Landscapes of Non-gradient Dynamics Without Detailed Balance: Stable Limit Cycles and Multiple Attractors

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Abstract

Landscape is one of the key notions in literature on biological processes and physics of complex systems with both deterministic and stochastic dynamics. The large deviation theory (LDT) provides a possible mathematical basis for the scientists’ intuition. In terms of Freidlin-Wentzell’s LDT, we discuss explicitly two issues in singularly perturbed stationary diffusion processes arisen from nonlinear differential equations: (1) For a process whose corresponding ordinary differential equation has a stable limit cycle, the stationary solution exhibits a clear separation of time scales: an exponential terms and an algebraic prefactor. The large deviation rate function attains its minimum zero on the entire stable limit cycle, while the leading term of the prefactor is inversely proportional to the velocity of the non-uniform periodic oscillation on the cycle. (2) For dynamics with multiple stable fixed points and saddles, there is in general a breakdown of detailed balance among the corresponding attractors. Two landscapes, a local and a global, arise in LDT, and a Markov jumping process with cycle flux emerges in the low-noise limit. A local landscape is pertinent to the transition rates between neighboring stable fixed points; and the global landscape defines a nonequilibrium steady state. There would be nondifferentiable points in the latter for a stationary dynamics with cycle flux. LDT serving as the mathematical foundation for emergent landscapes deserves further investigations.

Keywords: large deviations; limit cycle; multistability; nonequilibrium steady state; singularly perturbed diffusion process.
Stochastic nonlinear approaches to dynamics has attracted great interests from physicists, biologists, and mathematicians in current research. More than 70 years ago, Kramers has developed a diffusion model characterizing the molecular dynamics along a reaction coordinates, via a barrier crossing mechanism, and calculated the reaction rate for an emergent chemical reaction. The work explained the celebrated Arrhenius relation as well as Eyring’s concept of “transition state”. Kramers’ theory, however, is only valid for stochastic dynamics in closed systems with detailed balance (i.e., a gradient flow), where the energy landscape gives the equilibrium stationary distribution via Boltzmann’s law. It is not suitable for models of open systems. Limit cycle oscillation is one of the most important emergent behaviors of nonlinear, non-gradient systems. The large deviation theory from probability naturally provides a basis for the concept of a “landscape” in a deterministic nonlinear, non-gradient dynamics. In the present study, we initiate a line of studies on the dynamics of and emergent landscape in open systems. In particular, using singularly perturbed diffusion on a circle as a model, we study systems with stable limit cycle as well as systems with multiple attractors with nonzero flux. A seeming paradox concerning emergent landscape for limit cycle is resolved; a local theory for transitions between two adjacent attractors, à la Kramers, is discussed; and a “λ-surgery” to obtain nonequilibrium steady state (NESS) landscape for multiple attractors is described.

1 Introduction

Stochastic nonlinear dynamics (SND) of biochemical reaction systems at the cellular and subcellular level has received much interests in recent years from applied mathematicians, physicists, as well as biologists [27]. In terms of stochastic processes, the main mathematical approaches to biochemical SND are either diffusion processes i.e., the chemical Langevin equation, or the chemical master equation [40, 1, 15] which characterizes the evolution of probability distribution for a Markov jump process that can be simulated by the method of Gillespie algorithm. For both stochastic models, their infinite large system (macroscopic) limit is a system of nonlinear ordinary differential equations (ODE) based on the Law of Mass Action [19, 7]. These mathematical models have provided a unique opportunity for comparative studies of corresponding nonlinear dynamics in small and in large systems.

In quantitative biology and in statistical physics, there is an emerging notion of “landscape” for dynamics [8], as both a metaphor and as an analytical device [41, 45, 13]. Landscape for a gradient system is of course natural, and the well-known Kramers’ rate theory directly follows [13] (Fig. 1). For a non-gradient system, a Lyapunov function [25], if exists, can still be visualized as a landscape for the dynamics. The real question is whether there always exists such a landscape function for non-gradient system.
and whether there is a corresponding Kramers’-like rate formula for the inter-attractoral transition rates. For bistable systems, the answer to this question is yes \cite{9, 12, 13, 28}. The rate formula is exactly the same as the classical Kramers’ formula (Fig. 1). But how could one perceive a “landscape” for a periodically oscillatory system? Furthermore, is there any difference between multistable and bistable systems? These questions are the motivations of the present paper.

The case of a system with a limit cycle is in defiance of the intuition \cite{45}, and there are other serious, but subtle arguments against the general notion of landscape for systems with limit cycles. Noting that a same landscape is used for a deterministic dynamics as well as the stationary probability of its stochastic counterpart, one objection can be stated as follows: Let $\phi(x)$ be a landscape of a system with limit cycle $\Gamma$. Then $\phi(x)$ has to be a constant on $\Gamma$. However, since the landscape is also expected to represent the probability of a stochastic system: Lower $\phi$ corresponds to higher probability. Combining the two lines of reasoning, one arrives at equal probability along $\Gamma$. Now according to the ergodic theory, equal probability on $\Gamma$ implies uniform velocity on the limit cycle. This suggests that only uniform rotation is compatible with the notion of a landscape \cite{45}.

One of the aims of the present paper is to give an explicit resolution to this seeming paradox. The analysis reveals a separation of time scales for stochastic dynamics and its deterministic limit. Indeed around a limit cycle $\Gamma$, the probability $u_\epsilon(x) \simeq C_0(x)e^{-\phi(x)/\epsilon}$ in which $\phi(x) = 0$ along the $\Gamma$. The dynamics on the limiting set $\Gamma$, therefore, is determined by $C_0(x)$.

For presenting the results, we choose to be insightful rather than thorough and rigorous. Hence we shall only discuss the problem in terms of singularly perturbed diffusion processes. The insights we obtain, however, are qualitatively applicable also to other systems, even though technically they might be much more difficult to handle. More precisely, we would like to carry out an analysis of the singularly perturbed stationary diffusion equation in the form

$$\nabla \cdot (\epsilon \nabla u_\epsilon(x) - u_\epsilon(x)F(x)) = 0, \quad (x \in \mathbb{R}^N). \tag{1}$$

In Eq. (1) $\epsilon$ is a small positive parameter.

For a discussion of limit cycles in the Chemical Master Equation, see \cite{30, 38}, and the diffusion approximation in general, see \cite{35}. The singularly perturbed 2nd order linear elliptic equation is a well-studied problem in mathematics. However, there are still several important issues remaining unclear, even for the one-dimensional circle $S^1$. Here we wish to further explicitly illustrate some of them in connection to the case of a stable limit cycle or multiple attractors without detailed balance, sometime using examples. There would be two kinds of landscapes in the case of multiple attractors: one is for the Kramers’-like rate formula and the other is for the global stationary distribution. In a broad sense, both problems are very closely related to statistical dynamics and thermodynamics of nonequilibrium steady state \cite{43, 14}. 
2 General stationary solution and WKB approximation

We assume the function \( u_\epsilon(x) \) in Eq. (1) to be \( L^1 \) integrable throughout \( \mathbb{R}^N \). And we further assume that \( F(x) \) is sufficiently well behaved and that a stationary probability density exists. See [29] for appropriate conditions. It is understood that for Eq. (1), in addition to the stationary probability \( u_\epsilon(x) \), the system also possesses a non-trivial flux vector \( J \):

\[
J = u_\epsilon(x)F(x) - \epsilon \nabla u_\epsilon(x),
\]

satisfying \( \nabla \cdot J = 0 \). It can be shown that \( J = 0 \) if and only if \( F = -\nabla U \) is a gradient system [29], which is called detailed balance or equilibrium. In that case, there will be no limit cycle and in fact \( u_\epsilon(x) = A \exp(-U(x)/\epsilon) \), where \( A \) is a normalization constant.

Now in terms of the small parameter \( \epsilon \), let us first assume that the limit

\[
\lim_{\epsilon \to 0} \epsilon \ln u_\epsilon(x) = -\phi(x) \leq 0
\]

exists. Note that the limit \( \phi(x) \) has to be zero on the entire set where \( u_\epsilon(x) \) has a nontrivial limit. In probability theory, \( \phi(x) \) is known as the large deviation rate function [9, 4, 32]. Furthermore, we also assume that the solution has the general form

\[
u_\epsilon(x) = C_\epsilon(x)e^{-\phi(x)/\epsilon},
\]

and that there exists a positive constant \( \nu \) such that

\[
\lim_{\epsilon \to 0} \epsilon^\nu C_\epsilon(x) = C_0(x), \quad 0 < C_0(x) < +\infty.
\]

Therefore, we have the fundamental asymptotic representation

\[
u_\epsilon(x) = \epsilon^{-\nu} (C_0(x) + \epsilon C_1(x) + \cdots) e^{-\phi(x)/\epsilon},
\]

as in WKB theory [2, 17]. Substituting Eq. (6) into Eq. (1), we formally have

\[
\frac{1}{\epsilon} C_0 (\nabla \phi + F) \cdot \nabla \phi
\]

\[
- (C_0 \nabla^2 \phi + 2\nabla C_0 \cdot \nabla \phi + \nabla C_0 \cdot F + C_0 \nabla \cdot F - C_1 (\nabla \phi)^2 - C_1 F \cdot \nabla \phi)
\]

\[
+ \epsilon (\nabla^2 C_0 - C_1 \nabla^2 \phi - 2\nabla C_1 \cdot \nabla \phi - \nabla C_1 \cdot F - C_1 \nabla \cdot F) + \cdots = 0.
\]

The leading order term in (7) yields

\[
C_0 (\nabla \phi + F) \cdot \nabla \phi = 0.
\]

Since \( C_0 \neq 0 \), this means

\[
F \cdot \nabla \phi = -(\nabla \phi)^2 \leq 0.
\]
Eq. (9) shows that for the ordinary differential equation
\[
\frac{dx}{dt} = F(x),
\tag{10}
\]
the function \(\phi(x)\) has the Lyapunov property:
\[
\frac{d\phi(x(t))}{dt} = \nabla \phi \cdot \frac{dx}{dt} = \nabla \phi \cdot F \leq 0.
\tag{11}
\]
This result was contained in \[22, 23, 32\] and first explicitly reported in \[10\] for chemical master equation.

The second-order term in Eq. (7) yields
\[
\nabla C_0 \cdot (2\nabla \phi + F) + C_0 (\nabla^2 \phi + \nabla \cdot F) = 0,
\tag{12}
\]
from which \(C_0(x)\) can be obtained. For example, if \(C_0(x)\) is a constant, independent of \(x\), then Eq. \[12\] implies that \(\nabla \phi + F = \gamma\) is a divergence-free vector field. Combining this with Eq. \[5\], we have
\[
F = -\nabla \phi + \gamma, \quad \nabla \phi \cdot \gamma = 0.
\tag{13}
\]
The vector field \(F\) thereby has an orthogonal Hodge decomposition \[21\]. In other words, if one assumes that the solution to Eq. \[1\] is in the form of \(e^{-w(x)/\epsilon}\), then \(w(x)\) has to be a function of \(\epsilon\) except when \(F\) has orthogonal Hodge decomposition (This is indeed the case for Boltzmann's law with \(\gamma = 0\)). The leading order expansion is also the starting point of several investigations carried out by Graham and coworkers \[16\]. By requiring an orthogonality condition between the gradient and the rotational parts of the decomposition of \(F(x)\), the existence of a smooth \(\phi(x)\) is related to the complete integrability of certain Hamiltonian system. The Lyapunov property of \(\phi(x)\) in Eq. \[9\], however, is more general.

3 The theory of diffusion on a circle

We now give a thorough treatment of the dynamics on the circle, which includes either a limit cycle or multiple fixed points. We consider the singularly perturbed, stationary diffusion equation on the circle
\[
\epsilon \frac{d^2 u}{d\theta^2} + \frac{d}{d\theta} \left\{ (U'(\theta) - f) u \right\} = 0,
\tag{14}
\]
in which \(U(\theta)\) is a given smooth periodic function, \(U(0) = U(1)\), with periodic boundary condition \(u(0) = u(1)\) \[24\], and \(f\) is a given constant. The general solution is
\[
u(\theta) = A_\epsilon \left( \int_0^{1+\theta} e^{\frac{U(z) - fz}{\epsilon}} dz \right) e^{-\frac{U(\theta) - f\theta}{\epsilon}},
\tag{15}
\]
in which $A_\epsilon$ is a normalization factor. An important quantity associated with the stationary process is the cycle flux

$$J = \epsilon A_\epsilon \left( 1 - e^{-f/\epsilon} \right),$$

(16)

which generalizes the rotation number for nonlinear dynamical systems on the circle [39]. When $f = 0$, the flux $J = 0$. This is the case of symmetric diffusion process in the theory of probability [29].

We note that in the limit of $\epsilon \to 0$, by Laplace’s method of integration [2], we have

$$\int_\theta^{1+\theta} e^{\frac{U(z)-fz}{\epsilon}} \, dz = C(\theta, f) e^{\nu \frac{V(\theta)}{\epsilon}},$$

(17)

where

$$U^*(\theta) = \sup_{\theta \leq z < 1+\theta} \{U(z) - fz\},$$

(18)

and $C$ is bounded. The parameter $\nu$ is either $\frac{1}{2}$ or 1 depending on whether the Laplace integral is evaluated at an interior or a boundary point of the domain.

$u(\theta)$ in Eq. (15), therefore, has the form

$$u(\theta) = A C(\theta, f) e^{\nu \frac{V(\theta)}{\epsilon}},$$

(19)

in which

$$V(\theta) = U^*(\theta) - U(\theta) + f\theta$$

(20)

is periodic. Fig. 2 shows one example of how $U^*(\theta)$ is obtained from $U(\theta) - f\theta$, and $V(\theta)$ is obtained from $U^*(\theta)$. In general, $V(\theta)$ will have points of non-differentiability.

If the periodic $V(\theta)$ is not a constant, then it reaches its global maximum at a certain $\theta^*$. Then in the limit of $\epsilon \to 0$, the stationary distribution $u(\theta) \to \delta(\theta - \theta^*)$.

However, if

$$f > \max_{\theta \in [0,1]} U'(\theta),$$

then $U(\theta) - f\theta$ is a monotonically decreasing function of $\theta$. In this case, $U^*(\theta) = U(\theta) - f\theta$ and $V(\theta) \equiv 0!$ Furthermore, $\nu = 1$ and $C(\theta, f) = 1/(f - U'(\theta))$. Thus in the limit of $\epsilon \to 0$, we have

$$u(\theta) = \left( \int_0^1 \frac{dz}{f - U'(z)} \right)^{-1} \frac{1}{f - U'(\theta)},$$

(21)

Thus, the stationary distribution $u(\theta)$ reflects the non-uniform velocity on the circle in accordance with ergodic theory. The nature of a stable limit cycle being an attractor, however, is reflected by the constant $\phi(x)$ on the limit cycle, which has a dynamics on a different time scale when $\epsilon$ is small.
3.1 A simple example of diffusion on a circle

We now give a simple example: A nonlinear dynamics on a circle $\dot{\theta} = f - \sin(2\pi\theta)$. The corresponding Eq. (13) has a $U(\theta) = -1/(2\pi)\cos(2\pi\theta)$, $\theta \in \mathbb{S}[0, 1]$. For $f < 1$, the deterministic dynamics has a stable fixed point at $\theta^*$ and unstable fixed point at $\frac{1}{2} - \theta^*$, where we denote $\theta^* = \frac{1}{2\pi} \arcsin f$, $\theta^* \in [0, \frac{1}{2}]$. But for $f > 1$, it has no fixed point; instead it has a limit cycle. With periodic boundary condition, the stationary solution to Eq. (14) is Eq. (15) in the form of

$$u_\epsilon(\theta) = A_\epsilon \left( \int_0^{1+\theta} e^{-\frac{1}{\epsilon}(\frac{1}{2\pi}\cos(2\pi z) + fz)} dz \right) e^{\frac{1}{\epsilon}(\frac{1}{2\pi}\cos(2\pi\theta) + f\theta)}, \quad (22)$$

in which

$$A_\epsilon = \left[ \int_0^1 \left( \int_0^{1+\theta} e^{-\frac{1}{\epsilon}(\frac{1}{2\pi}\cos(2\pi z) + fz)} dz \right) e^{\frac{1}{\epsilon}(\frac{1}{2\pi}\cos(2\pi\theta) + f\theta)} d\theta \right]^{-1}.$$  

It is easy to show that when $f > 1$, $1/(2\pi)\cos(2\pi z) + fz$ is a monotonically increasing function of $z$. Hence applying Laplace’s method near $z = \theta$ one has [2]

$$u_0(\theta) = \lim_{\epsilon \to 0} u_\epsilon(\theta) = \frac{\sqrt{f^2 - 1}}{f - \sin(2\pi\theta)}, \quad (f > 1). \quad (23)$$

This is the case with deterministic limit cycle. According to the ergodic theory, $u_0(\theta) \propto 1/\dot{\theta}$.

When $f \leq 1$, one again applies Laplace’s method. We introduce $\hat{\theta}$, which satisfies

$$\frac{1}{2\pi} \cos(2\pi\hat{\theta}) + f\hat{\theta} = -\frac{\cos(2\pi\theta^*)}{2\pi} + f(1/2 - \theta^*).$$

If $\hat{\theta} \in [0, \frac{1}{2} - \theta^*]$, then one has

$$u_\epsilon(\theta) \approx A_\epsilon \exp \left[ \frac{1}{\epsilon} \begin{cases} 0 & 0 \leq \theta \leq \hat{\theta} \\ \frac{\cos(2\pi\theta) + \cos(2\pi\theta^*)}{2\pi} + f \left( \theta + \theta^* - \frac{1}{2} \right) & \hat{\theta} \leq \theta \leq \frac{1}{2} - \theta^* \\ 0 & \frac{1}{2} - \theta^* \leq 1 \end{cases} \right]. \quad (24)$$

If $\hat{\theta} \in [-\frac{1}{2} + \theta^*, 0]$, then we denote $\hat{\theta} = 1 + \hat{\theta} \in [\frac{1}{2} + \theta^*, 1]$. Then we have

$$u_\epsilon(\theta) \approx A_\epsilon \exp \left[ \frac{1}{\epsilon} \begin{cases} \frac{\cos(2\pi\theta) + \cos(2\pi\theta^*)}{2\pi} + f \left( \theta + \theta^* - \frac{1}{2} \right) & 0 \leq \theta \leq \frac{1}{2} - \theta^* \\ 0 & \frac{1}{2} - \theta^* \leq \theta \leq \hat{\theta} \\ \frac{\cos(2\pi\theta) + \cos(2\pi\theta^*)}{2\pi} + f \left( \theta + \theta^* - \frac{3}{2} \right) & \hat{\theta} \leq \theta \leq 1 \end{cases} \right]. \quad (25)$$
Note that both Eqs. (24) and (25) are periodic function of \( \theta \) on \([0, 1] \). The exponents in both are non-negative with a “flat region” of zero as \( V(\theta) \) illustrated in Fig. 2C. Furthermore, the maximum of \( V(\theta) \) is located at \( \theta^* \), the stable fixed point of the nonlinear dynamics. Therefore, the normalized \( u_0(\theta) = \delta(\theta - \theta^*) \) for \( f < 1 \).

Note that the limit of Eq. (23)

\[
\lim_{f \to 1^+} \frac{\sqrt{f^2 - 1}}{f - \sin(2\pi \theta)} = \delta \left( \theta - \frac{1}{4} \right).
\]

(26)

This simple example has been discussed widely in the nonlinear dynamic literature on synchronization and neural networks [34]. As expected for the attractor of a nonlinear dynamics with limit cycle, the law of large numbers for the corresponding stationary process is not a set of Dirac-delta measures, but a continuous one. Fig. 3 shows \( u_0(\theta) \) in Eq. (23) for several different values of \( f \).

4 General derivation for high dimensional systems with a Limit Cycle

The existence of limit cycles is indicative of a system being far from equilibrium (detailed balance). It has been widely believed that systems with limit cycle can not have a Lyapunov function. This is certainly true according to the strict definition of a Lyapunov function [25]. However, in a broader sense, functions with Lyapunov properties can be constructed for systems with limit cycle. We shall now consider a multi-dimensional \( F(x) \) and its corresponding \( \phi(x) \), as defined above.

First, we observe that the Lyapunov property of \( \phi \) immediately leads to the conclusion that \( \phi(x) = \text{constant} \) if \( x \in \Gamma \), where \( \Gamma \) is a limit cycle of \( F \). To show this, we simply note that

\[
\oint_{\Gamma} \nabla \phi \cdot d\vec{\ell} = 0,
\]

(27)

where the integrand

\[
\nabla \phi \cdot d\vec{\ell} = \frac{\nabla \phi \cdot F}{||F||} \leq 0.
\]

(28)

Hence, \( \nabla \phi(x) = 0 \), i.e., \( \phi(x) = \text{const} \), where \( x \in \Gamma \). Moreover in any small neighborhood of \( \Gamma \), \( \phi(x) \) must be all greater or less than that on \( \Gamma \) in the cases of stable or unstable limit cycles respectively. Here we do not consider the very complicated case such as strange attractor.

We now compute \( C_0(x) \) on \( \Gamma \). First of all, according to Eq. (12) and \( \nabla \phi(x) = 0 \) on \( x \in \Gamma \), we have

\[
\nabla C_0 \cdot F + C_0 \nabla \cdot F = 0,
\]

i.e. \( \nabla \cdot (C_0 F) = 0 \). Then we could pick any continuous segment of \( \Gamma \), and consider its \( \delta \)-thickness neighborhood. When \( \delta \) tends to zero, the only fluxes remain are the influx
and outflux of the vector field $C_0 \mathbf{F}$ along $\Gamma$, hence according to Gauss’ theorem, the values of the function $C_0 \mathbf{F}$ at the two ends of the segment must be the same. Therefore, $\|C_0 \mathbf{F}\| = C_0 \|\mathbf{F}\|$ must be constant along $\Gamma$.

Now we restrict the dynamics to $\Gamma$ and introduce an specific angular variable $\theta$, $\theta \in S$: $\theta$ is just the arc length starting from some fixed point on $\Gamma$. In this case, we have $\sum_i (\frac{dx_i}{d\theta})^2 \equiv 1$.

Then let

$$\Theta(\theta) = F_i(x(\theta)) \left( \frac{dx_i}{d\theta} \right)^{-1} = \|F\|, \quad i = 1, 2, \ldots, N,$$  

(29)

where $x(\theta) = (x_1(\theta), x_2(\theta), \ldots, x_N(\theta))$ and

$$\|F\| = \sqrt{\sum_i (F_i)^2},$$

such that the differential equation on the limit cycle $\Gamma$ becomes

$$\frac{d\theta}{dt} = \Theta(\theta).$$

(30)

Hence

$$C_0(\theta) = \frac{A}{\Theta(\theta)},$$

(31)

where $A$ is a normalization constant, whose meaning is very clear: According to ergodic theory, the stationary probability distribution of $\theta$ is simply the inverse of the angular velocity. In fact, the period of the limit cycle is

$$T = \int_0^{2\pi} \frac{d\theta}{\Theta(\theta)}.$$  

(32)

This corresponds to the flux on $\Gamma$, i.e, the number of cycles per unit time, according to Eq. (2):

$$J = A = \left( \int_0^{2\pi} \frac{d\theta}{\Theta(\theta)} \right)^{-1}.$$  

(33)

$J$ is also known as the rotation number in nonlinear dynamics.

We shall note that while $\phi(x)$ has the Lyapunov property, the stationary probability in Eq. (6) does not. The stationary $u_\epsilon(x)$ can not be a Lyapunov function since in general it is not a constant on $\Gamma$ due to the contribution from $C_0(x)$ [45].

If one chooses another angular parameter $\theta'$, then the reciprocal of the velocity $\Theta(\theta')$ will not be $C_0$. It is straightforward to modify the analysis presented above.
4.1 Beyond limit cycle

The discussion in this section is only heuristic; a more detailed mathematical analysis remains to be developed. From the result above, it seems reasonable that for a high-dimensional nonlinear ordinary differential equation with vector field \( \mathbf{F}(\mathbf{x}) \), its entire center manifold has a constant \( \phi(\mathbf{x}) \), if it exists. Similarly, \( \phi(\mathbf{x}) \) will be a constant on an invariant torus, i.e., quasi-periodic motion occurs. This is easy to illustrate from the simple example:

\[
\begin{align*}
\frac{d\theta}{dt} &= \Theta dt + \sqrt{2\epsilon D_1} dB^{(1)}_t, \\
\frac{d\xi}{dt} &= \Xi dt + \sqrt{2\epsilon D_2} dB^{(2)}_t,
\end{align*}
\]

in which \((\theta, \xi) \in \mathbb{S}^2\). When \(\Theta/\Xi\) is irrational, the entire \(\mathbb{S}^2\) is an invariant torus. However, the stationary probability for Eq. (34) is separable in \(\theta\) and \(\xi\). Hence according to the above results on the limit cycle, \(\phi(\theta, \xi)\) is constant on the entire \(\mathbb{S}^2\).

5 Local and global landscapes in the case of multiple attractors and emergent nonequilibrium steady state

Nonlinear dynamics on a circle, i.e., Eq. (14), can only be one of the three types: (a) single stable fixed point (attractor), (b) multiple stable fixed points (attractors), and (c) oscillation. Our focus so far has been mainly on (c), and transition from (a/b) to (c). However, even within (a) and (b), there are further distinctions between gradient systems with \(f = 0\) and non-gradient system with \(f \neq 0\). The latter is known as irreversible diffusion processes [43]. To complete the analysis, we now consider (b). For small \(\epsilon\), the dynamics exhibits two different time scales: intra-attractor dynamics and inter-attractor dynamics. The major questions here are (i) the relative stability of these attractors and (ii) the transition rates between different attractors. Note the unique feature of dynamics on the circle \(\mathbb{S}\), which is different from one-dimensional \(\mathbb{R}\), is the possibility of non-gradient, i.e., no detailed balance. Stationary, reversible diffusion process on \(\mathbb{R}\) has a single global landscape which simultaneously provides answers to both (i), e.g., Boltzmann’s law, and (ii) via Kramers’ theory [18, 12, 13]. This is not the case for stationary diffusion process on \(\mathbb{S}\). Although the fundamental theorems by Freidlin and Wentzell have been developed for quite a long time [9], their relation to nonequilibrium thermodynamics is still unknown. The present study, thus, serves an initiation for this interesting problem.

According to Freidlin and Wentzell [9], in the high-dimensional case as well as in the one-dimensional compact manifold, there are two types of landscapes: The local landscape underlies a Kramers’ theory-like analysis for a single transition from one basin of attraction to another. The global landscape, on the other hand, is for the relative stability in nonequilibrium steady state [13, 14]. The well-known Kramers’ rate theory states that the barrier crossing time is exponentially dependent on the barrier height and...
nearly exponential distributed \([3]\) when the noise strength tends to zero. Then, putting together all the transition rate constants computed from the Kramers’ theory, one obtains a discrete-state continuous-time Markovian chain (in chemistry, this is called discrete chemical kinetics.). According to a key theorem in \([43]\), one could then realize that such a Markov chain is equilibrium if and only if the local landscapes derived from the Freidlin-Wentzell local actions in each attractive domain could be continuously pieced together; the function pieced together is just the global landscape; this could only be guaranteed with the detailed balance condition.

Next, for each single domain (or basin) of attraction associated with a stable fixed point, applying the large deviation theory of Freidlin and Wentzell, one builds a local landscape \(\phi_i(x), i = 1, 2, ..., N\). And then for each pair of neighbouring attractive domains \(\Omega_i\) and \(\Omega_j\), one obtains a pair of local transition rates from Kramers’ theory: the transition rate \(k_{ij}\) from \(\Omega_i\) to \(\Omega_j\) is proportional to \(e^{-\frac{V(i,j)}{\epsilon}}\), where \(V(i,j)\) is the lowest barrier height of \(\phi_i(x)\) along the boundary with the attractive domain \(\Omega_j\).

This way, we obtain an emergent, discrete-state Markov network with state space \(\{1, 2, ..., N\}\) and transition rates \(K = \{k_{ij}\}_{N \times N}\) for non-diagonal elements. The diagonal elements of \(K\) are determined by requiring all its rows summed to zero. A stationary distribution can then be solved, with \(\pi = \{\pi_i\}\) satisfying

\[
\pi K = 0,
\]

Now we are ready for a crucial step: to paste (reshuffle) the local landscapes together in order to build the global one. The reshuffle procedure is somewhat subtle. There is an illustrative example in \([9]\) (also see Fig. 4). Here we give a simple demonstration in terms of a 3-state Markov chain.

In this case, we have

\[
\pi_1 = \frac{k_{23}k_{31} + k_{31}k_{21} + k_{21}k_{32}}{\mathcal{D}},
\]

\[
\pi_2 = \frac{k_{31}k_{12} + k_{12}k_{32} + k_{32}k_{13}}{\mathcal{D}},
\]

\[
\pi_3 = \frac{k_{12}k_{23} + k_{23}k_{13} + k_{13}k_{21}}{\mathcal{D}},
\]

in which the denominator \(\mathcal{D}\) is determined by \(\pi_1 + \pi_2 + \pi_3 = 1\).

When \(\epsilon\) tends to zero, let

\[
W_1 = -\lim_{\epsilon \to 0} \epsilon \log \pi_1 = \min\{V(2, 3) + V(3, 1), V(3, 1) + V(2, 1), V(2, 1) + V(3, 2)\},
\]

\[
W_2 = -\lim_{\epsilon \to 0} \epsilon \log \pi_2 = \min\{V(3, 1) + V(1, 2), V(1, 2) + V(3, 2), V(3, 2) + V(1, 3)\},
\]

\[
W_3 = -\lim_{\epsilon \to 0} \epsilon \log \pi_3 = \min\{V(1, 2) + V(2, 3), V(2, 3) + V(1, 3), V(1, 3) + V(2, 1)\}.
\]

So the reshuffle rule is as follows

\[
W(x) = \min\{W_1 + V(1, x), W_2 + V(2, x), W_3 + V(3, x)\},
\]
where \( V(i, x) \) means the minimum of the Freidlin-Wentzell action along the path between the \( i \)-th attractive domain and the position \( x \). The intuitive understanding is that starting from each attractor, and compare their probabilities for arriving at \( x \).

In the case of three states, for instance, if \( x \) is in the first attractive domain, then
\[
V(1, x) = \phi_1(x), \quad V(2, x) = V(2, 1) + \phi_1(x) \text{ if the backward trajectory starting from } x \text{ would arrive at the boundary between the first and second attractive domains, otherwise } V(2, x) = V(2, 1) + \phi_1(x), \quad V(3, x) = V(3, 1) + \phi_1(x).
\]
Hence, \( W(x) \) is continuous, and the global invariant distribution
\[
u(x) \propto e^{-(W(x) - \min_i \{W_i\})/\epsilon}. \tag{35}
\]
So \( W(x) - \min_i \{W_i\} \) is the global landscape, which also satisfies the Lyapunov property.

Furthermore, we know that the emergent Markovian chain is in equilibrium, if and only if \( k_{12}k_{23}k_{31} = k_{21}k_{32}k_{13} \) \[13\], which means \( V(1, 2) + V(2, 3) + V(3, 1) = V(2, 1) + V(3, 2) + V(1, 3) \), i.e. \( W_2 - W_1 = V(1, 2) - V(2, 1) \), \( W_3 - W_2 = V(2, 3) - V(3, 2) \) and \( W_1 - W_3 = V(3, 1) - V(1, 3) \). Hence the local landscape \( \phi_i(x) \) would be continuously connected at the boundaries in this case.

The above result is a generalization of the celebrated work of Kramers \[18\]. In Kramers’ theory, the underlined nonlinear diffusion process is the atomic dynamics along the reaction coordinate, while the emergent discrete dynamics is exactly the discrete chemical kinetics. For systems with detailed balance, Kramers’ rate constants are consistent with Boltzmann’s law for conformational probabilities. However, when detailed balance are not satisfied, we have clearly demonstrated here an essential difference between local and global landscapes: The former is related to individual state-to-state transition, while the latter is associated with a systems’ long-time dynamics. Their disagreement is the origin of nonequilibrium steady states \[13\], \[14\], \[26\].

6 Conclusions

Multi-dimensional diffusion processes and Markov jump processes with chemical master equations are two mathematical models for studying mesoscopic, nonequilibrium physical and biochemical dynamics with multiscale phenomena and emergent organizations \[27\]. In the past, our understandings of dynamics in terms of its molecular constituents have been mainly derived from theories of macroscopic, deterministic dynamics in the thermodynamic limit \[19\] or statistical mechanics of closed systems which are necessarily equilibrium. Much to be learned from the two types of stochastic models for the mesoscopic dynamics in open systems, especially the relationship between their asymptotic dynamics and emergent nonequilibrium steady state. Even for the simplest case of one-dimensional circle, there were important questions to be addressed and answered. In the present work, insights have been gained from applying methods of singular perturbation and the theory
of large deviations. It has been shown that the intuitive notion of a landscape can be further secured by applying the mathematical theories. Combining the insights with a wide range of existing applied mathematical techniques (see several reviews [21, 33, 24, 23]), the study illustrated here can be further taken into several directions. Strengthening the tie [32] between the abstract theory of large deviations [9] and more applied singular perturbation techniques [2, 17, 20] will yield further understandings for stochastic nonlinear dynamics (SND). In particular, the large deviation behavior of Delbrück-Gillespie process [27] is still poorly understood. The results from many previous workers synthesized in the present work also provides a glimpse of how to develop an alternative structural stability theory for nonlinear dynamical systems, as called by E.C. Zeeman many years ago [42]. One naturally wonders what $\phi$ and $C_0$ will be for a chemical reaction system that possesses a chaotic attractor [6]. Is there any regularity in the asymptotic behavior of the invariant measure for such systems [20, 5, 31, 44]? These are hard problems; but they are no longer impossible to conceive.

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Figure 1: Landscape $\phi(x)$ and related Kramers’ rate theory for a bistable system. The local minima correspond to stable fixed points of a deterministic dynamics $\dot{x} = -d\phi(x)/dx$ while the maximum corresponds to an unstable fixed (saddle) point. The $V(1,2)$ and $V(2,1)$ represent the energy barriers for exiting energy wells 1 and 2, respectively. For very small $\epsilon$, the stationary probability distribution for stochastic dynamics with Brownian motion $B(t)$, $dx = -(d\phi(x)/dx)dt + \sqrt{2\epsilon} \ dB(t)$, is $u_\epsilon(x) \propto \exp\left(-\frac{\phi(x)}{\epsilon}\right)$. The Kramers theory yields transition rates between the two attractors: $k_{12} \propto e^{-V(1,2)/\epsilon}$ and $k_{21} \propto e^{-V(2,1)/\epsilon}$. According to Freidlin-Wentzell’s LDT, this theory still applies for every pair of neighbouring attractors of a non-gradient system in terms of a local landscape. However, the stationary probability distribution follows a different, global landscape. Also see Fig. 4.
Figure 2: (A) The thin solid line is $U(\theta) - f\theta$, where $f$ is represented by the slope of the dashed line. The thick solid line is $U^*(\theta) = \sup_{z \in [\theta, \theta+1)} \{U(z) - fz\}$. When combining $U^*(\theta)$ with $-U(\theta) + f\theta$, as shown in (B), one obtains $V(\theta)$ given in (C). $V(\theta)$ is periodic but contains non-differentiable points. If $U(\theta) - f\theta$ is monotonically decreasing, then $U^*(\theta) = U(\theta) - f\theta$ and $V(\theta) = 0$. 
Figure 3: The limiting distribution $u_0(\theta)$ according to Eq. (23) for nonlinear dynamics on a circle $\dot{\theta} = f - \sin(2\pi \theta)$, with $f = 5, 2, 1.1$ and 1.05. With $f \to 1^+$, it approaches to $\delta(\theta - 0.25)$. For $f \leq 1$, the distribution is $\delta(\theta - \theta^*)$ where $\theta^* = 1/(2\pi \arcsin(f))$. 
Figure 4: (a) Pairwise local landscapes; (b) A simple “pasting together” leads to discontinuous matched case; (c) The global landscape is obtained by a “λ-surgery and pasting” procedure: The surgery lifts the well-2 with respect to well-1 an amount of $\ln(\pi_1 k_{12})/(\pi_2 k_{21})$, which is precisely the free energy difference $\Delta \mu_{12}$ for well-2 with respect to well-1 in nonequilibrium steady state. Similarly it lifts the amount of $\Delta \mu_{23} = \ln(\pi_2 k_{23})/(\pi_2 k_{32})$ for well-3 with respect to well-2, and $\Delta \mu_{31} = \ln(\pi_3 k_{31})/(\pi_1 k_{13})$ for well-1 with respect to well-3. Therefore, the total lift is $\Delta \mu_{12} + \Delta \mu_{23} + \Delta \mu_{31} = \ln(k_{12} k_{23} k_{31})/(k_{21} k_{32} k_{13})$; (d) The final global landscape. Note that $-V(x)$ in Fig. 2C is just one example of such a global landscape. It is a piecewise smooth function with “flat regions” at its local maxima.