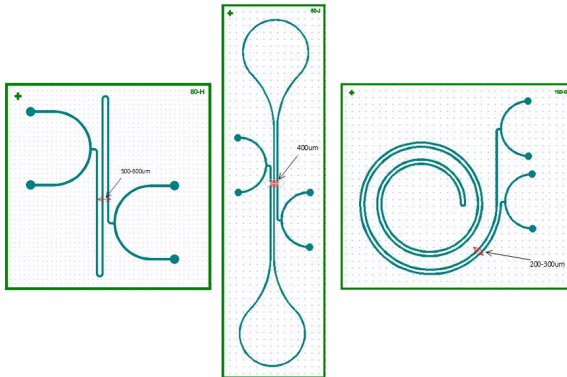
Where Particle Sizes are found on the Retention Ratio Curve



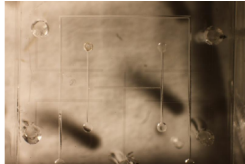
- Particle Sizes we have ($R(\mu\text{m}) = 0.49, 0.76, 3.01, 7.75$)
- Particle Sizes we are ordering ($R(\mu\text{m}) = 2.0, 6.0, 10.0$)

Microfluidic Devices

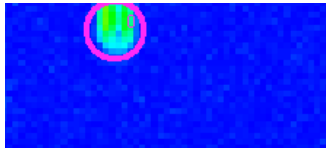
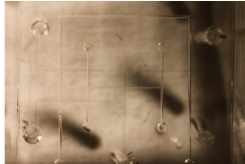
Mask Designs



Video Microscopy



Video Microscopy

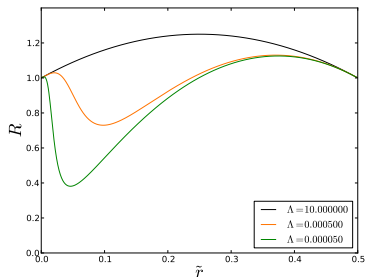


Approximations

$$R = \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \mathcal{L} \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) + \mathcal{F}_f = \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \left[\coth \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) - \frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right] + \mathcal{F}_f$$

$$\mathcal{F}_f = 6\tilde{r} \left(1 - \frac{4}{3}\tilde{r} \right)$$

Limits



Approximations

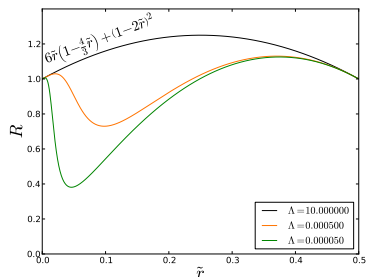
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Limits

● $\tilde{r} \rightarrow 0$

$$R \approx \mathcal{F}_f + (1 - 2\tilde{r})^2$$



Approximations

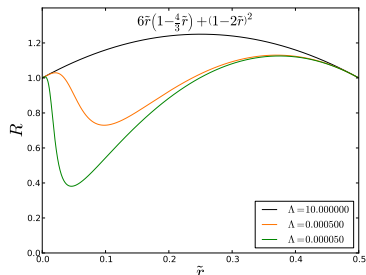
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Limits

● $\tilde{r} \rightarrow 0$ $R \approx \mathcal{F}_f + (1 - 2\tilde{r})^2$

● $\Lambda \gg 1$ $R \approx \mathcal{F}_f + (1 - 2\tilde{r})^2$



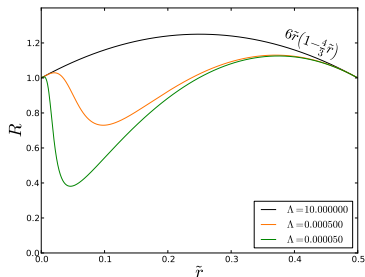
Approximations

$$R = \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \mathcal{L} \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) + \mathcal{F}_f = \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \left[\coth \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) - \frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right] + \mathcal{F}_f$$

$$\mathcal{F}_f = 6\tilde{r} \left(1 - \frac{4}{3}\tilde{r} \right)$$

Limits

- $\tilde{r} \rightarrow 0$ $R \approx \mathcal{F}_f + (1 - 2\tilde{r})^2$
- $\Lambda \gg 1$ $R \approx \mathcal{F}_f + (1 - 2\tilde{r})^2$
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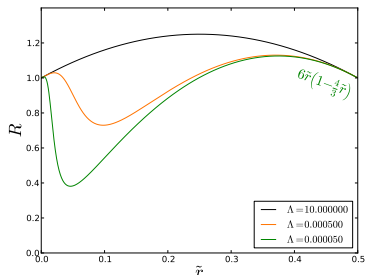
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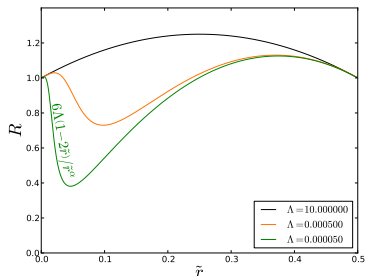
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Quadratic Approximations of Transition

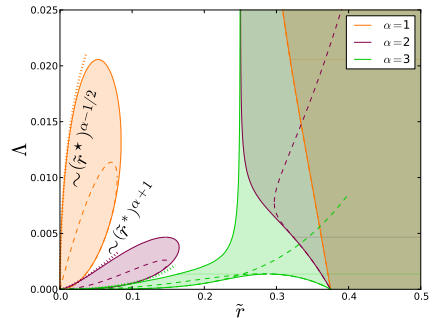
$\Lambda \ll 1$ ($\tilde{r}^*$ dashed)

$$\frac{\partial R}{\partial \tilde{r}} \approx 6 \left[a + b\Lambda + c\Lambda^2 \right]$$

$$a \equiv 1 - \frac{8}{3}\tilde{r}$$

$$b \equiv \frac{2\alpha\tilde{r} - 2\tilde{r} - \alpha}{\tilde{r}^{\alpha+1}}$$

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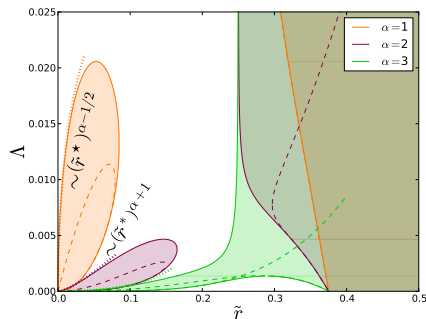
Quadratic Approximations of Transition

$\tilde{r} \ll 1$ ($\tilde{r}^*$ dotted)

$$\frac{\partial R}{\partial \tilde{r}} = A + \frac{B}{\Lambda^2} + \mathcal{O}(\tilde{r}^{2\alpha+2})$$

$$A \equiv 2 - 8\tilde{r}$$

$$B \equiv \frac{1}{5} \left[-\frac{\alpha}{6} \tilde{r}^{2\alpha-1} + \frac{2(2\alpha+1)}{3} \tilde{r}^{2\alpha} - 4(\alpha+1) \tilde{r}^{2\alpha+1} \right].$$



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Normal Mode
Steric Mode
Elution Order
HC Mode
Faxén's Mode
Verification

Real Units
Microparticles
Microchannel
Microscopy
Approximations
Conclusion

Thank you,



uOttawa



- Michel Godin
- Andrew Todd

- Mykyta Chubynsky
- Yu-guo Tao
- Henk de Haan

- David Sean
- Owen Hickey

Flow Perturbation and Wall Interaction

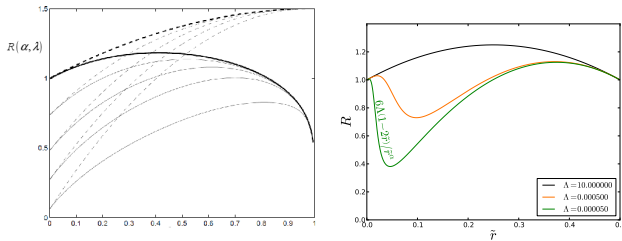


Figure: Stolen this morning from Dr. Martin. His transition to Faxén's Mode occurs at $\tilde{r} \approx 2$ which is lower than the ideal prediction of 2.5 – 4. But the slope is steeper suggesting better resolution than one would expect from the ideal theory.

Expansions

Retention Ratio

$$\begin{aligned}
 R &= \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \mathcal{L} \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) + \mathcal{F}_f \\
 &= \frac{6\Lambda}{\tilde{r}^\alpha} [1 - 2\tilde{r}] \left[\coth \left(\frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right) - \frac{[1 - 2\tilde{r}] \tilde{r}^\alpha}{2\Lambda} \right] + 6\tilde{r} \left(1 - \frac{4}{3}\tilde{r} \right)
 \end{aligned}$$

Expansions

$$\coth(x) \approx \frac{1}{x} + \frac{x}{3} - \frac{x^3}{45} + \dots$$

$$\begin{aligned}
 \coth(x) &= \frac{e^{2x} + 1}{e^{2x} - 1} \approx (1 + e^{2x}) (1 + e^{-2x} + e^{-4x} \dots) \\
 &\approx 1
 \end{aligned}$$

Retention Ratio Derivative

$$\begin{aligned} \frac{\partial R}{\partial \tilde{r}} = & \left(-\frac{6\alpha\Lambda}{\tilde{r}^{\alpha+1}} \right) \left\{ \coth \left([1 - 2\tilde{r}] \frac{\tilde{r}^\alpha}{2\Lambda} \right) (1 - 2\tilde{r}) - \frac{2\Lambda}{\tilde{r}^\alpha} + \left(1 - \frac{4}{3}\tilde{r} \right) \frac{\tilde{r}^{\alpha+1}}{\Lambda} \right\} \\ & + \left[\left\{ 1 - \coth^2 \left([1 - 2\tilde{r}] \frac{\tilde{r}^\alpha}{2\Lambda} \right) \right\} \left([1 - 2\tilde{r}] \frac{\alpha \tilde{r}^\alpha}{2\Lambda \tilde{r}} - \frac{\tilde{r}^\alpha}{\Lambda} \right) [1 - 2\tilde{r}] \right. \\ & \left. - 2 \coth \left([1 - 2\tilde{r}] \frac{\tilde{r}^\alpha}{2\Lambda} \right) + \frac{2\alpha\Lambda}{\tilde{r}^{\alpha+1}} + [1 + \alpha] \left(1 - \frac{4}{3}\tilde{r} \right) \frac{\tilde{r}^\alpha}{\Lambda} - \frac{4}{3} \frac{\tilde{r}^{\alpha+1}}{\Lambda} \right] \frac{6\Lambda}{\tilde{r}^\alpha} \end{aligned}$$

Approximations of Derivative

$$\Lambda \ll 1$$

$$\frac{\partial R}{\partial \tilde{r}} \approx 6 \left[a + b\Lambda + c\Lambda^2 \right]$$

$$a \equiv 1 - \frac{8}{3}\tilde{r}, \quad b \equiv \frac{2\alpha\tilde{r} - 2\tilde{r} - \alpha}{\tilde{r}^{\alpha+1}}, \quad c \equiv \frac{4\alpha}{\tilde{r}^{2\alpha+1}},$$

$$\tilde{r} \ll 1$$

$$\frac{\partial R}{\partial \tilde{r}} = A + \frac{B}{\Lambda^2} + \mathcal{O}(\tilde{r}^{2\alpha+2})$$

$$A \equiv 2 - 8\tilde{r},$$

$$B \equiv \frac{1}{5} \left[-\frac{\alpha}{6}\tilde{r}^{2\alpha-1} + \frac{2(2\alpha+1)}{3}\tilde{r}^{2\alpha} - 4(\alpha+1)\tilde{r}^{2\alpha+1} \right].$$

Concentrations

Normal

$$\langle \tilde{y} \rangle = \lambda + \frac{1}{1 - e^{1/\lambda}}$$

Steric

$$\langle \tilde{y} \rangle = \lambda + \frac{1}{1 - e^{(1-2\tilde{r})/\lambda}} + \tilde{r} \coth \left(\frac{1 - 2\tilde{r}}{2\lambda} \right)$$

Plate Height and Resolution

$$H = \frac{D_{\text{eff}}}{R \langle v \rangle}$$

$$= \frac{D_{\text{mol}}}{R \langle v \rangle} \left[1 + \chi \frac{R}{2} \left(\frac{\langle v \rangle w}{D_{\text{mol}}} \right)^2 \right]$$

$$\chi \propto \Lambda^3 r^{3\alpha}$$

$$D_{\text{mol}} \propto r$$

$$R_S = \frac{1}{2(H_1 + H_2)} \left(\frac{1}{\langle v_2 \rangle} - \frac{1}{\langle v_1 \rangle} \right)$$