An annealed model of diffusion in semi-permeable domains

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Reading progress

- *Conformal Field Theory* (CFT) - Chpt. 5
  - Goal: Complete reading Chpt. 1-7, 10, 12, 13-15 before end of term
- *Critical Dynamics* (CD) - Chpt. 4
  - Goal: Complete reading Chpt. 1-9 before end of term
- *Quantum Field Theory and the Standard Model* (QFT) - Chpt. 18
  - Goal: Complete reading Chpt. 14-23 before end of term
- *Field Theory of Non-Equilibrium Systems* (FTNE) - DNS
  - Goal: Complete reading Chpt. 1-2, 6, 9-12 before end of term
- Working on finishing typesetting for CFT Chpt. 4 & CD Chpt. 3

The model

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- Reflected Brownian motion (RBM) occurs, governed by the Skorokhod equation[1]

$$dX_t^{(1)} = \sqrt{2D_1}\, dW_t + n(x)\, d\ell_t^{(1)}, \tag{1}$$

until the boundary local time,

$$\ell_t^{(1)} := 2Dn \int_{\partial\mathcal{B}_1} d^{n-1}x \int_0^t \delta(X_s - x)\, ds \tag{2}$$

exceeds a random variable $\hat{\ell}^{(1)}$, whose probability law obeys $\mathbb{P}\{\hat{\ell}^{(1)} > \ell\} = e^{-q_1\ell}$, for $q_1 = \kappa_1/D_1$

The model

- Once the (boundary) *reaction time* $\mathcal{T}_1 = \inf\left\{t > 0 : \ell_t^{(1)} > \hat{\ell}^{(1)}\right\}$ for the domain has been reached, an new RBM initiates at the centre of another sphere $\mathcal{B}_2$ with sampled parameters (D_2, r_2, κ_2)
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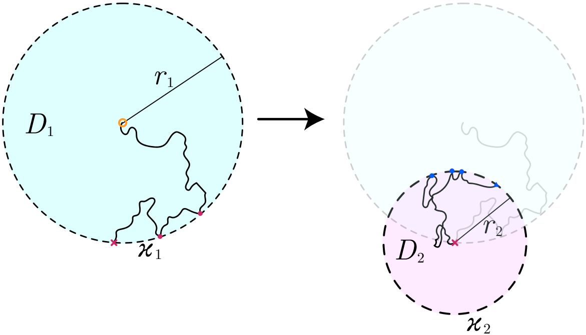

The exit time density

- Coarse graining: view the exit from a domain as a single step on a (continuous) "lattice". Assume that direction of step is unbiased (symmetry of sphere)[2]

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- $\mathbb{P}_{x_0}\{\ell_t > \ell\} \iff \mathbb{P}_{x_0}\{t > \mathcal{T}_\ell\}$ and so

$$U(\ell, t \mid x_0) = \frac{\partial \mathbb{P}_{x_0}\{t > \mathcal{T}_\ell\}}{\partial t} = \int_\ell^\infty d\ell' \, \frac{\partial \rho(\ell', t \mid x_0)}{\partial t}, \tag{3}$$

where ρ is the PDF of the boundary local time[1]

The Dirichlet-to-Neumann operator

- After taking a temporal Laplace transform ($t \xrightarrow{\mathcal{L}} \alpha$), it can be shown that U permits a spectral representation in terms of the eigenfunctions $v_n^{(\alpha)}(x)$ of the Dirichlet-to-Neumann (DtN) operator[1],

$$\tilde{U}(\ell, \alpha \mid x_0) = \sum_n e^{-\mu_n^\alpha \ell} [V_n^{(\alpha)}(x_0)]^* \int_{\partial\Omega} d^{n-1}y \, V_n^{(\alpha)}(y) \qquad (4)$$

with V_n denoting the projection of the eigenfunction onto the absorbing diffusive current,

$$V_n^{(\alpha)}(x) = \int_{\partial\Omega} d^{d-1}y \, v_n^{(\alpha)}(y) \tilde{j}_\infty(y, \alpha \mid x) \qquad (5)$$

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  - Let w be a solution to the Helmholtz equation $(\alpha - D\Delta)w = 0$ with Dirichlet BC $w|_{\partial\Omega} = f$. The DtN operator $\mathcal{M}_\alpha$ is defined by $[\mathcal{M}_\alpha f](x) = \partial_n w|_{x\in\partial\Omega}$

DtN eigenvalues in circular domains

- In general we can write the eigenequation as

$$\mathcal{M}_\alpha v_n^{(\alpha)}(x) = \mu_n^{(\alpha)} v_n^{(\alpha)}(x) \tag{6}$$

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- We allow for arbitrary boundary data $f(\theta)$. Let $u_n(r,\phi) = Q_n(r)e^{in\phi}$ with $\lambda = \sqrt{\alpha/D}$. The modified Helmholtz equation reads

$$r^2 Q_n''(r) + rQ_n'(r) - \left(n^2 + \lambda^2 r^2\right) Q_n(r) = 0$$

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- Solutions are modified Bessel functions. Enforcing regularity,

$$u(r,\phi,\alpha) = \sum_{n=-\infty}^{\infty} c_n I_{|n|}(\lambda r) e^{in\phi} \tag{7}$$

DtN eigenvalues in circular domains

- But now we note that $e^{in\phi}$ is not altered by the normal derivative. As such, we may identify the eigenfunctions $v_n(\phi) = e^{in\phi}$ as the Fourier modes and compute

$$[\mathcal{M}_\alpha v_n](\phi) = \partial_r u_n(R,\phi) = \lambda \frac{I'_{|n|}(\lambda R)}{I_{|n|}(\lambda R)} e^{in\phi},$$

so we can write the eigenvalues

$$\mu_n^{(\alpha)} = \lambda \frac{I'_{|n|}(\lambda R)}{I_{|n|}(\lambda R)} = \lambda \frac{d}{dz} \log I_{|n|}(\lambda R), \qquad \lambda = \sqrt{\alpha/D} \tag{8}$$

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- We can approximate these eigenvalues at long times, $\lambda R \ll 1$ as

$$\mu_n^{(\alpha)} \sim \frac{n}{R} + \frac{\lambda^2 R}{2(n+1)} - \frac{\lambda^4 R^3}{8(n+1)^2(n+2)} + \mathcal{O}(\lambda^5 R^5) \tag{9}$$

The absorbing propagator

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- In order to evaluate the perfectly-reactive (absorbing) diffusive current, we must determine its propagator. Assuming all motion begins at the centre of the circular domain of radius R, we find

$$\tilde{G}_\infty(r, \alpha \mid r_0 = 0) = \frac{1}{\pi R^2} \sum_n \frac{1}{\alpha + D j_n^2 / R} \frac{J_0\left(r j_n / R\right)}{[J_1(j_n)]^2} \tag{10}$$

where J_α is a Bessel function of the first kind and j_n is the nth zero of the function J_0

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  where J_α is a Bessel function of the first kind and j_n is the nth zero of the function J_0
- Computing the normal derivative and multipling by $-D$,

$$-\frac{\tilde{j}_\infty(y,t \mid x_0)}{D} = \left.\frac{\partial \tilde{G}_\infty(x,\alpha \mid x_0)}{\partial n}\right|_{x=y} = \frac{1}{\pi R^3}\sum_n \frac{j_n}{\alpha + \frac{Dj_n^2}{R}}\frac{J_0'\left(j_n\right)}{[J_1(j_n)]^2}$$

Projection operators

- The *projection operator* of the (circular) DtN eigenfunctions are the Fourier transform of the absorptive current,

$$V_n^{(\alpha)}(r_0, \phi_0) = \int_0^{2\pi} d\phi \, e^{in\phi} \tilde{j}_\infty(\phi, \alpha \mid r_0, \phi_0)$$

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- Assuming motions start from $r_0 = 0$, $\partial_\phi \tilde{j} = 0$ so these integrals vanish for all $n \neq 0$:

$$V_n^{(\alpha)}(r_0 = 0) = -\frac{2D}{\pi R^3} \delta_{0,n} \sum_m \frac{j_m}{\alpha + D j_m^2 / R} \frac{J_0'(j_m)}{[J_1(j_m)]^2} \tag{11}$$

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- Substituting into the equation for the exit time density,

$$\tilde{U}(\ell, \alpha \mid x_0) = \frac{8\pi D^2}{R^6} e^{-\lambda \ell \frac{d}{dz} \log I_0(\lambda R)} \left(\sum_m \frac{j_m}{\alpha + \frac{D j_m^2}{R}} \frac{J_0'(j_m)}{[J_1(j_m)]^2} \right)^2 \quad (12)$$

References

[1] D. S. Grebenkov, Phys. Rev. Lett. **125**, 078102 (2020).

[2] P. Massignan, C. Manzo, J. A. Torreno-Pina, M. F. García-Parajo, M. Lewenstein, and G. J. Lapeyre, Phys. Rev. Lett. **112**, 150603 (2014).

[3] D. S. Grebenkov, J. Chem. Phys. **151**, 104108 (2019).

Details of spectral representation

Probabilistically, we can understand diffusion within a domain Ω with semi-permeable boundary of permissivity $\kappa(x)$ as the BVP[3]

$$\frac{\partial G(x,t \mid x_0)}{\partial t} - D\Delta G(x,t, \mid x_0) = 0, \quad x \in \Omega \tag{13a}$$

$$G(x,t=0 \mid x_0) = \delta(x - x_0) \tag{13b}$$

$$\left(D\frac{\partial}{\partial n_x} + \kappa(x)\right) G(x,t \mid x_0) = 0, \quad x \in \partial\Omega. \tag{13c}$$

If we are to take the Laplace transform with respect to time,

$$\tilde{G}(x,\alpha \mid x_0) = \int_0^\infty e^{-\alpha t} G(x,t \mid x_0)\, dt, \tag{14}$$

this reduces Eq. (13a) to the inhomogeneous Helmholtz problem:

$$(\alpha - D\Delta)\tilde{G}(x,\alpha \mid x_0) = \delta(x - x_0). \tag{15}$$

Details of spectral representation

Since the problem is linear, we are free to take

$$\tilde{G}(x,\alpha \mid x_0) = \tilde{G}_0(x,\alpha \mid x_0) + \tilde{g}(x,\alpha \mid x_0), \tag{16}$$

where $\tilde{G}_\infty$ is the propagator with absorbing boundary conditions,

$$(\alpha - D\Delta)\tilde{G}_\infty(x,\alpha \mid x_0) = \delta(x - x_0), \quad x \in \Omega \tag{17a}$$

$$\tilde{G}_\infty(x,\alpha \mid x_0) = 0, \quad x \in \partial\Omega. \tag{17b}$$

Thus the unknown contribution which reacts with the boundary satisfies

$$(\alpha - D\Delta)\tilde{g}(x,\alpha \mid x_0) = 0, \quad x \in \Omega \tag{18a}$$

$$\left(D\frac{\partial}{\partial n_x} + \kappa(x)\right)\tilde{g}(x,\alpha \mid x_0) = \tilde{j}_\infty(x,\alpha \mid x_0), \quad x \in \partial\Omega, \tag{18b}$$

where $\tilde{j}_0$ is the Laplace transform of the diffusive flux density at time t on a point of the perfectly reactive surface.

Details of spectral representation

Consider the Dirichlet BVP

$$(\alpha - D\Delta)u(x,\alpha) = 0, \quad x \in \Omega \tag{19a}$$

$$u(x,\alpha) = f(x,\alpha), \quad x \in \partial\Omega. \tag{19b}$$

We define the Dirichlet-to-Neumann (DtN) $\mathcal{M}_\alpha$ as

$$[\mathcal{M}_\alpha f](x) = \left(\frac{\partial u}{\partial n}\right)_{x\in\partial\Omega}, \tag{20}$$

which determines the flux produced at the boundary by u (Neumann BC) given boundary value f. To this end, on the boundary, we may write the boundary conditions for the unknown contribution to the propagator Eq. (18a) as (with $\mathcal{Q} = \kappa(y)/D$)

$$(\mathcal{M}_\alpha + \mathcal{Q})\,\tilde{g}(y,\alpha \mid x_0) = \frac{1}{D}\tilde{j}_\infty(y,\alpha \mid x_0), \quad y \in \partial\Omega. \tag{21}$$

Details of spectral representation

Formally, we may invert Eq. (21),

$$\tilde{g}(y, \alpha \mid x_0) = (\mathcal{M}_\alpha + \mathcal{Q})^{-1} \frac{\tilde{j}_\infty(y, \alpha \mid x_0)}{D}. \tag{22}$$

This is now the form of a Dirichlet problem, allowing us to write the bulk solution

$$\tilde{g}(x, \alpha \mid x_0) = \int_{\partial\Omega} d^{n-1}y \, \tilde{j}_\infty(y, \alpha \mid x)\tilde{g}(y, \alpha \mid x_0), \quad x \in \Omega. \tag{23}$$

If we let the surface propagation operator be written $\mathcal{O} = (\mathcal{M}_\alpha + \mathcal{Q})^{-1}$,

$$\tilde{G}(x, \alpha \mid x_0) = \tilde{G}_\infty(x, \alpha \mid x_0) + \int_{\partial\Omega} d^{n-1}y \, \tilde{j}_\infty(y, \alpha \mid x) \frac{\mathcal{O}}{D} \tilde{j}_\infty(y, \alpha \mid x_0). \tag{24}$$

Details of spectral representation

The identity $\tilde{j}_\infty(y,\alpha \mid y_0) = \delta(y - y_0)$ for $y, y_0 \in \partial\Omega$ implies that the propagator between two surface points

$$D(\mathcal{M}_\alpha + \mathcal{Q})\tilde{G}(y,\alpha \mid y_0) = \delta(y - y_0). \tag{25}$$

Now we may express the bulk propagator as

$$\begin{aligned}\tilde{G}(x,\alpha \mid x_0) = \tilde{G}_\infty(x,\alpha \mid x_0) + \\ \int_{\partial\Omega} dy_1\, dy_2\, \tilde{j}_\infty(y_2,\alpha \mid x)\frac{\tilde{G}(y_2,\alpha \mid y_1)}{D}\tilde{j}_\infty(y_1,\alpha \mid x_0).\end{aligned} \tag{26}$$

In braket notation, eigenvalues of the DtN operator satisfy the eigenequation

$$\mathcal{M}_\alpha \left| v_n^\alpha(x) \right\rangle = \mu_n^\alpha \left| v_n^\alpha(x) \right\rangle . \tag{27}$$

Details of spectral representation

Provided $\partial\Omega$ is bounded, the eigenvalues are non-negative and the eigenfunctions form a complete, orthonormal basis on the boundary. We may hence resolve the identity and insert it into Eq. (26):

$$\tilde{G}(x,\alpha \mid x_0) = \tilde{G}_\infty(x,\alpha \mid x_0) \\ + \frac{1}{D}\sum_{n,m}\int_{\partial\Omega} dy_1\, dy_2\, \tilde{j}_\infty \left|v_n^{(\alpha)}(s)\right\rangle \left\langle v_n^{(\alpha)}(s)\middle|\mathcal{O}\middle|v_m^{(\alpha)}(s')\right\rangle \left\langle v_m^{(\alpha)}(s')\right| \tilde{j}_\infty \tag{28}$$

If the permissivity is taken to be constant, the matrix is diagonal

$$\left\langle v_n^{(\alpha)}(s)\middle|(\mathcal{M}_\alpha + q)^{-1}\middle|v_m^{(\alpha)}(s')\right\rangle = \left(\mu_n^{(\alpha)} + q\right)^{-1} \delta_{n,m}. \tag{29}$$

Details of the spectral representation

Inserting the element into the propagator,

$$\tilde{G}(x,\alpha \mid x_0) = \tilde{G}_\infty(x,\alpha \mid x_0) + \frac{1}{D}\sum_n \frac{[V_n^{(\alpha)}(x_0)]^* V_n^{(\alpha)}(x)}{\mu_n^{(\alpha)} + q}, \quad (30)$$

where we have defined the projections

$$V_n^{(\alpha)}(x) = \int_{\partial\Omega} d^{n-1}y \, v_n^{(\alpha)}(y)\tilde{j}_\infty(y,\alpha \mid x). \quad (31)$$

Bessel ratio asymptotic form

We begin with the standard power series

$$I_n(z) = \sum_{k=0}^{\infty} \frac{1}{k!\Gamma(n+k+1)} \left(\frac{z}{2}\right)^{n+2k}$$
$$= \frac{1}{n!} \left(\frac{z}{2}^{n}\right)^{n} \left[1 + \frac{z^2}{4(n+1)} + \frac{z^4}{32(n+1)(n+2)} + \mathcal{O}(z^6)\right].$$

Taking the logarithm, performing an expansion $\log(1+x) \sim x - \frac{x^2}{2} + \frac{x^3}{3} + \mathcal{O}(x^4)$, and differentiating,

$$\frac{I_n'(z)}{I_n(z)} = \frac{n}{z} + \frac{z}{2(n+1)} - \frac{z^3}{8(n+1)^2(n+2)} + \mathcal{O}(z^5) \qquad (32)$$

The absorbing diffusive propagator

The absorbing propagator $G_\infty(x, t \mid x_0)$ makes its appearance in many of the equations for such semi-permeable systems to account for the probability of a walker never interacting with the boundary. Determining such a G is a relatively simple task in PDEs,

$$\begin{aligned}\frac{\partial G_\infty(x, t \mid x_0)}{\partial t} &= D\Delta G_\infty(x, t \mid x_0), \quad x \in \Omega \\ G_\infty(x, t = 0 \mid x_0) &= \delta(x - x_0) \\ G_\infty(x, t \mid x_0) &= 0, \quad x \in \partial\Omega.\end{aligned}$$

We perform a separation of variables $G_\infty = TP\Phi$ to find the equivalent differential equation problems. If we choose $r_0 = 0$, the initial conditions become $\delta(r)/2\pi r$ which is independent of θ and so we can neglect this term:

$$\dot{T} = -D\lambda T, \quad \rho\frac{d}{d\rho}\left(\rho\frac{dP}{d\rho}\right) + \lambda\rho^2 P = 0$$

The absorbing diffusive propagator

The first equation is trivially exponential while the second is a Bessel equation with $\alpha = 0$. Enforcing the Dirichlet condition on the eigenfunctions ensures that $\lambda = j_n^2$ where j_n is the nth solution of J_0. Summing over the eigenfunctions,

$$G_\infty(r, t \mid r_0 = 0) = \sum_n c_n J_0\left(\frac{r j_n}{R}\right) e^{-D j_n^2 t/R}.$$

The coefficients are uniquely determined via the orthogonality relation for Bessel functions with Dirichlet BC,

$$\int_0^R x J_\alpha\left(\frac{x\mu_k}{R}\right) J_\alpha\left(\frac{x\mu_n}{R}\right)\, dx = \frac{R^2}{2}[J_{\nu+1}(\mu_n)]^2 \delta_{k,n}$$

The absorbing diffusive propagator

Multiplying both sides of the propagator by $rJ_0(rj_k/R)$ and integrating from 0 to R, we find $c_n = 1/\pi R^2[J_1(j_n)]^2$ and so

$$G_\infty(r, t \mid r_0 = 0) = \frac{1}{\pi R^2} \sum_n \frac{J_0\left(rj_n/R\right)}{[J_1(j_n)]^2} e^{-Dj_n^2 t/R}. \qquad (33)$$

Since $Dj_n/R > 0$, the Laplace transform converges,

$$\tilde{G}_\infty(r, \alpha \mid r_0 = 0) = \frac{1}{\pi R^2} \sum_n \frac{1}{\alpha + Dj_n^2/R} \frac{J_0\left(rj_n/R\right)}{[J_1(j_n)]^2}. \qquad (34)$$