Field Theory Approach to Diffusion-Limited Reactions:

1. Models and Mappings

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Field Theory Approach to Diffusion-Limited Reactions

1. Models and Mappings

How to turn stochastic particle models into a field theory, with no phenomenology.

2. Single-Species Annihilation

Field theoretic renormalization group calculation for $A+A \rightarrow 0$ reaction in gory detail.

3. Applications

Higher order reactions, disorder, Lévy flights, two-species reactions, coupled reactions.

4. Active to Absorbing State Transitions

Directed percolation, branching and annihilating random walks, and all that.

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Diffusion-Limited Reactions

One or more species of particles undergoing random walks on a lattice, with a reactions occurring for particles on the same lattice site



Example: for $A + A \rightarrow 0$ the density of particles decays as

$$\rho(t) = \begin{cases} Ct^{-1} & \text{for } d > 2\\ \tilde{A}\ln t/Dt & \text{for } d = 2\\ A_d(Dt)^{-d/2} & \text{for } d < 2 \end{cases}$$

where A_d and \tilde{A} are universal numbers!

Field Theory Approach to Diffusion-Limited Reactions: 1. Models and Mappings

Master Equation for Lattice Models

Doi Representation

Mapping to Field Theory

Stochastic Classical Particles on a Lattice

Consider a set of lattice sites labeled $i = 1, 2, 3, \ldots$, and each site is occupied by n_1, n_2, n_3, \ldots particles.



Define

- $\alpha = a$ particular state, i.e., $\alpha = \{n_1, n_2, n_3, \dots\}$
- $P(\alpha, t) =$ the probability of obtaining state α at time t.

Probability Master Equation

Dynamical processes processes (hops, reactions, decays) will cause a change of state from α to β .

 $w_{\alpha \rightarrow \beta} =$ rate of transition from α to β , defines dynamics

Master Equation

$$\frac{d}{dt}P(\alpha,t) = \sum_{\beta} \left[\underbrace{w_{\beta \to \alpha} P(\beta,t)}_{\text{flow into } \alpha} - \underbrace{w_{\alpha \to \beta} P(\alpha,t)}_{\text{flow out of } \alpha} \right]$$

 $\blacktriangleright \sum_{\alpha} P(\alpha,t) = 1$ preserved by the master equation

▶ Initial conditions $P(\alpha, 0)$ need to be specified

Master Equation for $A \rightarrow 0$ Decay

Consider a single lattice site that contains some number of identical particles. These particles decay at rate $\lambda.$

The rate for a transition from n to m particles is

$$w_{n \to m} = egin{cases} 0 & \mbox{for } m
eq n-1 \ n\lambda & \mbox{for } m = n-1 \end{cases}$$

and the master equation is

$$\frac{d}{dt}P(n,t) = \lambda \Big[(n+1)P(n+1,t) - n P(n,t) \Big]$$

Wait! That doesn't look like exponential decay

Master Equation to Differential Equation

Let $\rho(t)=\langle n\rangle=\sum_n n\,P(n,t)$ be the average number of particles at time t. Then

$$\begin{split} \dot{\rho} &= \sum_{n} n\dot{P}(n,t) = \sum_{n} n \bigg[\lambda(n+1)P(n+1,t) - \lambda nP(n,t) \bigg] \\ &= \lambda \sum_{n} n(n+1)P(n+1,t) - \lambda \sum_{n} n^{2}P(n,t) \\ &= \lambda \sum_{m} (m-1)mP(m,t) - \lambda \sum_{n} n^{2}P(n,t) \\ &= -\lambda \sum_{m} mP(m,t) \\ &= -\lambda \rho \end{split}$$

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$A + A \rightarrow 0$ Reaction

Again, consider a single lattice site, with the rule that a pair of particles may annihilate each other. The rates are

$$w_{n \to m} = \begin{cases} 0 & \text{for } m \neq n-2\\ n(n-1)\lambda & \text{for } m = n-2 \end{cases}$$

and the master equation is

$$\frac{d}{dt}P(n,t) = \lambda \Big[(n+2)(n+1)P(n+2,t) - n(n-1)P(n,t) \Big]$$

Hop



Diffusion

Consider a one-dimensional chain of lattice sites i = 1, 2, ... and let all particles hop left or right with rate Γ . The master equation is



$$\frac{d}{dt}P(\alpha,t) = \Gamma \sum_{\langle ij \rangle} \left[(n_i+1)P(n_i+1,n_j-1,\ldots,t) - n_i P(\alpha,t) \right]$$
$$(n_i+1)P(n_i-1,n_j+1,\ldots,t) - n_i P(\alpha,t)$$

Define $\rho(x,t) = \sum_{\alpha} n_i P(\alpha,t)$ where $x = i\Delta x$.

For small Δx this becomes the diffusion equation:

$$\frac{\partial \rho}{\partial t} = D \frac{\partial^2 \rho}{\partial x^2}$$
 $D = \Gamma \Delta x^2 = \text{diffusion constant.}$

Master Equation for $A + A \rightarrow 0$ Diffusion-Limited Reaction

$$\begin{split} \frac{d}{dt} P(\{n\},t) &= \\ & \frac{D}{\Delta x^2} \sum_{\langle ij \rangle} \Big[(n_i+1) P(\dots,n_i+1,n_j-1,\dots,t) - n_i P(\{n\},t) \\ & + (n_j+1) P(\dots,n_i-1,n_j+1,\dots,t) - n_j P(\{n\},t) \Big] \\ & + \lambda \sum_i \Big[(n_i+2)(n_i+1) P(\dots,n_i+2,\dots,t) \\ & - n_i(n_i-1) P(\{n\},t) \Big] \\ & \text{with } P(\{n\},0) = \prod_i \frac{n_0^{n_i} e^{-n_0}}{n_i!} \text{ for random initial conditions.} \\ & \textbf{Yuck!} \end{split}$$

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Field Theory Approach to Diffusion-Limited Reactions:

1. Models and Mappings

Master Equation for Lattice Models

Doi Representation

Mapping to Field Theory

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Doi Representation, part II

We can pack the probability P into a Fock space state:

$$|\phi(t)\rangle = \sum_{\{n\}} P(\{n\}, t) |\{n\}$$

and re-write the master equation in Schrödinger-like form:

$$\frac{d}{dt}|\phi(t)\rangle = -\hat{H}|\phi(t)\rangle$$

Why do this? Because it is a simpler description of the dynamics. For $A+A\to 0$ diffusion-limited reaction we get

$$\hat{H} = \Gamma \sum_{\langle ij \rangle} (a_i^{\dagger} - a_j^{\dagger})(a_i - a_j) - \lambda \sum_i (1 - a_i^{\dagger 2})a_i^2$$

and formal solution $|\phi(t)\rangle = \exp(-\hat{H}t)|\phi(0)\rangle$.

Doi Occupation Number Representation (Fock Space)

For a single lattice site:

- ▶ Introduce a creation operator \hat{a}^{\dagger} and annihilation operator \hat{a} , with commutator $[\hat{a}, \hat{a}^{\dagger}] = 1$.
- \blacktriangleright Represent the state of zero particles by $|0\rangle,$ defined via $\hat{a}|0\rangle=0.$
- Represent a state of n particles by $|n\rangle = \hat{a}^{\dagger n}|0\rangle$. (Note: normalization differs from usual QM.)
- ► For this state

$$\hat{a}^{\dagger} \left| n \right\rangle = \left| n + 1 \right\rangle \qquad \hat{a} \left| n \right\rangle = n \left| n - 1 \right\rangle \qquad \hat{a}^{\dagger} \hat{a} \left| n \right\rangle = n \left| n \right\rangle$$

For multiple lattice sites: introduce a pair \hat{a}_i , \hat{a}_i^{\dagger} at each site

$$\{n\} = (n_1, n_2, \dots) \quad \Leftrightarrow \quad |\{n\}\rangle = \prod_i \hat{a}_i^{\dagger n_i} |0\rangle$$

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$A+A \rightarrow 0$ on a Single Site

Master equation:

$$\frac{d}{dt}P(n,t) = \lambda \Big[(n+2)(n+1)P(n+2,t) - n(n-1)P(n,t) \Big]$$

Multiply by $|n\rangle$ and sum over $n{:}$

$$\begin{aligned} \frac{d}{dt} |\phi(t)\rangle &= \lambda \sum_{n} P(n+2,t) \left(n+2\right)(n+1) |n\rangle - \lambda \sum_{n} P(n,t) n(n-1) |n\rangle \\ &= \lambda \sum_{n} P(n+2,t) \hat{a}^{2} |n+2\rangle - \lambda \sum_{n} P(n,t) \hat{a}^{\dagger 2} \hat{a}^{2} |n\rangle \\ &= \lambda (\hat{a}^{2} - \hat{a}^{\dagger 2} a^{2}) \sum_{n} P(n,t) |n\rangle \\ &= \lambda (1 - \hat{a}^{\dagger 2}) \hat{a}^{2} |\phi(t)\rangle = -\hat{H} |\phi(t)\rangle \end{aligned}$$

Hop from Site 1 to Site 2

Master Equation

$$\frac{d}{dt}P(n_1, n_2, t) = \Gamma\Big[(n_1+1)P(n_1+1, n_2-1, t) - n_1P(n_1, n_2, t)\Big]$$

Multiply by $|n_1, n_2\rangle$ and sum over n_1 and n_2 :

$$\begin{aligned} \frac{d}{dt} |\phi(t)\rangle &= \Gamma \sum_{n_1, n_2} P(n_1 + 1, n_2 - 1, t) (n_1 + 1) |n_1, n_2\rangle \\ &- \Gamma \sum_{n_1, n_2} P(n_1, n_2, t) n_1 |n_1, n_2\rangle \\ &= \Gamma \sum_{n_1, n_2} P(n_1 + 1, n_2 - 1, t) \hat{a}_2^{\dagger} \hat{a}_1 |n_1 + 1, n_2 - 1\rangle \\ &- \Gamma \sum_{n_1, n_2} P(n_1, n_2, t) \hat{a}_1^{\dagger} \hat{a}_1 |n_1, n_2\rangle \\ &= \Gamma(\hat{a}_2^{\dagger} - \hat{a}_1^{\dagger}) \hat{a}_1 |\phi(t)\rangle \end{aligned}$$

Diffusion

► Hop from site 1 to site 2:

$$\hat{H}_{1\to 2} = \Gamma(\hat{a}_1^{\dagger} - \hat{a}_2^{\dagger})a_1$$

► Allow for the reverse hop with the same rate:

$$\hat{H}_{1\leftrightarrow 2} = \Gamma(\hat{a}_1^{\dagger} - \hat{a}_2^{\dagger})(\hat{a}_1 - \hat{a}_2)$$

► For hops between all neighboring lattice sites:

$$\hat{H}_D = \frac{D}{(\Delta x)^2} \sum_{\langle ij \rangle} (\hat{a}_i^{\dagger} - \hat{a}_j^{\dagger}) (\hat{a}_i - \hat{a}_j)$$

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Multiple Species

With two species reactions, such as $A+B\to 0$ reaction, the master equation is defined in terms of A-particle and B-particle occupation numbers

$$P(\{m\},\{n\},t)$$

For the Doi representation, we introduce distinct creation and annihilation operators for each species: \hat{a}_i , \hat{a}_i^{\dagger} , \hat{b}_i , and \hat{b}_i^{\dagger} , and define state

$$|\phi(t)\rangle = \sum_{\{m\},\{n\}} P(\{m\},\{n\},t) \prod_{i} \hat{a}_{i}^{\dagger m_{i}} \hat{b}_{i}^{\dagger n_{i}}|0\rangle$$

Doi Hamiltonians

Each process contributes two terms to \hat{H} , of the form

$$(\mathsf{rate}) \Big[(\mathsf{reactants}) - (\mathsf{reaction}) \Big]$$

(reactants) = creation and annihilation operator for each reactant, normal ordered

(reaction) = annihilation operator for each reactant, creation operator for each product, normal ordered

Examples:

$$\begin{array}{ll} A + A \to 0 & \lambda [\hat{a}^{\dagger 2} \hat{a}^2 - \hat{a}^2] & A + A \to A & \lambda [\hat{a}^{\dagger 2} \hat{a}^2 - \hat{a}^{\dagger} \hat{a}^2] \\ \\ A \to A + A & \lambda [\hat{a}^{\dagger} \hat{a} - \hat{a}^{\dagger 2} \hat{a}] & A + B \to C & \lambda [\hat{a}^{\dagger} \hat{b}^{\dagger} \hat{a} \hat{b} - \hat{c}^{\dagger} \hat{a} \hat{b}] \\ \\ \\ \text{Hop } 1 \to 2 & \Gamma [\hat{a}_1^{\dagger} a_1 - \hat{a}_2^{\dagger} a_1] \end{array}$$

Observables

For our classical particle system, averages are given by

$$\langle A(t)\rangle = \sum_{\{n\}} A(\{n\}) P(\{n\},t)$$

To map this to the Doi representation, we need a projection state $\langle \mathcal{P}|=\langle 0|e^{\sum_i \hat{a}_i},$ which has properties

$$\langle \mathcal{P}|0 \rangle = 1 \qquad \langle \mathcal{P}|\hat{a}_i^{\dagger} = \langle \mathcal{P}| \quad \Rightarrow \quad \langle \mathcal{P}|n \rangle = 1$$

Then, for the operator $\hat{A} = A(\{n_i \rightarrow \hat{a}_i^{\dagger} \hat{a}_i\})$ we get

$$\langle A(t) \rangle = \langle \mathcal{P} | \hat{A} | \phi(t) \rangle = \langle \mathcal{P} | \hat{A} e^{-\hat{H}t} | \phi(0) \rangle$$

Note: for Poisson initial conditions, $|\phi(0)\rangle = \prod_i e^{-n_0 + n_0 \hat{a}_i^{\dagger}} |0\rangle$

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Probability Conservation

Check Normalization

For some initial state $P(\{n\}, 0)$ we average the identity operator:

$$\langle \mathcal{P}|\hat{1}|\phi(0)\rangle = \langle \mathcal{P}|\sum_{\{n\}} P(\{n\},0)|\{n\}\rangle = \sum_{\{n\}} P(\{n\},0) = 1 \checkmark$$

Check Probability Conservation

What is the condition on \hat{H} ? We need

$$1 = \langle \mathcal{P} | \hat{1}e^{-\hat{H}t} | \phi(0) \rangle = \langle \mathcal{P} | (1 - \hat{H}t + \frac{1}{2}t^2\hat{H}^2 - \dots) | \phi(0) \rangle$$

for all t, so we require $\langle \mathcal{P}|\hat{H}=0.$ Equivalently, $\hat{H}\rightarrow 0$ as $\hat{a}_i^\dagger\rightarrow 1.$

Note: \hat{H} need not be hermitian!

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Coherent States

The coherent state $|\phi\rangle=\exp(-\frac{1}{2}|\phi|^2+\phi a^\dagger)|0\rangle$ with complex ϕ has properties

$$\hat{a}|\phi
angle = \phi|\phi
angle, \qquad \langle \phi|\hat{a}^{\dagger} = \langle \phi|\phi^{\dagger}\rangle$$

and the overlap relation

$$\langle \phi_1 | \phi_2 \rangle = \exp(-\frac{1}{2} |\phi_1|^2 - \frac{1}{2} |\phi_2|^2 + \phi_1^* \phi_2)$$

Can construct a resolution of the identity operator

$$\hat{1} = \sum_{n} \frac{1}{n!} |n\rangle \langle n| = \sum_{m,n} \frac{1}{n!} |n\rangle \langle m| \delta_{mn} = \int \frac{d^2 \phi}{\pi} |\phi\rangle \langle \phi|$$

by use of

$$\delta_{mn} = \frac{1}{m!\pi} \int \phi^{*m} \phi^n e^{-|\phi|^2} d^2\phi$$

Coherent State Representation

Given $\hat{H}(\hat{a}_i^{\dagger},\hat{a}_i)$, take formal solution $|\phi(t)\rangle=e^{-\hat{H}t}|\phi(0)\rangle$ and divide time t into a number of small increments Δt via

$$e^{-\hat{H}t} = \exp(-\hat{H}\Delta t)^{t/\Delta t}$$

Insert $\hat{1} = \int \frac{d^2 \phi}{\pi} |\phi\rangle \langle \phi|$ between each successive time step:

$$\int \dots |\phi_{t+\Delta t}\rangle \langle \phi_{t+\Delta t}| e^{-\hat{H}\Delta t} |\phi_t\rangle \langle \phi_t| e^{-\hat{H}\Delta t} |\phi_{t-\Delta t}\rangle \langle \phi_{t-\Delta t}| \dots$$

Now focus on a single matrix element:

$$\begin{split} \langle \phi_t | e^{-\hat{H}\Delta t} | \phi_{t-\Delta t} \rangle &= e^{-H(\phi_t^*,\phi_{t-\Delta t})\Delta t} \langle \phi_t | \phi_{t-\Delta t} \rangle \\ &\simeq e^{-H(\phi_t^*,\phi_t)\Delta t} \ e^{\phi_t^*\phi_{t-\Delta t} - \frac{1}{2}|\phi_t|^2 - \frac{1}{2}|\phi_{t-\Delta t}|^2} \\ &\simeq e^{-H(\phi_t^*,\phi_t)\Delta t} \ e^{-\phi_t^*\partial_t\phi_t\Delta t} \end{split}$$

Path Integral

String together many time slices in the limit $\Delta t \rightarrow 0$ and we get

$$e^{-\hat{H}t} \to \int \mathcal{D}\phi^* \mathcal{D}\phi \exp\left(-\int_0^t dt' [\phi^*\partial_{t'}\phi + H(\phi^*,\phi)]\right)$$

where $\prod_{j} \left(\frac{d^2 \phi_j}{\pi} \right) \to \mathcal{D} \phi^* \mathcal{D} \phi$

Generalize to multiple lattice sites:

$$e^{-\hat{H}t} \to \int \prod_{j} (\mathcal{D}\phi_j^* \mathcal{D}\phi_j) e^{-S[\{\phi_j^*\}, \{\phi_j\}]}$$

with

$$S = \int_0^t \sum_j dt' \Big[\phi_j^* \partial_{t'} \phi_j + H(\{\phi_j^*\}, \{\phi_j\}) \Big]$$

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Path Integral Observables

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$$\langle A(t) \rangle = \langle \mathcal{P} | \hat{A} e^{-\hat{H}t} | \phi(0) \rangle = \mathcal{N}^{-1} \int \prod_{j} (\mathcal{D} \phi_{j}^{*} \mathcal{D} \phi_{j}) A(\phi(t)) e^{-S[\phi^{*},\phi]} d\phi(t)$$

with action \boldsymbol{S} given by

$$S = \sum_{i} \left\{ -\phi_{i}(t) + \int_{0}^{t} dt' \Big[\phi_{i}^{*} \partial_{t'} \phi_{i} + H(\{\phi_{i}^{*}\}, \{\phi_{i}\}) \Big] - n_{0} \phi_{i}^{*}(0) \right\}$$

Can eliminate projection state term by field shift $\phi^* \rightarrow 1 + \tilde{\phi}$:

$$\int_0^t (1+\tilde{\phi})\,\partial_{t'}\phi\,\,dt' = \phi(t) - \phi(0) + \int_0^t \tilde{\phi}\,\partial_t\phi\,\,dt'$$

which takes $H \to H(\{1+\tilde{\phi}_j\},\{\phi_j\})$

Diffusion

$$S_{D} = \int dt \left[\sum_{i} \tilde{\phi}_{i} \partial_{t} \phi_{i} + \frac{D}{\Delta x^{2}} \sum_{\langle ij \rangle} (\tilde{\phi}_{i} - \tilde{\phi}_{j})(\phi_{i} - \phi_{j}) \right] - \sum_{i} n_{0} \tilde{\phi}_{i}(0)$$
$$= \int dt \, d^{d}x \left[\tilde{\phi} \partial_{t} \phi + D \nabla \tilde{\phi} \cdot \nabla \phi - n_{0} \tilde{\phi} \, \delta(t) \right]$$
$$= \int dt \, d^{d}x \left[\tilde{\phi} (\partial_{t} - D \nabla^{2}) \phi - n_{0} \tilde{\phi} \, \delta(t) \right]$$

Action is linear in $\tilde{\phi}$. Extremum:

$$\frac{\delta S_D}{\delta \tilde{\phi}} = \partial_t \phi - D \nabla^2 \phi - n_0 \,\delta(t) = 0$$

is the plain old diffusion equation: $\partial_t \phi = D \nabla^2 \phi + n_0 \, \delta(t)$

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Diffusion-Limited $A + A \rightarrow 0$ Reaction

The reaction part of the hamiltonian is

$$H_{\rm reac} = -\lambda \sum_i (1 - \phi_i^{*2}) \phi_i^2 \to \int d^d x (2\lambda_0 \tilde{\phi} \phi^2 + \lambda_0 \tilde{\phi}^2 \phi^2$$

with the field shift $\phi^* \rightarrow 1 + \tilde{\phi}$ and $\lambda_0 = \lambda / \Delta x^d$.

Thus the complete $A + A \rightarrow 0$ action is

 $S = \int d^d x \, dt \left[\tilde{\phi} (\partial_t - D\nabla^2) \phi + 2\lambda_0 \tilde{\phi} \phi^2 + \lambda_0 \tilde{\phi}^2 \phi^2 - n_0 \tilde{\phi} \, \delta(t) \right]$

Now we're ready to do some calculations!

The $A + A \rightarrow 0$ Stochastic PDE

Can make the action linear in $\tilde{\phi}$ via an auxiliary field η :

$$e^{-\lambda_0 \tilde{\phi}^2 \phi^2} \propto \int d\eta \, \exp\left\{-\frac{1}{2}\eta^2 + i\eta \sqrt{2\lambda_0} \, \tilde{\phi} \, \phi\right\}$$

resulting in averages

$$\int \mathcal{D}\eta \, e^{-\eta^2/2} \int \mathcal{D}\phi \int \mathcal{D}\tilde{\phi} \, e^{-\int \tilde{\phi}(\partial_t - D\nabla^2)\phi + 2\lambda_0 \tilde{\phi}\phi^2 + i\eta\sqrt{2\lambda_0} \, \tilde{\phi}\phi}$$

The $\tilde{\phi}$ integration creates a δ -function that enforces

$$\partial_t \phi = D\nabla^2 \phi - 2\lambda_0 \phi^2 + i\sqrt{2\lambda_0} \phi \eta$$

A stochastic reaction-diffusion equation with multiplicative noise that is complex!?

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Summary and Observations

- Mapping to Doi representation simplifies the master equation by getting rid of pesky factors involving n.
- This Fock space description natural for identical particles acting independently, not restricted to quantum mechanics
- Fock space dynamics can be converted to a field theory without resorting to Langevin-type phenomenology
- Technique can easily include multiple species, long-range hops, birth/death processes, convected fields
- Mechanical forces not so easily included. *Ĥ* is not an energy but rather rates.
- SPDE's are fraught with peril!

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Exercises

- 1. Diffusion equation:
 - (a) Show that the diffusion master equation is equivalent to

$$\frac{d}{dt}\langle n_i\rangle = \frac{D}{\Delta x^2} \sum_j (\langle n_j\rangle - \langle n_i\rangle)$$

where the sum on j runs over nearest neighbors of site i.

- (b) Show that $\rho(x,t) = \langle n_i \rangle$ with $x = i \Delta x$ satisfies $\partial_t \rho = D \partial_x^2 \rho$ as $\Delta x \to 0$.
- (c) Generalize the result to a d-dimensional hypercubic lattice.
- 2. Consider the decay $A \to 0$ on a single lattice site. Map the problem from the master equation to the Doi hamiltonian to the shifted field theory. Show that the $\mathcal{D}\tilde{\phi}$ integration yields the expected result

$$\partial_t \phi = -\lambda \phi + n_0 \,\delta(t)$$

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Exercises

3. Write down the Doi hamiltonian for the reversible reaction $\ell A + mB \rightleftharpoons nC$, with rates λ for the forward reaction and μ for the reverse reaction. Here ℓA , for example, means ℓA particles are required for the reaction.

4. Coherent states:

- (a) Determine the coefficients C_n in the expansion of the coherent state $|\phi\rangle=\sum_n C_n|n\rangle$
- (b) Confirm the identity

$$\delta_{mn} = \frac{1}{m!\pi} \int \phi^{*m} \phi^n e^{-|\phi|^2} d^2 \phi$$

(c) Use the results from (a) and (b) confirm $\hat{1} = \int \frac{d^2\phi}{\pi} |\phi\rangle\langle\phi|$. Note that $d^2\phi = d(\text{Re}\phi) d(\text{Im}\phi)$.