Geometry controlled dispersion in periodic channels

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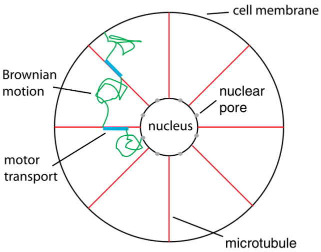
How to characterize the dispersion of small particles in channels ?

Some examples of dispersion in confined media 1

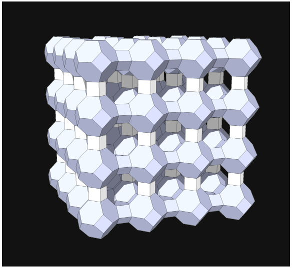
- How fast does a cloud of tracer particles disperse in heterogeneous media ?
- Active field of research.
- Dispersion in confined media :

biological cells



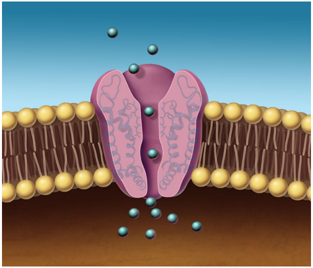
Bressloff and Newby, RMP (2013)

zeolites



Karger and Ruthven (1992)

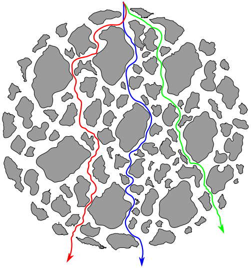
ion channels



Some examples of dispersion in confined media 2

- Dispersion in confined media :

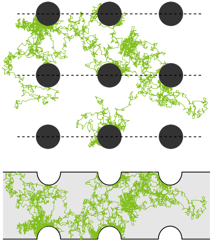
contaminant spreading in porous media



Tzella and Vanneste, PRL (2016)

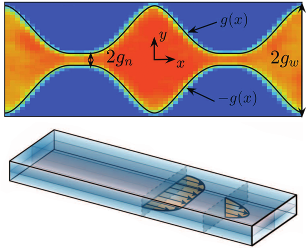
Leitmann and Franosch, PRL (2017)

mapping onto diffusion in channels



Dagdug *et al.*, JCP (2012)

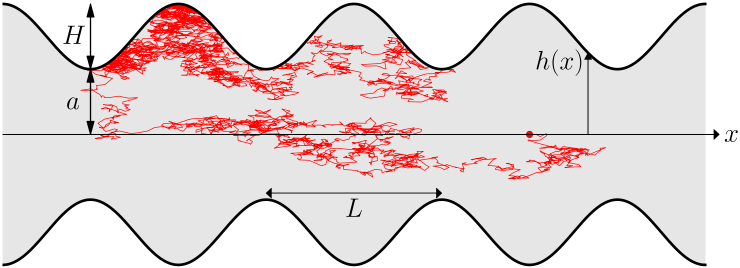
microfluidic devices



Yang *et al.*, PNAS (2017)

Aminian *et al.*, Science (2016)

The dispersion in symmetric two-dimensional periodic channels



- Periodicity of the channel : $\boldsymbol{L}$.
- Height of the channel : $\boldsymbol{h(x) = a\zeta(x)}$, where a is the minimum height.
- We define $\boldsymbol{\varepsilon = a/L}$ and $\boldsymbol{\xi = H/a}$, where H is the variation of height.
- Microscopic, homogeneous and isotropic, diffusivity D_0. What global dispersion appears at long time ?

Dynamics of overdamped Brownian particles

- The overdamped Langevin equation for the position

$$\frac{d\mathbf{X}_t}{dt} = \sqrt{2D_0}\boldsymbol{\eta}$$

$$\overline{\eta_i(t)\eta_j(t')} = \delta_{ij}\delta(t-t')$$

- The Fokker-Planck equation for the probability density function (pdf)

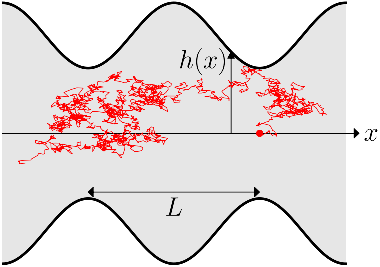
$$\frac{\partial p}{\partial t} = D_0\boldsymbol{\nabla}^2 p = -\nabla\cdot\mathbf{J}$$

$$\boldsymbol{n}\cdot\boldsymbol{\nabla}p = \boldsymbol{n}\cdot\mathbf{J} = 0$$

Stationary properties studied

- Mean squared displacement (MSD) : $\overline{\mathbf{X}^2(t)}$.
- Long-time effective diffusivity along the channel : $D_e = \lim_{t\to\infty}\frac{\overline{x^2(t)}}{2t}$.

Famous traditional approach : reduction to one-dimensional problem



- narrow regions = entropic barriers
- wide regions = entropic traps
- Entropic trapping of particles
- Slowing down of dispersion

$$D_e \leq D_0$$

- Marginal probability $p(x) \propto h(x) \propto \exp[-\beta\varphi(x)]$
- Effective entropy $s(x) = -\beta\varphi(x) = \ln h(x)$
- Long-time effective diffusivity

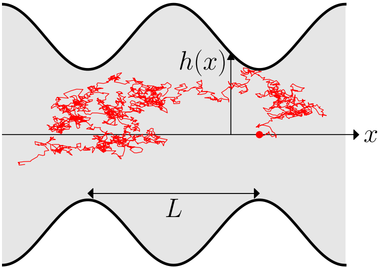
$$\frac{D_e}{D_0} = \frac{1}{\langle \exp(-\beta\varphi)\rangle\langle \exp(\beta\varphi)\rangle} = \frac{1}{\langle h\rangle\langle h^{-1}\rangle} \leq 1 \text{ [Jensen's inequality]}$$

Lifson and Jackson, JCP (1961)

- Spatial average

$$\langle h\rangle = \frac{1}{L}\int_0^L dx\ h(x)$$

Fick-Jacobs' one-dimensional equation



- Reduced one-dimensional pdf (assuming fast equilibration in y)

$$p^*(x;t) = \int_{-h(x)}^{h(x)} dy\ p(x,y;t) \simeq 2h(x)p_0(x;t)$$

- Fick-Jacobs' equation

$$\frac{\partial p^*}{\partial t} = D_0 \frac{\partial}{\partial x}\left[\frac{\partial p^*}{\partial x} - \beta\varphi'(x)p^*\right] = D_0 \frac{\partial}{\partial x}\left[\frac{\partial p^*}{\partial x} - \frac{h'(x)}{h(x)}p^*\right]$$

Jacobs, Diffusion Processes (1935)

Existing improvements of Fick-Jacobs approximation

- Modified Fick-Jacobs' equation

$$\frac{\partial p^*}{\partial t} = \frac{\partial}{\partial x} D(x) \left[\frac{\partial p^*}{\partial x} - \frac{h'(x)}{h(x)} p^* \right]$$

- Effective one-dimensional diffusivity

$$D(x) = D_0 \left(1 - \frac{1}{3} h'(x)^2 + \cdots \right) \simeq D_0 \frac{\arctan h'(x)}{h'(x)}$$

- Long-time effective diffusivity

$$D_e = \frac{1}{\langle h \rangle \langle D^{-1} h^{-1} \rangle} = \frac{D_0}{\langle h \rangle \langle h^{-1} \rangle} \left(1 - \frac{1}{3} \frac{\langle h'^2 / h \rangle}{\langle h^{-1} \rangle} + \cdots \right)$$

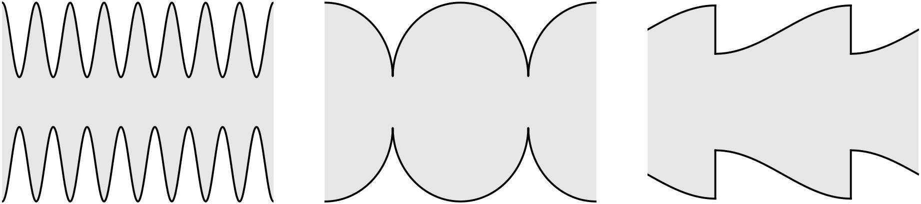
- Valid when $h'(x) \ll 1$, for continuous channels.
- Effectively one-dimensional Markovian process.

Zwanzig, JPC (1991)
Reguera and Rubí, PRE (2001)

Kalinay and Percus, PRE (2006)
Bradley, PRE (2009)
Dorfman and Yariv, JCP (2014)

Limits of Fick-Jacobs improvements

- The FJ approximation is valid only for slowly varying channels ($\varepsilon \ll 1$).
- The effective one-dimensional process is clearly non Markovian, the definition of an effective diffusivity $D(x)$ is not valid.



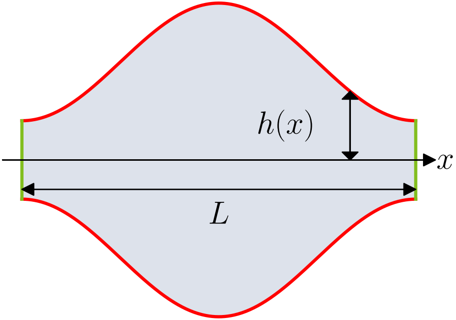
- What happens for wide channels ($\varepsilon \gg 1$) ? the intermediate range of ε ? sharp-neck channels ? discontinuous channels ?

Starting point : Exact formula of D_e, non relying on 1d-reduction

- Explicit equation for the long-time effective diffusivity

$$\frac{D_e}{D_0} = 1 - \frac{1}{\langle h \rangle} \int_0^L dx\ h'(x)\ f(x, h(x))$$

- Partial differential equations for the auxilliary function $f(x, y)$



$$\frac{\partial^2 f}{\partial x^2} + \frac{\partial^2 f}{\partial y^2} = 0$$

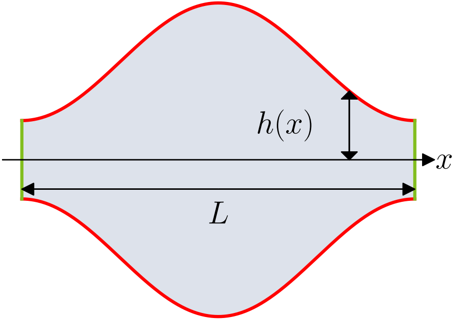
$$h'(x)\ \frac{\partial f}{\partial x}\Big|_{y=h(x)} - \frac{\partial f}{\partial y}\Big|_{y=h(x)} = h'(x)$$

$$f(x+L, y) = f(x, y)$$

Guérin and Dean, PRE (2015)

Simplifications using complex analysis

- Auxilliary function f as an analytic function $w(z = x + iy)$



$$f(x, y) = \frac{1}{2}[w(x + iy) + w(x - iy)]$$
$$= \mathrm{Re}\, w(x + iy)$$

$$\mathrm{Im}\, w(x + ih(x)) = h(x) - C$$

$$w(z + L) = w(z)$$

- Long-time effective diffusivity

$$\frac{D_e}{D_0} = \frac{C}{\langle h \rangle}$$

Back to the Fick Jacobs approximation when $\varepsilon \ll 1$

- Leading order of perturbation :

$$\mathrm{Im}\, w(x + ih(x)) = h(x) - C$$

$$h(x) w'(x) = h(x) - C$$

$$w'(x) = 1 - C h(x)^{-1}$$

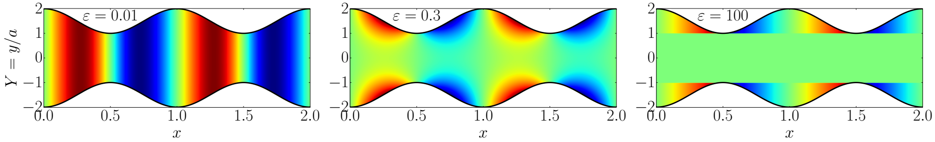
- The periodicity gives $C = \langle h^{-1} \rangle^{-1}$
- Fick-Jacobs' effective diffusivity

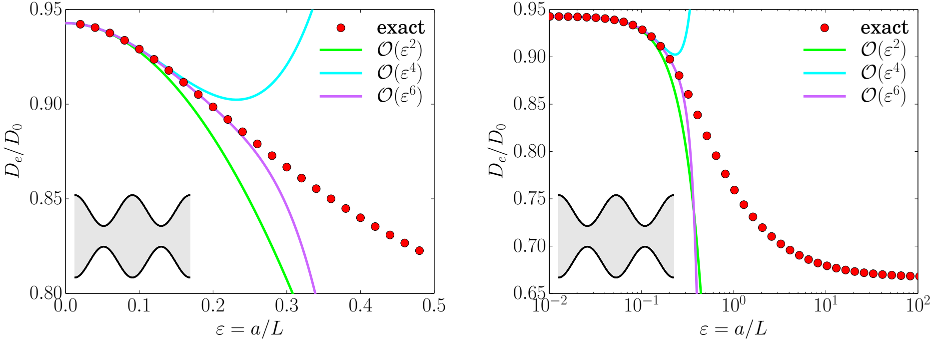
$$\frac{D_e}{D_0} = \frac{C}{\langle h \rangle} = \frac{1}{\langle h \rangle \langle h^{-1} \rangle}$$

- Corrections up to $\mathcal{O}(\varepsilon^6)$:

$$\frac{D_e}{D_0} = \frac{1}{\langle \zeta \rangle} \Bigg[\langle \zeta^{-1} \rangle + \frac{\varepsilon^2}{3} \langle \zeta'^2 / \zeta \rangle - \frac{\varepsilon^4}{45} \left(4 \langle \zeta'^4 / \zeta \rangle + \langle \zeta''^2 \zeta \rangle \right) + \frac{\varepsilon^6}{945} \left(44 \langle \zeta'^6 / \zeta \rangle + 5 \langle \zeta^2 \zeta''^3 \rangle + 45 \langle \zeta'^3 \zeta''^2 \rangle + 2 \langle \zeta^3 \zeta'''^2 \rangle \right) \Bigg]^{-1}$$

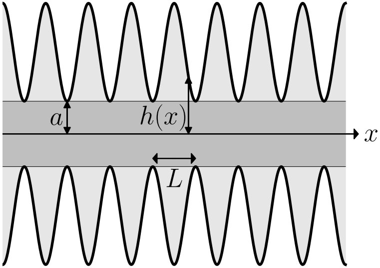
Numerical results : validity of the perturbative expansion





- Improvements of D_e at each order.
- What happens for wide channels ($\varepsilon \gg 1$) ?

Entropic trapping of particles in wide channels



- Particles trapped in regions $|y| > a$

$$\langle x^2(t)\rangle = 2D_e t \geq 2D_0 t'$$

- t' is the time spend in $|y| < a$ region.

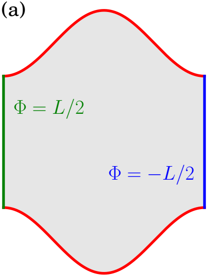
- The long-time effective diffusivity is bounded

$$1 \geq \frac{D_e}{D_0} \geq \frac{t'}{t} = \frac{a}{\langle h\rangle}$$

- For $a \gg L$, the lower bound becomes exact.
- Presence of a boundary layer at $y = a$.

Finding the finite corrections in the wide channel regime

(a)

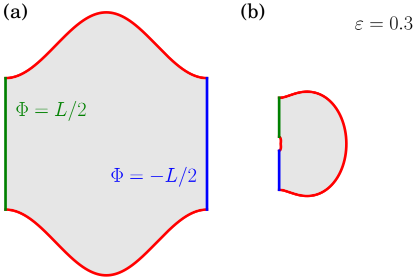


$$\widetilde{w}(z) = w(z) - z = \Phi(x, y) + i\Psi(x, y)$$

$$\mathrm{Im}\,\widetilde{w}(x + ih(x)) = -C$$

$$\widetilde{w}(z + L) = \widetilde{w}(z) - L$$

Finding the finite corrections in the wide channel regime



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- Conformal mapping $G(z) = \exp[-i\pi(z - ia)]$

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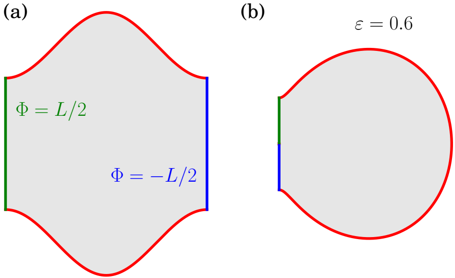

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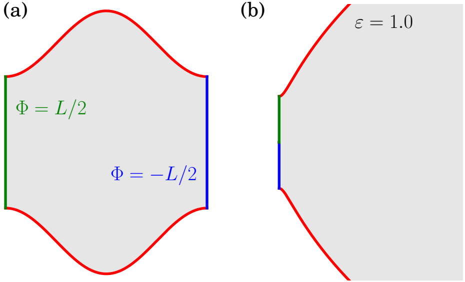

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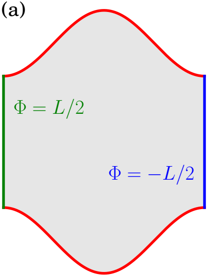
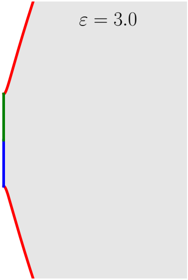


(b)



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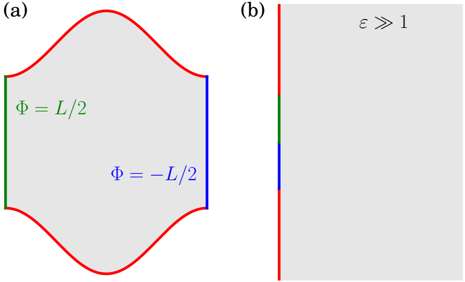
Finding the finite corrections in the wide channel regime



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$$\widetilde{w}(z + L) = \widetilde{w}(z) - L$$

- Conformal mapping $G(z) = \exp[-i\pi(z - ia)]$
- Analogy to an electrostatic problem $\widetilde{w}(z) = \frac{1}{\pi} \arcsin\left[iG(z)^{-1}\right]$
- Long-time effective diffusivity, with a correction $\mathcal{O}(1/\varepsilon)$

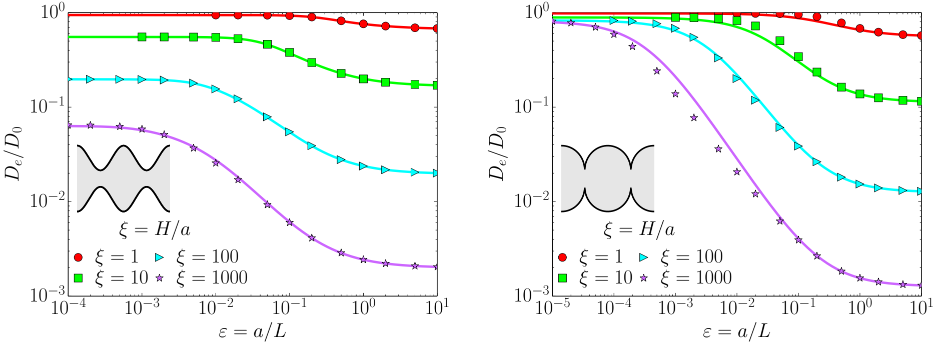
$$\frac{D_e}{D_0} = \frac{a}{\langle h \rangle}\left(1 + \frac{\ln 2}{\pi\varepsilon}\right)$$

- Identification of a universal constant $\ln 2/\pi$.

Intermediate regime of dispersion

- Padé approximant with $\varepsilon \ll 1$ and $\varepsilon \gg 1$ expansions

$$\frac{D_e}{D_0} = \frac{1}{\langle h \rangle \langle h^{-1} \rangle} \frac{1 + a_1\varepsilon + a_2\varepsilon^2 + \cdots}{1 + b_1\varepsilon + b_2\varepsilon^2 + \cdots}$$

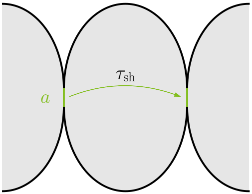


- The Padé approximant fit well the numerical data for smooth channels.
- What happens in the large-ξ limit, in presence of small openings ?

Narrow escape regime for small openings - $\varepsilon \ll 1$ and $\xi \gg 1$

Behavior of the height near the neck : $\zeta(x) \simeq 1 + \xi x^\nu$

- Sharp-neck channels ($\nu < 1$)

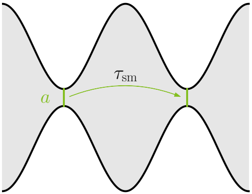


- τ_{sh} : FPT to reach a small opening
- Long-time effective diffusivity

$$\frac{D_e}{D_0} = \frac{L^2}{2D_0\tau_{\text{sh}}} = \frac{L}{\langle h\rangle}\frac{\pi}{2\ln(K/\varepsilon)}$$

- D_{FJ} independant of ξ.
- Expression valid for smooth channels if $H/L = \varepsilon\xi$ is not too big.

- Smooth-neck channels ($\nu > 1$)

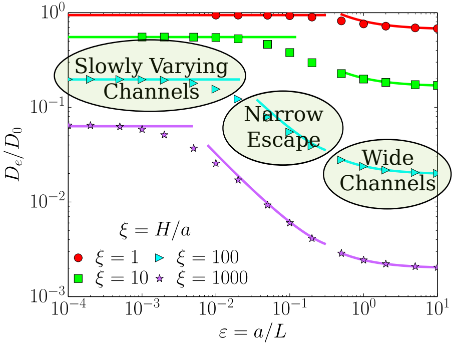


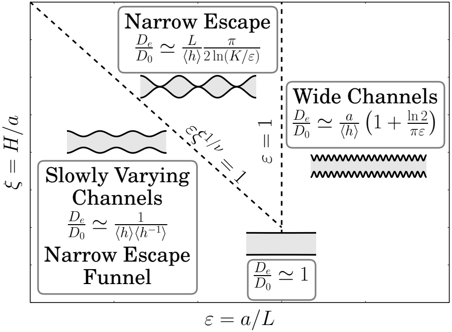
- τ_{sm} : FPT to reach a small opening.
- Fick-Jacobs' diffusivity is

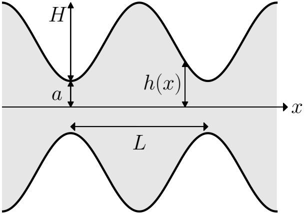
$$D_{\text{FJ}} = \frac{L^2}{2\tau_{\text{sm}}} \simeq \xi^{1/\nu-1}$$

- Valid approach for $H/L = \varepsilon\xi \gg 1$.

Classification of dispersion regimes for smooth-neck channels ($\nu > 1$)

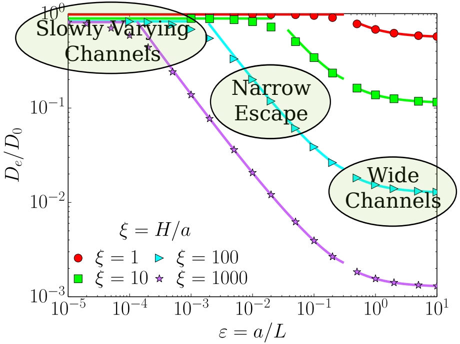


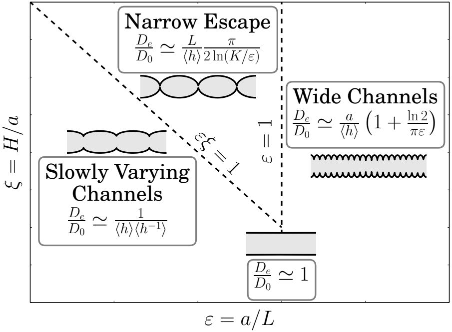


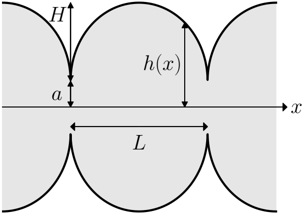


- Three regimes of dispersion ($d = 2$ and 3).
- Geometry controlled dispersion in channels.
- What happens for discontinuous channels ?

Classification of dispersion regimes for sharp-neck channels ($\nu < 1$)







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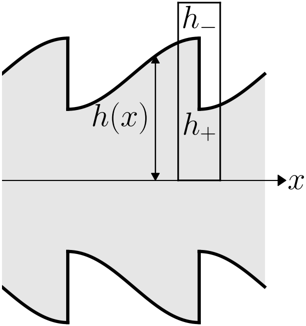
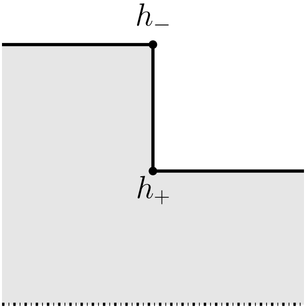
Failure of perturbation theory ($h'(x) = \infty$)



- Discontinuity at $x = 0$. Perturbative solution valid at $x \neq 0$.
- Boundary layer at the discontinuity.

Failure of perturbation theory ($h'(x) = \infty$)



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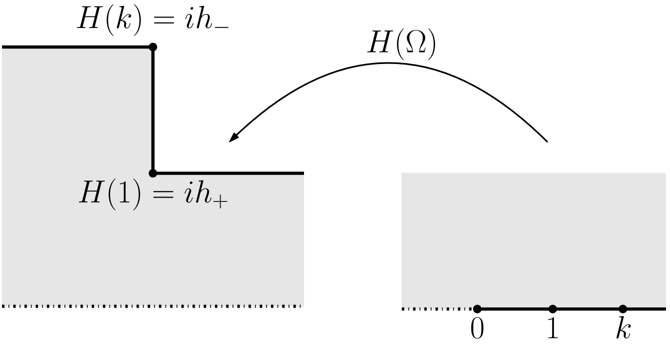


- Discontinuity at $x = 0$. Perturbative solution valid at $x \neq 0$.
- Boundary layer at the discontinuity.
- Conformal mapping $z = H(\Omega)$ via a Schwarz-Christoffel transformation

$$H'(\Omega) = H_0 \frac{\sqrt{\Omega - 1}}{\Omega\sqrt{\Omega - k}}, \; k^2 = h_-/h_+$$

Trapping rate associated to discontinuities

- Long-time effective diffusivity, with a correction $\mathcal{O}(\varepsilon)$ instead of $\mathcal{O}(\varepsilon^2)$!

$$\frac{D_e}{D_0} = \frac{1}{\langle h \rangle \langle h^{-1} \rangle} \left(1 - \frac{\varepsilon}{\pi \langle \zeta^{-1} \rangle} \sum_i \gamma(\nu_i) \right)$$

- The discontinuity plays the role of a local trap.
- Exact function $\gamma(\nu)$, $\nu = h_+/h_- \leq 1$

$$\gamma(\nu) = \frac{1+\nu^2}{\nu} \ln \frac{1+\nu}{1-\nu} - 2 \ln \frac{4\nu}{1-\nu^2}$$

- Berezhkovskii's numerical interpolation

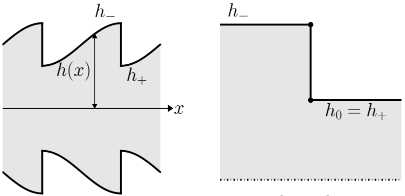
$$\gamma(\nu) = 2(1-\nu)^2 \ln \left(2.6 + \frac{0.7}{\nu} \right)$$

- Kalinay and Percus only give $\gamma(1/2) = 0.7848$.

Berezhkovskii *et al.*, JCP (2009)

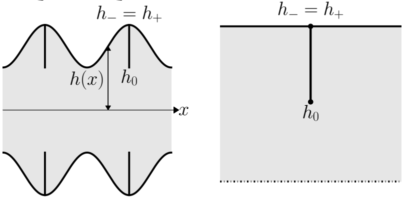
Kalinay and Percus, PRE (2010)

Three kinds of discontinuities



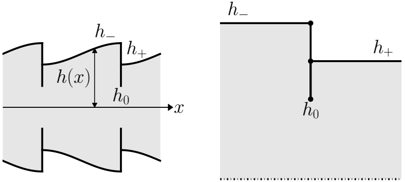
$$\gamma(\nu) = \frac{1+\nu^2}{\nu} \ln \frac{1+\nu}{1-\nu} - 2 \ln \frac{4\nu}{1-\nu^2}$$

$$\nu = h_+/h_- \leq 1$$



$$\gamma(\nu) = -4 \ln \sin \frac{\pi\nu}{2}$$

$$\nu = h_0/h_- \leq 1$$



$$\gamma(\nu, \tilde{\nu}) = \frac{1+(\nu\tilde{\nu})^2}{\nu\tilde{\nu}} \ln \frac{\nu+\tilde{\nu}}{\nu-\tilde{\nu}} - 2 \ln \frac{4\nu\tilde{\nu}}{\nu^2-\tilde{\nu}^2}$$

ν and $\tilde{\nu}$ functions of h_0/h_- and h_0/h_+.

What we have characterized

- Exact PDEs to get the effective diffusivity, simplified by the use of complex analysis in two dimensions through a compact formalism.
- Fick-Jacobs approximation revisited and wide channels limit characterized by a universal constant.
- Classification of all regimes of dispersion for periodic smooth-neck and sharp-neck channels.
- Discontinuities associated to an effective local trapping.
- The dispersion of particles is controlled by the geometry of the channel.

Geometry controlled dispersion in periodic corrugated channels, EPL **118** (2017).

Dispersion in two dimensional channels — the Fick-Jacobs approximation revisited, JSM (2017).

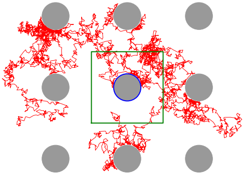
Dispersion in two dimensional periodic channels with discontinuous profiles, in preparation.

What is the next step ?

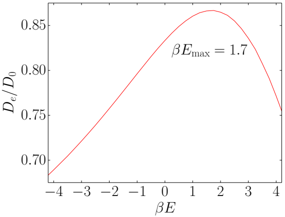
- What happens with a longitudinal force ? In presence of hydrodynamics flows for example.
- What happens with a normal force ? In presence of gravity for example.
- How to characterize the dispersion in moving channels $h(x,t)$? For biological systems for example.
- Is the complex analysis possible in 3-dimensions ?

Thank you for your attention !

Dispersion of particles in periodic lattice of spheres

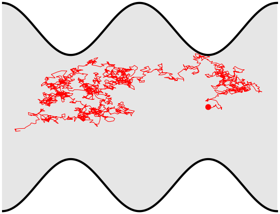


- attractive sphere $\mathbf{F} = -\boldsymbol{\nabla} V$
- $V(r) = -E \exp(-\lambda(r-R))\theta(r-R)$
- non monotonic dispersion



Putzel *et al.*, PRL (2014)

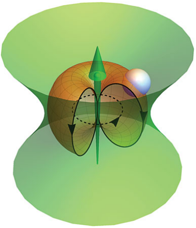
Dispersion of particles into periodic channels

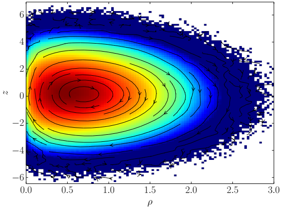


- no drift $\mathbf{F} = \mathbf{0}$
- entropic effects of the boundaries
- slowing down of dynamics
- control of the dispersion

Mangeat *et al.*, EPL (2017), JSP (2017)

Optical trapping of diffusive particles





- $\mathbf{F} = -\boldsymbol{\nabla} V + \mathbf{F}_{\rm nc}$
- Non conservative force

$$\mathbf{F}_{\rm nc} = F_0 \left(1 - \frac{\rho^2}{a^2}\right) \mathbf{e_z}$$

- non equilibrium state
- presence of a stationary current

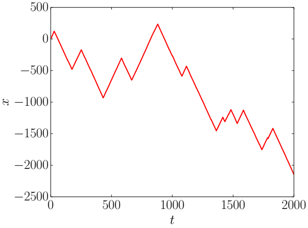
Grier *et al.*, PRL (2008), RSTA (2017)

Self-propelled autochemotactic walker

- Force due to the concentration of walkers $\mathbf{F} = -\lambda \boldsymbol{\nabla} c$

$$\frac{d\mathbf{X}_t}{dt} = \Lambda \int_0^t ds\ \mathbf{K}(\mathbf{X}_s, s) + \sqrt{2D_0}\boldsymbol{\eta}$$

- non Markovian dynamics
- intermediate time ballistic dynamics



Löwen *et al.*, PRE (2009)