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Abstract

We consider a system of independent particles on a finite state space, and prove a dynamic large-deviation principle for the empirical measure-empirical flux pair, taking the specific fluxes rather than net fluxes into account. We prove the large deviations under deterministic initial conditions, and under random initial conditions satisfying a large-deviation principle. We then show how to use this result to generalise a number of principles from Macroscopic Fluctuation Theory to the finite-space setting.

1 Introduction

A well-known strengthening of the Second Law states that for thermodynamically closed systems not only is the free energy non-increasing in time, but the system is *driven* by its free energy. Exactly how the free energy drives the system can often be made precise by linear response theory, that is, a constitutive relation between thermodynamic driving forces, defined as the derivative of the free energy, and the thermodynamic velocities, that is, $\dot{c}(t) = -K\big(c(t)\big)D\mathcal{F}\big(c(t)\big)$. If the state-dependent operator $K(c)$ is symmetric and positive definite, then such equations are actually gradient flows on the manifold with inverse metric tensor $K(c)$, and the Second Law follows as a consequence. Moreover, one then also has the free energy-dissipation balance, that is, the dissipation of free energy equals the free energy production. Since the work of Onsager it is well-known that the symmetry of the operator $K(c)$ is closely connected to detailed balance of an underlying microscopic system [Ons31] via large-deviation principles. In a general setting, one can always derive free energy-dissipation balances from microscopic systems satisfying detailed balance, although this may lead to non-linear response theories, and non-quadratic dissipations, in particular on discrete spaces [MPR14].

For microscopic systems that are not in detailed balance with macroscopic systems that are not thermodynamically closed, this is no longer true. A detailed balance condition can be violated either due to bulk forces, or due to boundary effects. Typical examples of the latter are the boundary-driven systems studied in for example [BDSG$^+$02, BDSG$^+$03, DLE03]. We will focus our attention on bulk forces. The typical case is a system with a uniform stationary state and an external non-conservative force field that causes mass to flow around in cycles, leaving the stationary state intact. Hence the work done by the external force field may have no effect on the concentration, and an energy-dissipation balance as described above is not to be expected. Naturally, the work done is not simply lost, but the resulting effect cannot be observed unless both the concentration and the fluxes are taken into account. This idea is the core of Macroscopic Fluctuation Theory (MFT), which allows to derive many thermodynamic properties for systems that are not in detailed balance, see the recent overview paper [BDSG$^+$15] and the references therein. The aim of the current paper is to provide a few steps towards a more general Macroscopic Fluctuation Theory, as follows.

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Firstly, most known results in MFT are stated for systems on a continuous space (at least macroscopically). Therefore the stochastic noise is usually driven by some noise that is approximately white, leading to Freidlin-Wentzell-type large deviations that can be written as a squared Hilbert norm. Of course, having a natural Hilbert structure at hand can be beneficial in many ways. For example, with a Hilbert structure one can split the large deviations into orthogonal components: gradient parts and solenoidal parts. However, it is more natural for processes on discrete spaces to be driven by Poisson noise; in that case the large deviations will no longer be quadratic. Nevertheless, in this paper we will see that many arguments of MFT are also valid in the non-quadratic case.

Secondly, it should be noted that most known MFT studies net fluxes only. In the current paper we distinguish between forward and backward fluxes through an edge, which we call *one-way* fluxes. This has a few advantages. Firstly, there could be forces that produce strong fluxes without changing the net flux; such forces speed up the mixing behaviour of the system and can therefore be important quantities to study. Secondly, it turns out that on discrete spaces the large deviation rate for the one-way fluxes takes on a very explicit form, whereas the large deviations for the net fluxes is defined implicitly, either via a convex dual, or via a minimisation problem. Indeed, this minimisation comes from a simple contraction principle, which can always be applied whenever only the net fluxes are of interest.

Thirdly, we study generalised gradient structures and what we call *dissipative structures* that are induced by the large-deviation Lagrangians. We show that in an abstract setting, such gradient structures must always be related to the adjoint of the continuity equation.

Some of the MFT principles that we consider are also studied in the work [BMN09], which is more physics-oriented and focuses on net fluxes rather than one-way fluxes.

1.1 Microscopic models

In this paper we consider two classes of models: (A) independent particles on a finite state space, and (B) reacting particles on a finite state space, where we allow for mean-field interaction and a broad range of possible reactions including annihilation, creation, coagulation and fragmentation. Of course the independent particle model is a subclass of the reacting particle model. For the independent case we will rigorously prove the many-particle limit and the corresponding large deviations, which is – maybe surprisingly given the independence of the particles – not completely trivial. For the reacting particle model the large deviations will only be calculated formally. We can then use the large deviations to study MFT for the more general setting of reacting particles.

(A) Let $X_1(t), X_2(t), \dots$ be independent copies of a Markov chain on a finite state space I, with generator matrix $\mathsf{Q} \in \mathbb{R}^{I\times I}$. We first assume the following initial conditions:

> Fix a $0 < \mu \in \mathcal{P}(I)$ and deterministic initial positions in I: $X_1(0) = x_1, X_2(0) = x_2, \dots$ in I, such that
> $C^{(n)}(0) := \frac{1}{n}\sum_{k=1}^n \mathbb{1}_{x_k} \xrightarrow[n\to\infty]{} \mu.$ (1)

Later we will consider random initial conditions as well. Throughout this paper, all processes will be of bounded variation, where we implicitly assume càdlàg representatives. Therefore the (random) set $\mathrm{jump}_t(X_k) := \{\hat{t} \in (0,t); X_k(\hat{t}^-) \neq X_k(\hat{t})\}$ of any Markov chain X_k will be at most countable

(actually finite a.s.). Define the empirical measure and the empirical integrated (one-way) flux by

$$C^{(n)}(t) := \frac{1}{n}\sum_{k=1}^{n} \mathbb{1}_{X_k(t)}, \qquad \text{and} \qquad \mathsf{W}^{(n)}(t) := \frac{1}{n}\sum_{k=1}^{n} \sum_{\hat{t}\in \mathrm{jump}_t(X_k)} \mathbb{1}_{\left(X_k(\hat{t}^-),X_k(\hat{t})\right)}.$$

Then the pair $\big(C^{(n)}(t), \mathsf{W}^{(n)}(t)\big)$ is a Markov process in $\mathcal{P}(I) \times \ell^1_+(I\times I)$ with generator

$$(\mathcal{Q}^{(n)}\phi)(c,\mathsf{w}) := n\sum_{i\neq j}\sum c_i \mathsf{Q}_{ij}\big[\phi(c^{i\to j}, \mathsf{w}^{ij+}) - \phi(c,\mathsf{w})\big], \tag{2}$$

and deterministic initial condition $\big(C^{(n)}(0), \mathsf{W}^{(n)}(0)\big) = \big(n^{-1}\sum_{k=1}^n \mathbb{1}_{x_k}, 0\big)$, where $c^{i\to j} := c - \frac{1}{n}\mathbb{1}_i + \frac{1}{n}\mathbb{1}_j$ and $\mathsf{w}^{ij+} := \mathsf{w} + \frac{1}{n}\mathbb{1}_{ij}$. This notation will only be used for fixed n, and so we can repress the dependence on that parameter. Here, $\ell^1_+(I\times I)$ is the space of non-negative matrices $\mathbb{R}_+^{I\times I}$ equipped with the norm $|\mathsf{w}|_1 = \sum\sum_{i\neq j}\mathsf{w}_{ij}$. By a slight abuse of notation we ignore diagonal elements in this space.

(B) The reacting particle model that we study is a classic model in the study of chemical reactions, which is sometimes know as the Chemical Master Equation (although this usually signifies a model with specific mass-action reaction rates), see [AK11] and the references therein. In this model the number of particles is no longer conserved, and so to avoid confusion we replace the parameter n by a parameter V (for volume) which controls the *order* of the number of particles in the system. Similar to initial condition (1.1) we fix a $\mu \in \ell^1_+(I)$, not necessarily a probability measure, such that initially the number of particles in the system is approximately $V|\nu|_1$. The dynamics will consist of a finite number of possible reactions $r \in \mathcal{R}$. A reaction r corresponds to removing of a number of particles $(\alpha_1^{(r)}, \dots, \alpha_I^{(r)}) \in \mathbb{N}^I$ and adding a number of particles $(\beta_1^{(r)}, \dots, \beta_I^{(r)}) \in \mathbb{N}^I$, so that the net result of a reaction r can be described by the *effective stoichiometric vector* $\gamma^{(r)} = \beta^{(r)} - \alpha^{(r)} \in \mathbb{Z}^I$. A reaction r takes place with a rate $k^{(V,r)}\big(C^{(V)}(t)\big)$ that depends on the empirical measure

$$C^{(V)}(t) := \frac{1}{V}\sum_{k=1}^{N^{(V)}(t)} \mathbb{1}_{X_k(t)},$$

where $N^{(V)}(t)$ denotes the number of particles in the system at time t. Similarly as above, we can now define the integrated flux as

$$\mathsf{W}_r^{(V)}(t) := \tfrac{1}{V}\#\big\{\text{reactions } r \text{ that occurred in time } (0,t]\big\}.$$

The pair $(C^{(V)}, \mathsf{W}^{(V)})$ is then a Markov process in $\ell^1_+(I)\times\ell^1_+(\mathcal{R})$, with generator

$$(\mathcal{Q}^{(V)}\phi)(c,\mathsf{w}) := \sum_r k^{(V,r)}(c)\big[\phi(c+\tfrac{1}{V}\gamma^{(r)}, \mathsf{w} + \tfrac{1}{V}\mathbb{1}_r) - \phi(c,\mathsf{w})\big]. \tag{3}$$

Observe that this indeed includes the independent case: $\mathcal{R} = I\times I\backslash\{i=j\}$, $k^{(n,(i,j))}(c) = nc_i\mathsf{Q}_{ij}$ and $\gamma^{(i,j)} = \mathbb{1}_j - \mathbb{1}_i$ and $V = n$.

1.2 Macroscopic models and large deviations

(A) In the many-particle limit, the independent particle process $(C^{(V)}, \mathsf{W}^{(V)})$ converges to the solution of

$$\dot{c}(t) + \operatorname{div}\dot{\mathsf{w}}(t) = 0, \tag{4a}$$
$$\dot{\mathsf{w}}(t) = c(t)\otimes\mathsf{Q}, \tag{4b}$$

with initial condition $\big(c(0),\mathsf{w}(0)\big)=\big(\mu,0\big)$, using the notation $(c\otimes\mathsf{Q})_{ij}:=c_i\mathsf{Q}_{ij}$ and $(\operatorname{div}\mathsf{w})_i:=\sum_{j\in I}\mathsf{w}_{ij}-\mathsf{w}_{ji}$. We will make this convergence result rigorous in Proposition 3.3. Observe that the net integrated flux through an edge (i,j) is just the anti-symetric matrix $\mathsf{w}_{ij}-\mathsf{w}_{ji}$, so that the one-way fluxes really encode more information. Equation (4a) is the *continuity equation*, or conservation law, and it plays an important role in MFT. Naturally, the microscopic equation also satisfies the continuity equation, hence microscopic fluctuations can only occur around (4b).

In Theorems 4.1 and 4.2 we prove large-deviation principles that characterise the dynamic microscopic fluctuations around the macroscopic limit:

$$\operatorname{Prob}\Big(\big(C^{(n)}(\cdot),\mathsf{W}^{(n)}(\cdot)\big)\approx(c,\mathsf{w})\Big)\sim e^{-n\mathcal{I}_0(c(0))-n\mathcal{J}_{(0,T)}(c,\mathsf{w})}\qquad\text{as } n\to\infty,$$

$$\mathcal{J}_{(0,T)}(c,\mathsf{w}):=\begin{cases}\int_0^T\mathcal{S}\big(\dot{\mathsf{w}}(t)|c(t)\otimes\mathsf{Q}\big)\,dt, & \text{if } (c,\mathsf{w})\in\\ & \qquad W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)\\ & \text{and } \dot c+\operatorname{div}\dot{\mathsf{w}}=0,\\ & \text{and } \lim_{t\searrow0}\mathsf{w}(t)=0,\\ \infty, & \text{otherwise.}\end{cases}\tag{5}$$

Here $\mathcal{S}(\hat{\mathsf{A}}|\mathsf{A}):=\sum\sum_{i\neq j}\lambda_B(\hat{\mathsf{A}}_{ij}/\mathsf{A}_{ij})\mathsf{A}_{ij}$ is the usual relative entropy and $\lambda_B(z):=z\log z-z+1$ for $z>0$ and $\lambda_B(0):=0$ is the Boltzmann function. We first prove the large deviations under initial condition (1.1), in which case

$$\mathcal{I}_0\big(c(0)\big)=\begin{cases}0, & c(0)=\mu,\\ \infty, & \text{otherwise.}\end{cases}\tag{6}$$

After that we will prove the large-deviation principle for more general initial conditions, where $C^{(n)}(0)$ is assumed to be random, and satisfying a large-deviation principle with some given rate functional $\mathcal{I}_0$.

(B) For the reacting particle model we assume that the average reaction rates converge $\frac{1}{V}k^{(r,V)}\to\bar k^{(r)}$ as $V\to\infty$. Then under suitable assumptions, see for example [Kur70, Kur72, SW95, PR16], the process $(C^{(V)},\mathsf{W}^{(V)})$ converges to the solution of

$$\dot c(t)=\Gamma\mathsf{w}(t),\tag{7a}$$
$$\dot{\mathsf{w}}(t)=\bar k(c(t)),\tag{7b}$$

where

$$\Gamma:=\big[\gamma^{(1)}\dots\gamma^{(\mathcal{R})}\big].\tag{7c}$$

Of course, (7a) can again be interpreted as a continuity equation. Formally, we shall see that the pair $(C^{(V)},\mathsf{W}^{(V)})$ satisfies a large-deviation principle,

$$\operatorname{Prob}\Big(\big(C^{(V)}(\cdot),\mathsf{W}^{(V)}(\cdot)\big)\approx(c,\mathsf{w})\Big)\sim e^{-V\mathcal{I}_0(c(0))-V\mathcal{J}_{(0,T)}(c,\mathsf{w})}\qquad\text{as } V\to\infty,$$

$$\mathcal{J}_{(0,T)}(c,\mathsf{w}):=\begin{cases}\int_0^T\mathcal{S}\big(\dot{\mathsf{w}}(t)|\bar k(c(t))\big)\,dt, & \text{if } (c,\mathsf{w})\in W^{1,1}\big(0,T;\mathfrak{l}^1_+(I)\times\mathfrak{l}^1_+(\mathcal{R})\big)\\ & \text{and } \dot c-\Gamma\mathsf{w}=0,\\ & \text{and } \lim_{t\searrow0}\mathsf{w}(t)=0,\\ \infty, & \text{otherwise.}\end{cases}\tag{8}$$

1.3 Overview

The paper is organised into two parts. In the first part we rigorously prove the flux large deviations for the independent particles case, and formally for the reacting particle system. In Section 2 we introduce the space, the topology and the sigma algebra that will be used for the large deviations. In Section 3 we discuss well-posedness for the microscopic and macroscopic models, and show that the microscopic process converges to the deterministic macroscopic equation; these results are needed for the large deviations. In Section 4 we rigorously prove the dynamic large-deviation principle large-deviation principle (5) for the state-flux pair in the independent case, first under deterministic initial conditions and then under random initial conditions. As common in dynamical large deviation theory, the difficulty lies in showing that the rate functional can be approximated by a set of sufficiently regular paths in order to prove the lower bound. It is also common to either regularise the perturbation factor [PR16], or replace the probabilities by exponentially equivalent ones [SW95, Ch. 5]. However, in the current work we directly regularise the paths (c, w), and exploit the explicit formulation (5) of the rate functional. Unfortunately this argument is not easily generalisable to the reacting particle model, see Remark 4.19. Nevertheless we will formally derive those large deviations at the end of Section 4. In the second part of the paper, i.e. Section 5, we will work in the more general setting, assuming the large deviations for the reacting particle model hold. We then explore what these large deviations imply for MFT on discrete spaces with one-way fluxes. In particular, we derive a number of time-reversal symmetries. After this we introduce the concept of dissipative structures and its relation with generalised gradient structures, and apply these concepts to the setting of this paper, where we will need the net fluxes rather than the one-way fluxes.

2 Preliminaries

2.1 Paths of bounded variation

Throughout this paper we consider paths of bounded variation on an arbitrary time interval $(0,T)$. For any Banach space valued function $f \in L^1(0,T;X^*)$, the essential pointwise variation is

$$\mathrm{epvar}(f) := \inf_{g=f \text{ a.e.}} \sup_{0<t_1<\ldots<t_K<T} \sum_{k=1}^{K} \|g(t_k) - g(t_{k-1})\|_{X^*},$$

where the supremum runs over all finite partitions of the interval $(0,T)$. The space of paths of bounded variation is defined as

$$\mathrm{BV}(0,T;X^*) := \Big\{ f \in L^1(0,T;X^*) : \mathrm{epvar}(f) < \infty \Big\}.$$

Any function of bounded variation f has a weak derivative $\dot f \in \mathcal{M}(0,T;X^*)$ which is a X^*-valued bounded measure on $(0,T)$ with total variation norm $\|\dot f\|_{\mathrm{TV}} = \mathrm{epvar}(f)$ [AFP06, Prop. 3.6 & Th. 3.27]. In addition, if a function of bounded variation is absolutely continuous, then f lies in $W^{1,1}(0,T;X^*)$ [AFP06, p. 139]. Moreover, any path of bounded variation $f \in \mathrm{BV}(0,T;X^*)$ has a càdlàgrepresentative, that satisfies $f(t) = f(0) + \dot f\big((0,t]\big)$ [AFP06, Th. 3.28].

We will be particarly concerned with the set

$$\mathrm{BV}_{\mathrm{flux}} := \mathrm{BV}_{\mathrm{flux}}\big(0,T;\mathcal{P}(I) \times \mathfrak{l}^1(I\times I)\big) := \\ \Big\{ (c,\mathsf{w}) \in \mathrm{BV}\big(0,T;\mathcal{P}(I) \times \mathfrak{l}^1(I\times I)\big) : \dot c = -\operatorname{div} \dot{\mathsf{w}} \Big\}.$$

Observe that the continuity equation in this definition is in general a measure-valued equation in time, i.e. $\dot{c}(dt) = -\operatorname{div} \dot{\mathsf{w}}(dt)$. In this sense both the microscopic model and the macroscopic model satisfies the continuity equation.

Throughout the paper we will need the following three simple estimates for any $(c, \mathsf{w}) \in \mathrm{BV}_{\mathrm{flux}}$:

$$\operatorname{epvar}(c) = \|\dot{c}\|_{TV} = \|\operatorname{div} \dot{\mathsf{w}}\|_{TV} \leq 2 \|\dot{\mathsf{w}}\|_{TV} = 2 \operatorname{epvar}(\mathsf{w}), \tag{9}$$

$$\|c\|_{L^1(0,T;\mathcal{P}(I))} = \int_0^T |c(t)|_1 \, dt = T, \tag{10}$$

$$\begin{aligned} \|\mathsf{w}\|_{L^1(0,T;\mathfrak{l}^1(I\times I))} &= \int_0^T |\mathsf{w}(t)|_1 \, dt = \int_0^T |\dot{\mathsf{w}}((0,t])|_1 \, dt \\ &\leq \int_0^T |\dot{\mathsf{w}}|_1((0,T]) \, dt = T \, \|\dot{\mathsf{w}}\|_{TV} = T \operatorname{epvar}(\mathsf{w}). \end{aligned} \tag{11}$$

Remark 2.1. Inequality (9) actually becomes an equality if w does not have simultaneous jumps. For microscopic paths $(C^{(n)}, \mathsf{W}^{(n)})$ with finite n this is indeed almost surely the case. □

2.2 The hybrid topology and Borel sigma-algebra

In this paper we always equip the space of paths of bounded variation with the *hybrid topology*. This topology is defined via the convergent nets as follows. We say that a sequence (or net) $(c^n, \mathsf{w}^n)_n$ in $\mathrm{BV}\big(0,T;\mathcal{P}(I) \times \mathfrak{l}^1(I\times I)\big)$ *converges hybridly* to a path of bounded variation (c, w) whenever

$$\begin{aligned} &(c^n, \mathsf{w}^n) \to (c, \mathsf{w}) \text{ strongly in } L^1\big(0,T;\mathcal{P}(I) \times \mathfrak{l}^1(I\times I)\big) \qquad \text{and} \\ &(\dot{c}^n, \dot{\mathsf{w}}^n) \to (\dot{c}, \dot{\mathsf{w}}) \text{ vaguely,} \end{aligned}$$

that is, against all test functions in $C_0\big(0,T;\mathbb{R}^{I\times I\times I}\big)$. It should be noted that this topology is mostly known in the literature as the weak-* topology [AFP06, Def. 3.11]. The term hybrid topology was recently introduced in [HPR16] to distinguish it from the functional analytical weak-* topology; strictly speaking, the two topologies only coincide in finite dimensions and restricted to bounded variation balls.

The space $\mathrm{BV}\big(0,T;\mathcal{P}(I) \times \mathfrak{l}^1(I\times I)\big)$ equipped with the hybrid topology is not a Polish space - it is not metrisable. This could make it difficult to do probability theory on this space. It is, however, completely regular and perfectly normal [HPR16, Th. 3.15]. Therefore, the corresponding Borel sigma-algebra 'behaves nicely', and the standard tools from probability theory that we will need are still valid [HPR16, Sec. 4].

We choose to work with the space of paths of bounded variation rather than the usual Skorohod space since the compactness criteria are very easy. This simplifies the proof of the exponential tightness, Proposition 4.6, considerably. In particular, because of the estimates (9),(10) and (11) above,

Proposition 2.2 ([AFP06, Th. 3.23] and [HPR16, Th. 3.18])**.** *Let $\mathcal{F} \subset \mathrm{BV}_{\mathrm{flux}}$ be a subset of finite variation: $\sup_{(c,\mathsf{w})\in\mathcal{F}} \operatorname{epvar}(\mathsf{w}) < \infty$. Then $\mathcal{F}$ is hybrid-compact.*

A similar argument is used in [PR16] to prove large deviations for infinite chemical reaction networks, and related approaches can be found in [Jak97] and [BFG15].

3 Well-posedness and many-particle limit

We now discuss the well-posedness of both the microscopic and macroscopic model, and provide a short proof of the convergence from the micro to the macro model. The proofs of the results in this section are fairly standard, but we include them for completeness. For the proof of the large deviations in Section 4, we will in fact need these results for a slightly more general process, perturbed by an abitrary $u \in L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$, with generator:

$$(\mathcal{Q}^{(n)}_{u(t)}\phi)(c,\mathsf{w}) := n\sum_{i\neq j}\sum c_i \mathsf{Q}_{ij}u_{ij}(t)\big[\phi(c^{i\to j},\mathsf{w}^{ij+})-\phi(c,\mathsf{w})\big]. \tag{12}$$

3.1 Well-posedness

Since the jump rates $nc_iQ_{ij}u_{ij}(t)$ are uniformly bounded in t and the hybrid sigma-algebra is uniquely characterised by the finite-dimensional distributions [HPR16, Th. 4.5], the following result becomes trivial:

Proposition 3.1 (Existence for the microscopic system)**.** *Let $n\in\mathbb{N}$, $C^{(n)}(0)\in\mathcal{P}(I)\cap\frac{1}{n}\mathbb{N}^I$, and $\mathsf{W}^{(n)}(0)=0$, and let the process $(C^{(n)},\mathsf{W}^{(n)})$ be defined by the perturbed generator $\mathcal{Q}^{(n)}_{u(t)}$. Then there exists a unique probability measure for $(C^{(n)},\mathsf{W}^{(n)})$ on the space $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$, i.e. for any hybridly-measurable set $\mathcal{U}\subset\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$,*

$$\mathbb{P}^{(n)}_u(\mathcal{U}) := \mathrm{Prob}\left((C^{(n)},\mathsf{W}^{(n)})\in\mathcal{U}\right) \tag{13}$$

and similarly we define the measure $\mathbb{P}^{(n)}$ whenever $u\equiv 0$. Moreover, the process $(C^{(n)},\mathsf{W}^{(n)})$ lies almost surely in $\mathrm{BV}_{\mathrm{flux}}$.

The well-posedness of the macroscopic system is a bit more involved:

Proposition 3.2 (Well-posedness of the macroscopic system)**.** *(i) Let $u\in L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$, and let $(\mu,v)\in\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)$ be given. Then there exists a unique solution $(c,\mathsf{w})\in W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ to the perturbed system of equations*

$$\begin{cases}\dot c(t) = -\operatorname{div}\dot{\mathsf{w}}(t), & t\in(0,T)\\ \dot{\mathsf{w}}(t) = c(t)\otimes\mathsf{Q}\otimes u(t), & t\in(0,T)\\ \lim_{t\searrow 0}c(t)=\mu,\\ \lim_{t\searrow 0}\mathsf{w}(t)=v.\end{cases} \tag{14}$$

Moreover, the solution (c,w) also lies in $W^{1,\infty}\big(0,T;\mathfrak{l}^1(I\times I)\big)$.

(ii) Let $u\in L^\infty_+\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$. For any $t\in(0,T)$ the solution operator $\psi(t):\big(\mu,v\big)\mapsto\big(c(t),\mathsf{w}(t)\big)$ for the initial value problem (14) *is a linear and bounded operator mapping $\mathfrak{l}^1(I\times I\times I)$ into itself.*

Proof. (i) The proof is by a standard Picard-Lindelöf fixed point argument.

We first prove existence and uniqueness of solutions for the slowed-down system

$$\begin{cases}\dot{\mathsf{w}}(t) = \alpha\big(\mu-\operatorname{div}\mathsf{w}(t)\big)\otimes\mathsf{Q}\otimes u(t), & t\in(0,T),\\ \lim_{t\searrow 0}\mathsf{w}(t)=v,\end{cases} \tag{15}$$

where $0 < \alpha \leq 1$ will be determined later. Observe that for $\alpha = 1$ the problem (15) coincides with (14) once we retrieve the variable c through the continuity equation (4a). Denote

$$W^{1,1}_v := \Big\{ \mathsf{w} \in W^{1,1}\big(0,T; \mathfrak{l}^1(I\times I)\big) : \lim_{t\searrow 0} \mathsf{w}(t) = v \Big\},$$

and define the operator $A : W^{1,1}_v \to W^{1,1}_v$ by

$$A[\mathsf{w}](t) := v + \alpha \int_0^t \big(\mu - \operatorname{div} \mathsf{w}(s)\big) \otimes \mathsf{Q} \otimes u(s)\, ds.$$

Naturally, solutions of (15) are fixed points of A. We can estimate for any $\mathsf{w}, \hat{\mathsf{w}} \in W^{1,1}_v$:

$$\|A[\mathsf{w}] - A[\hat{\mathsf{w}}]\|_{\mathrm{BV}} \leq \alpha(T+1)\|\mathsf{w} - \hat{\mathsf{w}}\|_{L^1} |\mathsf{Q}|_1 \|u\|_{L^\infty}.$$

Hence for α small enough, the Banach Fixed Point Theorem gives the existence and uniqueness of a solution to the slowed-down system (15). By rescaling time we find a unique solution of the original system (14) up to time αT. Repeating this process a finite number of times gives existence and uniqueness in $W^{1,1}$ of the solution up to time T.

For the regularity in $W^{1,\infty}$, observe that $\dot{\mathsf{w}}(t) = c(t) \otimes \mathsf{Q} \otimes u(t)$ is uniformly bounded, and hence so are $\dot{c}(t) = -\operatorname{div} \dot{\mathsf{w}}(t)$ and $c(t)$ and $\mathsf{w}(t)$.

(ii) The linearity of ψ is immediate from the linearity of the system (14). The boundedness in the first variable is also trivial since $|c(t)|_1 = |\mu|_1$. For the boundedness in w, note that $u \geq 0$ implies that $\mathsf{w}(t)$ is non-negative and non-decreasing. Therefore $\frac{d}{dt}|\mathsf{w}(t)|_1 = |\dot{\mathsf{w}}(t)|_1 \leq |\mu|_1 |\mathsf{Q}|_1 \|u\|_\infty$, which proves that $|\mathsf{w}(t)|_1 \leq t|\mu|_1|\mathsf{Q}|_1\|u\|_\infty$. □

3.2 Many-particle limit

We now state that the microscopic system converges to the macroscopic system in the many-particle limit. Of course, this limit is an immediate consequence of the law of the large numbers, see for example [Dud89, Th. 11.4.1]. However, the proof below has a bit more structure that allows for generalisations.

Theorem 3.3 (Many-particle limit)**.** *Fix (deterministic)* $(C^{(n)}(0))_n$ *and* μ *in* $\mathcal{P}(I)$ *such that* $C^{(n)}(0) \to \mu$*, and* $\mathsf{W}^{(n)}(0) \equiv 0$*. Let* $0 \leq u \in L^\infty\big(0,T; \mathfrak{l}^\infty(I\times I)\big)$ *be non-negative and bounded, and let* $(C^{(n)}, \mathsf{w}^{(n)})$ *be the perturbed process with generator* (12) *starting from* $(C^{(n)}(0), 0)$*. Then* $(C^{(n)}, \mathsf{W}^{(n)})$ *converges in probability in* $\mathrm{BV}\big(0,T; \mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$ *with the hybrid topology to the (deterministic) solution* (c, w) *of the perturbed problem* (14)*.*

Proof. We first prove convergence of the finite-dimensional distributions by operator convergence. Let $\mathcal{Q}_{u(t)} : C^1_b\big(\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big) \to C_b\big(\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ be the deterministic generator corresponding to the system (14), i.e.:

$$(\mathcal{Q}_{u(t)}\phi)(c,\mathsf{w}) = \sum_{i\neq j}\sum c_i \mathsf{Q}_{ij} u_{ij}(t)\, \nabla\phi(c,\mathsf{w}) \cdot (\mathbb{1}_j - \mathbb{1}_i, \mathbb{1}_{ij}). \tag{16}$$

The corresponding semigroup is of course $\big(S(t)\phi\big)(c,\mathsf{w}) = \phi\big(\psi(t)(c,\mathsf{w})\big)$, where $\psi(t)$ is the solution operator. Because of Proposition 3.2(ii), the semigroup $S(t)$ maps $C^1_b\big(\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ into itself, and hence $C^1_b\big(\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ is a core for the limit generator $Q_{u(t)}$ [EK05, Ch. 1, Prop. 3.3].

Moreover, for any test function ϕ in this core we clearly have $\|\mathcal{Q}^{(n)}_{u(t)}\phi - \mathcal{Q}_{u(t)}\phi\|_{C_b(\mathcal{P}(I)\times \mathfrak{l}^\infty(I\times I))} \to 0$. Therefore, due to the Trotter-Kurtz Theorem [Kal97, Th. 17.25] all finite-dimensional distributions convergence.

By the tightness of the process $(C^{(n)}, \mathsf{W}^{(n)})$ in $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$, which will be proven in Proposition 4.6, we also have narrow convergence of the paths in $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$, see [HPR16, Prop. 4.8]. Finally, narrow convergence to a deterministic limit implies convergence in probability. □

4 Large deviations

In this section we rigorously prove the dynamic large-deviation principle. First we prove the dynamic large deviations under deterministic initial conditions:

Theorem 4.1 (Large-deviation principle I)**.** *Fix a $\mu \in \mathcal{P}(I), \mu > 0$ (coordinate-wise) satisfying initial condition* (1.1)*. Then the pair $(C^{(n)}, \mathsf{W}^{(n)})$ satisfies a large-deviation principle in $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$ with the hybrid topology, with good rate functional $\mathcal{J}_{(0,T)}$ from* (5)*, where we implicitly set $\mathcal{J}_{(0,T)}(c,\mathsf{w}) = \infty$ whenever $\lim_{t\searrow 0}\big(c(t),\mathsf{w}(t)\big) = \big(\mu,0\big)$ is violated.*

Next we derive the coupled large-deviation principle if the initial conditions satisfy a large-deviation principle themselves:

Theorem 4.2 (Large-deviation principle II)**.** *Let the random variables $C^{(n)}(0)$ satisfy a large-deviation principle in $\mathcal{P}(I)$ with rate functional $\mathcal{I}_0$, and assume that $\mathcal{I}_0$ is left-continuous at the lower boundary $\{c\in\mathcal{P}(I) : c_i = 0$ for some $i\in I\}$. Then the pair $(C^{(n)}, \mathsf{W}^{(n)})$ satisfies a large-deviation principle in $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$ with the hybrid topology, with good rate functional $(c,\mathsf{w})\mapsto \mathcal{I}_0\big(c(0)\big) + \mathcal{J}_{(0,T)}(c,\mathsf{W})$, where we implicitly set $\mathcal{J}_{(0,T)}(c,\mathsf{w}) = \infty$ whenever $\lim_{t\searrow 0}\mathsf{w} = 0$ is violated.*

We largely follow the ideas from [SW95, Ch. 5] and [PR16]. The assumption that $\mu > 0$ is believed to be technical, and has to do with the construction of the approximation sequence in the lower bound. A similar assumption is used in for example [BDSG$^+$07, Th. 2.1], although the approximation argument is very different. In our Theorem 4.2 above, this assumption is no longer needed.

In Subsection 4.2 we prove that the sequence is exponentially tight, in Subsection 4.3 we prove the large-deviation lower bound, and in Subsection 4.4 we prove the large deviation upper bound on compact sets. Together, these results imply the large-deviation principle Theorem 4.1, with a good rate functional [DZ87, Lem. 1.2.18]. In order to prove both bounds, we first need some results about the rate functional, which are proven in Subsection 4.1 below. In Subsection 4.5 we prove large-deviation Theorem 4.2 for the coupled system with random initial conditions.

4.1 Analysis of the rate functional

We now prove a number of properties of the rate functional that will be needed in the large-deviations proof. For some of these properties, it is helpful to consider the system as a chemical reacting particle system on the space $I\times I\times I$, where a one-particle jump i to j causes a state change of $\gamma^{(ij)} := (\mathbb{1}_j - \mathbb{1}_i, \mathbb{1}_{ij})$, which occurs with intensity $c_i\mathsf{Q}_{ij}$. Some conditions from the paper [PR16] are violated, but the ones that we will need in this section are easily verified. We can then use the following result:

Proposition 4.3. *Let* $(c,\mathsf{w}) \in \mathrm{BV}_{\mathrm{flux}}$ *such that* $\mathcal{J}_{(0,T)}(c,\mathsf{w}) < \infty$*. Then* (c,w) *is absolutely continuous, that is, we can identify the path* (c,w) *with a function in* $W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$.

Proof. For this theorem we can immediately apply [PR16, Prop. 4.1] after checking the following conditions:

- $1 \le \inf_{i,j\in I:i\neq j}|\gamma^{(ij)}|_1 = \sup_{i,j\in I:i\neq j}|\gamma^{(ij)}|_1 = 3 < \infty$,
- $\sup_{(c,\mathsf{w})\in\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)} \sum\sum_{i\neq j} c_i\mathsf{Q}_{ij} = \sum\sum_{i\neq j}\mathsf{Q}_{ij} < \infty$.

□

As often in large deviation theory it will be beneficial to have a dual formulation of the rate functional at hand. For any $c\in L^1\big(0,T;\mathcal{P}(I)\big)$, $\mathsf{j}\in L^1\big(0,T;\mathfrak{l}^1(I\times I)\big)$ and $\zeta\in L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$, let

$$G(c,\mathsf{j},\zeta) := {}_{L^\infty}\langle\zeta,\mathsf{j}\rangle_{L^1} - \int_0^T \mathcal{H}\big(c(t),\zeta(t)\big)\,dt \qquad \text{and}$$
$$\mathcal{H}(c,\zeta) := \sum\sum_{i\neq j} c_i\mathsf{Q}_{ij}\big(e^{\zeta_{ij}}-1\big), \tag{17}$$

denoting ${}_{L^\infty}\langle\zeta,\mathsf{j}\rangle_{L^1} := \sum\sum_{i\neq j}\int_0^T \zeta_{ij}(t)\,\mathsf{j}_{ij}(t)\,dt$, again excluding the diagonal. We then have the following:

Proposition 4.4. *For any* $(c,\mathsf{w}) \in W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)\cap \mathrm{BV}_{\mathrm{flux}}$ *with initial condition* $\lim_{t\searrow 0}(c(t),\mathsf{w}(t)) = (\mu,0)$*,*

$$\mathcal{J}_{(0,T)}(c,\mathsf{w}) = \sup_{\zeta\in C_b^2(0,T;\mathfrak{l}^\infty(I\times I))} G(c,\dot{\mathsf{w}},\zeta).$$

Moreover, if $\mathcal{J}_{(0,T)}(c,\mathsf{w}) < \infty$ *then*

$$\mathcal{J}_{(0,T)}(c,\mathsf{w}) = \sup_{\zeta\in C_c^2([0,T);\mathfrak{l}^\infty(I\times I))} G(c,\dot{\mathsf{w}},\zeta). \tag{18}$$

Proof. Fix a $c\in \mathrm{BV}\big(0,T;\mathcal{P}(I)\big)$. For any $\zeta\in L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$ there holds

$$\sup_{\mathsf{j}\in L^1(0,T;\mathfrak{l}^1(I\times I))} {}_{L^\infty}\langle\zeta,\mathsf{j}\rangle_{L^1} - \int_0^T \mathcal{S}\big(\mathsf{j}(t)|c(t)\otimes\mathsf{Q}\big)\,dt$$
$$\le \int_0^T \sup_{\mathsf{j}(t)\in\mathfrak{l}^1(I\times I)} \Big[\zeta(t)\cdot\mathsf{j}(t) - \mathcal{S}\big(\mathsf{j}(t)|c(t)\otimes\mathsf{Q}\big)\Big]\,dt = \int_0^T \mathcal{H}\big(c(t),\zeta(t)\big)\,dt. \tag{19}$$

The point-wise maximiser on the right-hand side is $\mathsf{j}:(i,j,t)\mapsto c_i(t)\mathsf{Q}_{ij}\big(e^{\zeta_{ij}(t)}-1\big)$, which lies in $L^1\big(0,T;\mathfrak{l}^1(I\times I)\big)$. Hence the point-wise maximiser is also the global maximiser of the left-hand side, and inequality (19) is in fact an equality. From the Moreau-Fenchel Theorem [Bré83, Th. I.10] it follows that

$$\int_0^T \mathcal{S}\big(\dot{\mathsf{w}}(t)|c(t)\otimes\mathsf{Q}\big)\,dt = \sup_{\zeta\in L^\infty(0,T;\mathfrak{l}^\infty(I\times I))} G(c,\dot{\mathsf{w}},\zeta).$$

We can now show that we can replace the supremum over L^∞-functionals by a supremum over C^2_b-functions. To prove this, take any $\zeta \in L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$ and approximate it by $\zeta^{(\epsilon)} := \zeta * \theta^{(\epsilon)}$ where

$$\theta^{(\epsilon)}(t) = \frac{1}{4\pi\epsilon} e^{-t^2/4\epsilon}, \tag{20}$$

such that $\zeta^{(\epsilon)} \in C^2_b\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$ and $\zeta^{(\epsilon)} \to \zeta$ weakly in $L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$. Note that by equality (19), the functional $\zeta \mapsto \int_0^T \mathcal{H}\big(c(t),\zeta(t)\big)\,dt$ is the supremum of weakly continuous functionals, and hence it is itself weakly lower semicontinuous in L^∞. Therefore we get

$$\begin{aligned}\limsup_{\epsilon\to 0} G(c,\dot{\mathsf{w}},\zeta^{(\epsilon)}) &\le \limsup_{\epsilon\to 0} {}_{L^\infty}\langle \zeta^{(\epsilon)},\dot{\mathsf{w}}\rangle_{L^1} - \liminf_{\epsilon\to 0}\int_0^T \mathcal{H}\big(c(t),\zeta^{(\epsilon)}(t)\big)\,dt\\ &\le G(c,\dot{\mathsf{w}},\zeta),\end{aligned}$$

which proves the claim.

Finally, we show (18), assuming $\mathcal{J}_{(0,T)} < \infty$. Take a $\zeta \in C^2_b\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$, and approximate it by $\zeta\eta^{(\epsilon)}$, where $\eta^{(\epsilon)} : (0,T)\to[0,1]$ is smooth such that $\eta^{(\epsilon)}|_{(0,T-\epsilon]} = 1$ and $\eta^{(\epsilon)}|_{(T-\frac12\epsilon,T]} = 0$. Let $g_{ij}(t,\zeta_{ij}) := \zeta_{ij}\dot{\mathsf{w}}_{ij}(t) - c_i(t)\mathsf{Q}_{ij}\big(e^{\zeta_{ij}}-1\big)$ and $s_{ij}(t) = \lambda_B\big(\dot{\mathsf{w}}_{ij}(t)/(c_i(t)\mathsf{Q}_{ij})\dot{\mathsf{w}}_{ij}(t)$. For almost every t and any coordinate pair $i\neq j$, we can distinguish three different cases (see Figure 1):

(i) if $\zeta_{ij}(t) < 0$, then

$$g_{ij}\big(t,\zeta_{ij}(t)\big) \le g_{ij}\big(t,\zeta_{ij}(t)\eta^{(\epsilon)}(t)\big) \le 0 \le s\big(\dot{\mathsf{w}}_{ij}(t)|c_i(t)\mathsf{Q}_{ij}\big),$$

(ii) if $\zeta_{ij}(t)\ge 0$ and $g_{ij}\big(t,\zeta_{ij}(t)\big)\ge 0$, then

$$0 \le g_{ij}\big(t,\zeta_{ij}(t)\eta^{(\epsilon)}(t)\big) \le s\big(\dot{\mathsf{w}}_{ij}(t)|c_i(t)\mathsf{Q}_{ij}\big),$$

(iii) if $\zeta_{ij}(t)\ge 0$ and $g_{ij}\big(t,\zeta_{ij}(t)\big) < 0$, then

$$g_{ij}\big(t,\zeta_{ij}(t)\big) \le g_{ij}\big(t,\zeta_{ij}(t)\eta^{(\epsilon)}(t)\big) \le s\big(\dot{\mathsf{w}}_{ij}(t)|c_i(t)\mathsf{Q}_{ij}\big).$$

In all cases we have the t-almost everywhere bounds

$$g^-(t) := \sum_{i\neq j}\sum g_{ij}\big(t,\zeta_{ij}(t)\big)\wedge 0 \le \sum_{i\neq j}\sum g_{ij}\big(t,\zeta_{ij}(t)\eta^{(\epsilon)}(t)\big) \le \sum_{i\neq j}\sum s_{ij}(t).$$

The right-hand side lies in $L^1(0,T)$ by the assumption that $\mathcal{J}_{(0,T)}(c,\mathsf{w}) < \infty$, and the left-hand as well since $\|g^-(t)\|_{L^1(0,T)} \le \mathcal{J}_{(0,T)} - G(c,\dot{\mathsf{w}},\zeta) < \infty$ where $G(c,\dot{\mathsf{w}},\zeta) > -\infty$ as ζ is bounded. Clearly $\zeta\eta^{(\epsilon)}$ converges pointwise to ζ; by dominated convergence it follows that $G(c,\dot{\mathsf{w}},\zeta\eta^{(\epsilon)}) \to G(c,\dot{\mathsf{w}},\zeta)$, which was to be proven. □

The functional $G(c,\dot{\mathsf{w}},\zeta)$ is generally not hybrid-continuous in (c,w), but all we will need is the following:

Lemma 4.5. *For any $\zeta \in C^2_c\big([0,T);\mathfrak{l}^\infty(I\times I)\big)$, the functional $(c,\mathsf{w}) \mapsto G(c,\dot{\mathsf{w}},\zeta)$ is hybrid-continuous on $\{\mathcal{J}_{(0,T)} < \infty\}$.*

Proof. Paths for which $\mathcal{J}_{(0,T)}$ is finite satisfy the initial condition $\big(c(0),\mathsf{w}(0)\big) = (\mu,0)$. As ζ is differentiable and 0 at the right boundary we can write:

$$G(c,\dot{\mathsf{w}},\zeta) = -{}_{L^\infty}\langle\dot\zeta,\mathsf{w}\rangle_{L^1} - \sum_{i\neq j}\sum\int_0^T c_i(t)\mathsf{Q}_{ij}(e^{\zeta_{ij}(t)}-1)\,dt,$$

which is strongly continuous in $L^1\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ and hence also hybrid-continuous. □

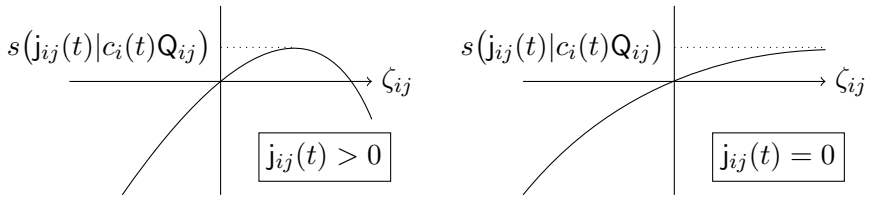

Figure 1: The function $g_{ij}(t, \zeta_{ij})$.

4.2 Exponential tightness

As mentioned in Section 2, with the choice of the hybrid topology, the proof for the exponential tightness becomes very simple. Define the total-variation balls:

$$B_r := \big\{ \big(c, \mathsf{w}\big) \in \mathrm{BV}_{\mathrm{flux}} : \mathrm{epvar}(\mathsf{w}) \le r \big\}. \tag{21}$$

Note that by (9) the variation $\mathrm{epvar}(c)$ is automatically uniformly bounded within such balls.

Proposition 4.6 (Exponential tightness)**.** *Let $u \in L^\infty\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$. For any $\eta > 0$, and for $r := \eta + T\,e\|u\|_{L^\infty}|\mathsf{Q}|_1$,*

$$\mathbb{P}_u^{(n)}(B_r^{\mathsf{c}}) \le e^{-n\eta},$$

and the balls B_r are hybrid-compact in $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$.

Proof. Observe that the perturbed Markov jump process $\sum\sum_{i\neq j}\mathsf{W}_{i,j}^{(n)}(t)$ is bounded by a Poisson process $\frac{1}{n}N_\lambda$ with intensity $\lambda := n\|u\|_{L^\infty}|\mathsf{Q}|_1 \ge n\sum\sum_{i\neq j}c_i(t)\mathsf{Q}_{ij}u_{ij}(t)$. A standard Chernoff bound therefore yields

$$\mathbb{P}^{(n)}(B_r^{\mathsf{c}}) = \mathbb{P}^{(n)}\big(\big\{\tfrac{1}{n}\#\,\mathrm{jump}(\mathsf{W}^{(n)}) > r\big\}\big) \le \mathrm{Prob}\,\big(N_\lambda > nr\big) \le e^{\lambda T e - nr} = e^{-n\eta}.$$

Moreover, by Proposition 2.2 the balls B_r are automatically hybrid-compact. □

4.3 Lower bound

We now prove the large-deviation lower bound. As usual in dynamic large deviations, the proof is based on a Girsanov transformation together with an approximation argument.

Lemma 4.7 (Girsanov transformation)**.** *Fix an $n \in \mathbb{N}$, $C^{(n)}(0) \in \mathcal{P}(I)\cap\frac{1}{n}\mathbb{N}^I$ and a function $\zeta \in C_b^2\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$. Let $\mathbb{P}^{(n)}_{e^\zeta,r}$ be the path measure for the process with initial conditions $(C^{(n)}(0),0)$ and generator*

$$(\mathcal{Q}^{(n)}_{e^{\zeta(t)},r}\phi)(c,\mathsf{w}) := n\sum\sum_{i\neq j} c_i\mathsf{Q}_{ij}e^{\zeta_{ij}(t)\mathbb{1}_{\{\mathsf{w}_{ij}(t)\le r-\frac{1}{n}\}}}\big[\phi(c^{i\to j},\mathsf{w}^{ij+}) - \phi(c,\mathsf{w})\big].$$

Then

$$\frac{1}{n}\log\frac{d\mathbb{P}^{(n)}_{e^\zeta,r}}{d\mathbb{P}^{(n)}}(c,\mathsf{w}) = -G_r(c,\mathsf{w},\zeta), \tag{22}$$

where

$$G_r(c,\mathsf{w},\zeta) := \sum\sum_{i\neq j}\int_0^{\inf\{\hat t\in(0,T):\mathsf{w}_{ij}(\hat t)\ge r\}}\Big[\zeta_{ij}(t)\dot{\mathsf{w}}_{ij}(dt) - c_i(t)\mathsf{Q}_{ij}\big(e^{\zeta_{ij}}-1\big)\,dt\Big].$$

Proof. We can apply the Girsanov Theorem for jump processes [KL99, Ch. A.1, Th. 7.3] to the functional $F_t(\mathsf{w}) := n\,\zeta(t)\cdot(\mathsf{w}\wedge r)$ for $\mathsf{w}\in\mathfrak{l}^1(I\times I)$, which lies in $C_b^2\big(0,T;L^\infty(\mathfrak{l}^1(I\times I))\big)$. □

Remark 4.8. The right-hand side of (22) does not involve the particle number n due to the independence of the particles. □

The next lemma shows that the following set constitutes of sufficiently regular paths that can be retrieved via the Girsanov transformation:

$$\mathcal{A} := \Big\{(c,\mathsf{w})\in W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big) : \mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty \quad\text{and}\\ t\mapsto \log\frac{\dot{\mathsf{w}}(t)}{c(t)\otimes\mathsf{Q}}\in C_c^2\big([0,T);\mathfrak{l}^\infty(I\times I)\big)\Big\}. \tag{23}$$

Lemma 4.9. *For any hybrid-open set* $\mathcal{U}\subset \mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *and any* $(c,\mathsf{w})\in\mathcal{U}\cap\mathcal{A}$,

$$\liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{U})\geq -\mathcal{J}_{(0,T)}(c,\mathsf{w}).$$

Proof. Let a hybrid-open $\mathcal{U}\subset \mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ and a $(c,\mathsf{w})\in\mathcal{U}\cap\mathcal{A}$ be given, and define $\zeta(t) := t\mapsto \log\frac{\dot{\mathsf{w}}(t)}{c(t)\otimes\mathsf{Q}}\in C_c^2\big([0,T);\mathfrak{l}^\infty(I\times I)\big)$. For an arbitrary $\epsilon>0$, let

$$\mathcal{U}_\epsilon(c,\mathsf{w}) := \big\{(\hat c,\hat{\mathsf{w}})\in \mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big) : G(\hat c,\hat{\mathsf{w}},\zeta)<G(c,\dot{\mathsf{w}},\zeta)+\epsilon\big\}.$$

By applying the transformation Lemma 4.7,

$$\begin{aligned}
\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{U}) &\geq \frac{1}{n}\log\mathbb{P}^{(n)}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n\big)\\
&\geq \frac{1}{n}\log\mathbb{P}^{(n)}_{e^\zeta,n}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n\big) \qquad (24)\\
&\qquad + \frac{1}{n}\log \mathop{\mathbb{P}^{(n)}\text{-}\operatorname{ess\,inf}}_{(\hat c,\hat{\mathsf{w}})\in\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n}\frac{d\mathbb{P}^{(n)}}{d\mathbb{P}^{(n)}_{e^\zeta,n}}(\hat c,\hat{\mathsf{w}})\\
&\geq \frac{1}{n}\log\mathbb{P}^{(n)}_{e^\zeta,n}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n\big) - \sup_{(\hat c,\hat{\mathsf{w}})\in\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n} G_n(\hat c,\hat{\mathsf{w}},\zeta)\\
&\geq \frac{1}{n}\log\mathbb{P}^{(n)}_{e^\zeta}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n\big) - \sup_{(\hat c,\hat{\mathsf{w}})\in\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n} G(\hat c,\hat{\mathsf{w}},\zeta). \qquad (25)
\end{aligned}$$

To bound the first term, observe that the perturbed limit equation (14) with $u=e^\zeta=\dot{\mathsf{w}}/(c\otimes\mathsf{Q})$ yields the given path (c,w). Hence by the many-particle limit (Theorem 3.3), $\mathbb{P}^{(n)}_{e^\zeta}\rightharpoonup\delta_{(c,\mathsf{w})}$. Furthermore, because of Lemma 4.5 the sets $\mathcal{U}_\epsilon(c,\mathsf{w})$ are hybrid-open, so that the Portemanteau Theorem gives $\liminf_n\mathbb{P}^{(n)}_{e^\zeta}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\big)\geq 1$. Then we also have, exploiting the exponential tightness (Proposition 4.6),

$$\liminf_{n\to\infty}\mathbb{P}^{(n)}_{e^\zeta}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n\big)\geq\liminf_{n\to\infty}\Big[\mathbb{P}^{(n)}_{e^\zeta}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\big)-\mathbb{P}^{(n)}_{e^\zeta}\big(B_n^{\mathsf{c}}\big)\Big]\geq 1,$$

which shows that the first term in (25) vanishes in the limes inferior.

For the second term, we use the definition of the set $\mathcal{U}_\epsilon(c,\mathsf{w})$ together with the fact that ζ maximises $G(c,\dot{\mathsf{w}},\cdot)$. Putting everything together we find:

$$\begin{aligned}\liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{U}) &\geq \liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}_{e^\zeta}\big(\mathcal{U}\cap\mathcal{U}_\epsilon(c,\mathsf{w})\cap B_n\big) - G(c,\dot{\mathsf{w}},\zeta)-\epsilon\\ &\geq -G(c,\dot{\mathsf{w}},\zeta)-\epsilon = \mathcal{J}_{(0,T)}(c,\mathsf{w})-\epsilon.\end{aligned}$$

Since ϵ was arbitrary the claim follows. □

The next three approximation lemmas show that the rate fuctional can be approximated by taking paths in $\mathcal{A}$.

Lemma 4.10 (Lower bound approximation I)**.** *Let* $(c,\mathsf{w})\in W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *with* $\mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty$*. Then there exists a sequence* $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})_{0<\epsilon<\hat\epsilon}\subset W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *such that* $\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})\xrightarrow[\epsilon\to0]{}\mathcal{J}_{(0,T)}(c,\mathsf{w})$ *and* $\|\dot{\mathsf{w}}^{(\epsilon)}_{ij}\|_{L^1(0,T)}=0\implies\|c_i^{(\epsilon)}\mathsf{Q}_{ij}\|_{L^1(0,T)}=0$ *for all* $i\neq j\in I\times I$ *and for all* $0<\epsilon<\hat\epsilon$*.*

Proof. If $\|\dot{\mathsf{w}}_{ij}\|_{L^1(0,T)}=0\implies\|c_i\mathsf{Q}_{ij}\|_{L^1(0,T)}=0$ for all $i,j\in I$ then the construction is trivial. Now assume that there is one and only one pair $i\neq j\in I\times I$ for which $\|c_i\|_{L^1}|\mathsf{Q}_{ij}|=\|c_i\mathsf{Q}_{ij}\|_{L^1}>0$ but $\|\dot{\mathsf{w}}_{ij}\|_{L^1}=0$. If there would be more pairs with this property then we can simply repeat the construction below for each such pair separately.

Since $\mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty$, by Proposition 4.3 the path c must be continuous, and so there exists an $\hat\epsilon>0$ and a $\hat t\in(0,T)$ such that, see Figure 2:

$$0\leq(t-\hat t)\mathbb{1}_{(\hat t,\hat t+\epsilon)}(t)\leq(t-\hat t)\mathbb{1}_{(\hat t,\hat t+\hat\epsilon)}(t)\leq c_i(t)-\hat\epsilon\qquad\text{for all }0<\epsilon<\hat\epsilon.$$

Define the sequence:

$$\begin{aligned}&\mathsf{w}^{(\epsilon)}(t):=\mathsf{w}(t)+(t-\hat t)\mathbb{1}_{(\hat t,\hat t+\epsilon)}(t)\mathbb{1}_{ij},\quad\text{and}\\ &c^{(\epsilon)}(t):=\mu-\operatorname{div}\mathsf{w}^{(\epsilon)}(t)=c(t)-(t-\hat t)\mathbb{1}_{(\hat t,\hat t+\epsilon)}(t)\mathbb{1}_i,\end{aligned}$$

and note that by construction $c^{(\epsilon)}(0)=\mu$, the continuity equation is satisfied, and we have that both $\dot{\mathsf{w}}^{(\epsilon)}_{ij}(t)>0$ and $c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}>0$, as required. To prove the convergence of the rate functional, we will use formulation (5) which is equal to $\mathcal{J}_{(0,T)}$ by Proposition 4.4. Recall that $\dot{\mathsf{w}}_{ij}\equiv0$ and note that the continuous function $c_i(t)$ is uniformly bounded by some $C>0$ on the compact interval $[\hat t,\hat t+\hat\epsilon]$, so that $0<\hat\epsilon\leq c_i^{(\epsilon)}(t)=c_i(t)-t+\hat t\leq C$ on $[\hat t,\hat t+\hat\epsilon]$. It follows that

$$\begin{aligned}&\big|\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})-\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})\big|\\ &\qquad\leq\int_{\hat t}^{\hat t+\epsilon}\Big|1\log\frac{1}{c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}}-1+c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}-c_i(t)\mathsf{Q}_{ij}\Big|\,dt\\ &\qquad\leq\int_{\hat t}^{\hat t+\epsilon}\Big(\big|\log(c_i^{(\epsilon)}(t)\mathsf{Q}_{ij})\big|+1+\hat t-t\Big)\,dt\to0.\end{aligned}$$

□

Lemma 4.11 (Lower bound approximation II)**.** *Let* $(c,\mathsf{w})\in W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *with* $\mu>0$*,* $\mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty$*, and* $\|\dot{\mathsf{w}}_{ij}\|_{L^1(0,T)}=0\implies\|c_i\mathsf{Q}_{ij}\|_{L^1(0,T)}=0$ *for all* $i\neq j\in I\times I$*. Then there exists a sequence* $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})_{\epsilon>0}\subset W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *such that* $\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})\xrightarrow[\epsilon\to0]{}\mathcal{J}_{(0,T)}(c,\mathsf{w})$ *and* $\log\frac{\dot{\mathsf{w}}^{(\epsilon)}}{c^{(\epsilon)}\otimes\mathsf{Q}}\in C^2_b\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$ *for all* $\epsilon>0$ *(with the convention that* $\log\frac{0}{0}\equiv0$*).*

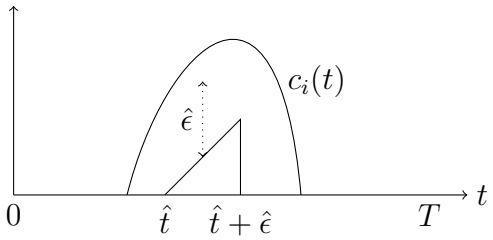

Figure 2: There exists a small triangle below the graph of $c_i(t)$.

Proof. Let $\theta^{(\epsilon)}$ be the heat kernel (20) and define the approximating sequence (coordinate-wise):

$$\mathsf{w}^{(\epsilon)}(t) := \frac{\mu}{(c*\theta^{(\epsilon)})(0)}\big[(\mathsf{w}*\theta^{(\epsilon)})(t)-(\mathsf{w}*\theta^{(\epsilon)})(0)\big], \qquad \text{and}$$
$$c^{(\epsilon)}(t) := \mu - \operatorname{div}\mathsf{w}^{(\epsilon)}(t) = \frac{\mu}{(c*\theta^{(\epsilon)})(0)}(c*\theta^{(\epsilon)})(t). \tag{26}$$

Here the convolutions run over the whole real line, where we extended $(c(t),\mathsf{w}(t)) = (\mu,0)$ for $t<0$ and $\big(c(t),\mathsf{w}(t)\big) = \big(c(T),\mathsf{w}(T)\big)$ for $t>T$; these values are well-defined since functions of bounded variation have left and right limits. Observe that by construction, the initial condition $\big(c^{(\epsilon)}(0),\mathsf{w}^{(\epsilon)}(0)\big) = (\mu,0)$ and the continuity equation are satisfied, $c^{(\epsilon)}(t)$ and $\dot{\mathsf{w}}^{(\epsilon)}(t)$ are non-negative, and so is $\mathsf{w}^{(\epsilon)}(t)$.

We first prove that $\log\frac{\dot{\mathsf{w}}^{(\epsilon)}_{ij}}{c_i^{(\epsilon)}\otimes\mathsf{Q}_{ij}} \in C_b^2(0,T)$ for all $\epsilon>0$ and $i,j\in I$. We distinguish between two cases. If the path $c_i(t)\mathsf{Q}_{ij}=0$ for almost every $t\in(0,T)$, then also $\dot{\mathsf{w}}_{ij}(t)=0$ for almost every t, since $\mathcal{J}_{(0,T)}<\infty$ implies $\dot{\mathsf{w}}(t)\ll c(t)\otimes\mathsf{Q}$. Then we also have $c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}=0$ and $\mathsf{w}_{ij}^{(\epsilon)}(t)=0$ for almost every $t\in(0,T)$, and hence $\log\frac{\dot{\mathsf{w}}^{(\epsilon)}_{ij}}{c_i^{(\epsilon)}\mathsf{Q}_{ij}} = \log\frac{0}{0} := 0 \in C_b^2(0,T)$. For the second case we can assume that $\|c_i\|_{L^1(0,T)}>0$ and $\mathsf{Q}_{ij}>0$, and so by the main assumption of the lemma also $\mathsf{w}_{ij}(T)=\|\dot{\mathsf{w}}_{ij}\|_{L^1(0,T)}>0$. Clearly $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})\in C_b^\infty\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^\infty(I\times I)\big)$, and in particular, the function and all its derivatives are uniformly bounded. More precisely, we have the following bounds from below, uniformly in t:

$$c_i^{(\epsilon)}(t) \ge \frac{\mu}{(c*\theta^{(\epsilon)})(0)}\theta^{(\epsilon)}(T)\|c_i\|_{L^1(0,T)}>0 \qquad \text{and}$$
$$\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t) \ge \frac{\mu}{(c*\theta^{(\epsilon)})(0)}\theta^{(\epsilon)}(T)\mathsf{w}_{ij}(T)>0. \tag{27}$$

Therefore the three functions

$$\log\frac{\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t)}{c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}}$$
$$\frac{d}{dt}\log\frac{\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t)}{c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}} = \frac{\ddot{\mathsf{w}}_{ij}^{(\epsilon)}(t)}{\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t)} - \frac{\dot c_i^{(\epsilon)}(t)}{c_i^{(\epsilon)}(t)}$$
$$\frac{d^2}{dt^2}\log\frac{\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t)}{c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}} = \frac{\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t)\dddot{\mathsf{w}}_{ij}^{(\epsilon)}(t)-\ddot{\mathsf{w}}_{ij}^{(\epsilon)}(t)^2}{\dot{\mathsf{w}}_{ij}^{(\epsilon)}(t)^2} - \frac{c_i^{(\epsilon)}(t)\ddot c_i^{(\epsilon)}(t)-\dot c_i^{(\epsilon)}(t)^2}{c_i^{(\epsilon)}(t)^2}$$

are all bounded and continuous which was to be shown.

We now show that $\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)}) \xrightarrow[\epsilon\to 0]{} \mathcal{J}_{(0,T)}(c,\mathsf{w})$. Because $(c,\mathsf{w}) \in W^{1,1}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$ we surely have $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)}) \xrightarrow[\epsilon\to 0]{W^{1,1}} (c,\mathsf{w})$. Since $\mathcal{J}_{(0,T)}$ is the supremum over $W^{1,1}$-continuous functionals, $\mathcal{J}_{(0,T)}$ is lower semi-continuous, and so

$$\liminf_{\epsilon\to 0} \mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)}) \geq \mathcal{J}_{(0,T)}(c,\mathsf{w}). \tag{28}$$

We use the relative entropy formulation of $\mathcal{J}_{(0,T)}$ to prove the inequality in the other direction. Since $\mathcal{S}(\cdot|\cdot)$ is jointly convex one finds by a two-dimensional Jensen inequality that

$$\begin{aligned}
&\int_0^T \mathcal{S}\big(\dot{\mathsf{w}}^{(\epsilon)}(t)|c^{(\epsilon)}(t)\otimes \mathsf{Q}\big)\,dt\\
&= \frac{\mu}{(c*\theta^{(\epsilon)})(0)} \int_0^T \mathcal{S}\Big(\int_{-\infty}^{\infty} \dot{\mathsf{w}}(t-s)\,\theta^{(\epsilon)}(s)\,ds \,\Big|\, \int_{-\infty}^{\infty} c(t-s)\theta^{(\epsilon)}(s)\,ds \otimes \mathsf{Q}\Big)\,dt\\
&\leq \frac{\mu}{(c*\theta^{(\epsilon)})(0)} \int_0^T \int_{-\infty}^{\infty} \mathcal{S}\big(\dot{\mathsf{w}}(t-s)|c(t-s)\otimes\mathsf{Q}\big)\theta^{(\epsilon)}(s)\,ds\,dt\\
&\xrightarrow[\epsilon\to 0]{} \int_0^T \mathcal{S}\big(\dot{\mathsf{w}}(t)|c(t)\otimes\mathsf{Q}\big)\,dt = \mathcal{J}_{(0,T)}(c,\mathsf{w}),
\end{aligned}$$

where the convergence follows from $(c*\theta^{(\epsilon)})(0)\to\mu$ together with the fact that the non-negative mapping $t\mapsto \mathcal{S}(\dot{\mathsf{w}}(t)|c(t)\otimes\mathsf{Q})$ lies in $L^1(0,T)$ since $\mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty$ (see for example [Eva98, App. C.4, Th. 6]). □

Lemma 4.12 (Lower bound approximation III)**.** *Let* $(c,\mathsf{w}) \in W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *with* $\mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty$ *and* $\log\frac{\dot{\mathsf{w}}}{c\otimes\mathsf{Q}} \in C_b^2\big(0,T;\mathfrak{l}^\infty(I\times I)\big)$ *(with the convention that* $\log\frac{0}{0}\equiv 0$*). Then there exists a sequence* $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})_{\epsilon>0}\subset W^{1,1}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ *such that* $\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})\to \mathcal{J}_{(0,T)}(c,\mathsf{w})$ *and* $\log\frac{\dot{\mathsf{w}}^{(\epsilon)}}{c^{(\epsilon)}\otimes\mathsf{Q}} \in C_c^2\big([0,T);\mathfrak{l}^\infty(I\times I)\big)$ *for all* $\epsilon>0$.

Proof. Let $\psi(t)$ be the solution map from Proposition 3.2(ii), (where the perturbation factor $u\equiv 0$), and define

$$(\hat{c}^{(\epsilon)}(t),\hat{\mathsf{w}}^{(\epsilon)}(t)) := \begin{cases} (c(t),\mathsf{w}(t)), & t\in(0,T-\epsilon),\\ \psi(t-T+\epsilon)\big[c(T-\epsilon),\mathsf{w}(T-\epsilon)\big], & t\in(T-\epsilon,T).\end{cases}$$

Then clearly $\log\frac{\dot{\hat{\mathsf{w}}}^{(\epsilon)}}{\hat{c}^{(\epsilon)}\otimes\mathsf{Q}}$ has C_b^2-regularity on $(0,T-\epsilon]$, and it is constant 0 on $(T-\epsilon,T)$. To deal with the lack of regularity at time $t=T-\epsilon$, we again mollify like in (26), but now with smooth, compactly supported bump functions $\eta^{(\epsilon)}\in C_c^\infty(-\frac{1}{2}\epsilon,\frac{1}{2}\epsilon)$, $\int\eta^{(\epsilon)}(t)\,dt=1$, i.e.

$$\begin{aligned}
\mathsf{w}^{(\epsilon)}(t) &:= \frac{\mu}{(\hat{c}^{(\epsilon)}*\eta^{(\epsilon)})(0)}\big[(\hat{\mathsf{w}}^{(\epsilon)}*\eta^{(\epsilon)})(t) - (\hat{\mathsf{w}}^{(\epsilon)}*\eta^{(\epsilon)})(0)\big], \qquad \text{and}\\
c^{(\epsilon)}(t) &:= \mu - \operatorname{div}\mathsf{w}^{(\epsilon)}(t).
\end{aligned}$$

Due to the small compact support, we still have that $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})$ follows the macroscopic flow on $(T-\frac{1}{2}\epsilon,T)$, and so $\log\frac{\dot{\mathsf{w}}^{(\epsilon)}}{c^{(\epsilon)}\otimes\mathsf{Q}}$ has the desired regularity.

We now prove that the rate functional converges. The convolution with the smooth kernel can be dealt with in exactly the same manner as in the proof of Lemma 4.11, so we only need to prove the convergence of $\mathcal{J}_{(0,T)}(\hat{c}^{(\epsilon)},\hat{\mathsf{w}}^{(\epsilon)})$. This follows immediately by monotone convergence:

$$\mathcal{J}_{(0,T)}(\hat{c}^{(\epsilon)},\hat{\mathsf{w}}^{(\epsilon)}) = \int_0^{T-\epsilon} \mathcal{S}\big(\dot{\hat{\mathsf{w}}}^{(\epsilon)}(t)|\hat{c}^{(\epsilon)}(t)\otimes\mathsf{Q}\big)\,dt \to \int_0^T \mathcal{S}\big(\dot{\hat{\mathsf{w}}}^{(\epsilon)}(t)|\hat{c}^{(\epsilon)}(t)\otimes\mathsf{Q}\big)\,dt.$$

□

Proposition 4.13 (Large-deviation lower bound)**.** *Assume $\mu > 0$. For any hybrid-open set $\mathcal{U} \subset \mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$,*

$$\liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{U}) \geq -\inf_{(c,\mathsf{w})\in\mathcal{U}}\mathcal{J}_{(0,T)}(c,\mathsf{w}).$$

Proof. Take an arbitrary hybrid-open $\mathcal{U} \subset \mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$. As a consequence of Lemmas 4.10, 4.11 and 4.12, any path with $\mathcal{J}_{(0,T)}(c,\mathsf{w})<\infty$ can be approximated by paths in the set $\mathcal{A}$ from (23) such that the rate functional also converges. In particular, due to this denseness, the set $\mathcal{U}\cap\mathcal{A}$ is never empty. Combining this approximation with Lemma 4.9 yields

$$\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{U}) \geq -\inf_{(c,\mathsf{w})\in\mathcal{U}\cap\mathcal{A}}\mathcal{J}_{(0,T)}(c,\mathsf{w}) = -\inf_{(c,\mathsf{w})\in\mathcal{U}}\mathcal{J}_{(0,T)}(c,\mathsf{w}).$$

□

4.4 Upper bound

We now prove the large-deviation weak upper bound via a standard covering technique.

Proposition 4.14. *For any compact set $\mathcal{K}\subset\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ and any $r>0$,*

$$\liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}\big(\mathcal{K}\cap B_r\big) \leq -\inf_{(c,\mathsf{w})\in\mathcal{K}\cap B_r}\mathcal{J}_{(0,T)}(c,\mathsf{w}).$$

where B_r is the bounded-variation ball (21)*.*

Proof. Fix an $\epsilon>0$, and observe that because of (18) one can find for any $(c,\mathsf{w})\in\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ a $\zeta[c,\mathsf{w}]\in C_c^2\big([0,T);\mathfrak{l}^\infty(I\times I)\big)$ such that $G(c,\dot{\mathsf{w}},\zeta[c,\mathsf{w}])\geq\mathcal{J}_{(0,T)}(c,\mathsf{w})-\epsilon$. Now define for each $(c,\mathsf{w})\in\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ the sets

$$\mathcal{V}_\epsilon(c,\mathsf{w}) := \Big\{(\tilde c,\tilde{\mathsf{w}})\in\mathrm{BV}_{\mathrm{flux}} : G(\tilde c,\tilde{\mathsf{w}},\zeta[c,\mathsf{w}])>G(c,\dot{\mathsf{w}},\zeta[c,\mathsf{w}])-\epsilon\Big\},$$

which are open by Lemma 4.5. Surely $\bigcup_{(c,\mathsf{w})\in\mathcal{K}}\mathcal{V}_\epsilon(c,\mathsf{w})\supset\mathcal{K}$ and so by compactness there exists a finite subcovering $\cup_{k=1}^K\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\supset\mathcal{K}$. We can then use Lemma 4.7 together with the fact that $G_r=G$ on B_r, to estimate for every $k=1,\dots,K$,

$$\begin{aligned}
&\limsup_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}\big(\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\cap B_r\big)\\
&\qquad\leq \limsup_{n\to\infty}\frac{1}{n}\underbrace{\log\mathbb{P}^{(n)}_{e^{\zeta[c^{(k)},\mathsf{w}^{(k)}]},r}\big(\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\cap B_r\big)}_{\leq 0}\\
&\qquad\qquad + \frac{1}{n}\log\mathbb{P}^{(n)}\text{-}\operatorname*{ess\,sup}_{(\tilde c,\tilde{\mathsf{w}})\in\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\cap B_r}\frac{d\mathbb{P}^{(n)}}{d\mathbb{P}^{(n)}_{e^{\zeta[c^{(k)},\mathsf{w}^{(k)}]},r}}(\tilde c,\tilde{\mathsf{w}})\\
&\qquad\leq -\inf_{(\tilde c,\tilde{\mathsf{w}})\in\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\cap B_r}G_r(\tilde c,\tilde{\mathsf{w}},\zeta[c^{(k)},\mathsf{w}^{(k)}])\\
&\qquad= -\inf_{(\tilde c,\tilde{\mathsf{w}})\in\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\cap B_r}G(\tilde c,\tilde{\mathsf{w}},\zeta[c^{(k)},\mathsf{w}^{(k)}])\\
&\qquad\leq -G(c^{(k)},\dot{\mathsf{w}}^{(k)},\zeta[c^{(k)},\mathsf{w}^{(k)}])\chi_{(c^{(k)},\mathsf{w}^{(k)})}(B_r)+\epsilon\\
&\qquad\leq -\mathcal{J}_{(0,T)}(c^{(k)},\mathsf{w}^{(k)})\chi_{(c^{(k)},\mathsf{w}^{(k)})}(B_r)+2\epsilon,
\end{aligned}$$

with the usual characteristic function: $\chi_{(c^{(k)},\mathsf{w}^{(k)})}(B_r) = 0$ for $(c^{(k)},\mathsf{w}^{(k)}) \in B_r$ and ∞ otherwise. Due to the finiteness of the covering one can use the Laplace Principle to get:

$$\begin{aligned}\limsup_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{K}\cap B_r) &\le \max_{k=1,\dots,K}\limsup_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}\big(\mathcal{V}_\epsilon(c^{(k)},\mathsf{w}^{(k)})\cap B_r\big)\\ &\le \max_{k=1,\dots,K} -\mathcal{J}_{(0,T)}(c^{(k)},\mathsf{w}^{(k)})\chi_{(c^{(k)},\mathsf{w}^{(k)})}(B_r)+2\epsilon\\ &\le -\inf_{(c,\mathsf{w})\in\mathcal{K}\cap B_r}\mathcal{J}_{(0,T)}(c^{(k)},\mathsf{w}^{(k)})+2\epsilon.\end{aligned}$$

Since ϵ was arbitrary, this proves the claim. □

Corollary 4.15 (Large-deviation weak upper bound)**.** *For any compact set* $\mathcal{K}\subset \mathrm{BV}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$,

$$\liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{K}) \le -\inf_{(c,\mathsf{w})\in\mathcal{K}}\mathcal{J}_{(0,T)}(c,\mathsf{w}).$$

Proof. This is a consequence of the exponential tightness Proposition 4.6 and Proposition 4.14, as follows. Note that the balls B_r are defined as subsets of $\mathrm{BV}_{\mathrm{flux}}$, and that $\mathcal{J}_{(0,T)}|_{\mathrm{BV}_{\mathrm{flux}}^{\mathsf{c}}}=\infty$. For any $\eta>0$ and $r:=\eta+T\,e|\mathsf{Q}|_1$ we can apply Laplace's principle:

$$\begin{aligned}\limsup_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{K}) &\le \limsup_{n\to\infty}\frac{1}{n}\log\big(\mathbb{P}^{(n)}(\mathcal{K}\cap B_r)+\mathbb{P}^{(n)}(B_r^{\mathsf{c}})\big)\\ &\le -\big(\inf_{\mathcal{K}\cap B_r}\mathcal{J}_{(0,T)}\wedge\eta\big)\le -\big(\inf_{\mathcal{K}}\mathcal{J}_{(0,T)}\wedge\eta\big).\end{aligned}$$

Since η was arbitrary the claim follows. □

4.5 Coupled large deviations

We can now use the conditional large deviations to prove the coupled large deviations.

Proof of Theorem 4.2. The coupled probabilities can be disintegrated as

$$\begin{aligned}&\mathrm{Prob}\big(C^{(n)},\mathsf{W}^{(n)}\in dc\,d\mathsf{w}\big)\\ &\qquad=\int\mathrm{Prob}\big(C^{(n)},\mathsf{W}^{(n)}\in dc\,d\mathsf{w}\mid C^{(n)}(0)=c(0)\big)\,\mathrm{Prob}\big(C^{(n)}(0)\in dc(0)\big),\end{aligned}$$

where initial probabilities satisfy a large-deviation principle with rate $\mathcal{I}_0$, and the conditional probabilities satisfy a large-deviation principle with rate $\mathcal{J}_{(0,T)}$ whenever $c(0)>0$. Therefore, apart from the condition $c(0)>0$, we can immediately apply [Big04] to get the large deviations for the coupled system. Observe that the condition $c(0)>0$ is not needed in the conditional upper bound, Corollary 4.15, and hence by [Big04] the coupled large-deviation upper bound holds. However, for the lower bound we only get for any hybrid-open set $\mathcal{U}\subset\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ that

$$\liminf_{n\to\infty}\frac{1}{n}\log\mathbb{P}^{(n)}(\mathcal{U})\ge -\inf_{\substack{(c,\mathsf{w})\in\mathcal{U}:\\ c(0)>0}}\mathcal{I}_0\big(c(0)\big)+\mathcal{J}_{(0,T)}(c,\mathsf{w}).$$

In order to replace the infimum above by $-\inf_{(c,\mathsf{w})\in\mathcal{U}}\mathcal{I}_0\big(c(0)\big)+\mathcal{J}_{(0,T)}(c,\mathsf{w})$, we need to show that any $(c,\mathsf{w})\in\mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ can be approximated by trajectories $(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})$ with $c^{(\epsilon)}(0)>0$ such that $\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)})\to\mathcal{J}_{(0,T)}(c,\mathsf{w})$.

Take an arbitrary $(c,\mathsf{w}) \in \mathrm{BV}\big(0,T;\mathcal{P}(I)\times\mathfrak{l}^1(I\times I)\big)$ where $c(0)$ lies on the boundary $\{c\in\mathcal{P}(I): c_i=0 \text{ for some } i\in I\}$. We apply two approximations. The first approximation is similar to (26) but slightly different since $c(0)$ might be zero:

$$\begin{aligned}\mathsf{w}^{(\epsilon)}(t) &:= (\mathsf{w}*\theta^{(\epsilon)})(t) - (\mathsf{w}*\theta^{(\epsilon)})(0),\\ \hat{c}^{(\epsilon)}(t) &:= (c*\theta^{(\epsilon)})(0) - \operatorname{div}\mathsf{w}^{(\epsilon)}(t) = (c*\theta^{(\epsilon)})(t),\end{aligned}$$

where as before we extend the functions constantly outside $(0,T)$. By the same argument as in Lemma 4.11 we have $\mathcal{J}_{(0,T)}(\hat{c}^{(\epsilon)},\mathsf{w}^{(\epsilon)}) \to \mathcal{J}_{(0,T)}(c,\mathsf{w})$; we will use this below. Moreover by the assumed continuity on the boundary we also have $\mathcal{I}_0\big(\hat{c}^{(\epsilon)}(0)\big) = \mathcal{I}_0\big((c*\theta^{(\epsilon)})(0)\big) \to \mathcal{I}_0\big(c(0)\big)$.

For the second approximation, let $\hat{i}\in I$ be such that $c_{\hat{i}}(0)>0$. For this coordinate, we have, similarly to (27), the lower bound $\hat{c}^{(\epsilon)}_{\hat{i}}(t) \geq \theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)} > 0$. We can then define

$$\begin{aligned}c^{(\epsilon)}(t) &:= c^{(\epsilon)}(0) - \operatorname{div}\mathsf{w}^{(\epsilon)}(t), \qquad \text{where}\\ c_i^{(\epsilon)}(0) &:= \begin{cases}\hat{c}^{(\epsilon)}_{\hat{i}}(0) - \epsilon\theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)}, & i=\hat{i},\\ \hat{c}^{(\epsilon)}_i(0) + \frac{\epsilon}{|I|-1}\theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)}, & i\neq\hat{i}.\end{cases}\end{aligned}$$

Observe that by construction $c^{(\epsilon)}(0)>0$ and $c^{(\epsilon)}(t)$ remains positive. Moreover $c^{(\epsilon)}(0)\to c(0)$ and hence $\mathcal{I}_0\big(c^{(\epsilon)}(0)\big)\to\mathcal{I}_0\big(c(0)\big)$. To prove convergence of $\mathcal{J}_{(0,T)}(c^{(\epsilon)})$, we exploit the lower semicontinuity as in (28); hence we only need to prove an upper bound. For any pair $i\neq\hat{i},j$ we get

$$\begin{aligned}&\int_0^T\Big[\dot{\mathsf{w}}^{(\epsilon)}_{ij}(t)\log\frac{\dot{\mathsf{w}}^{(\epsilon)}_{ij}(t)}{\underbrace{c_i^{(\epsilon)}(t)}_{\geq\hat{c}_i^{(\epsilon)}(t)}\mathsf{Q}_{ij}} - \dot{\mathsf{w}}^{(\epsilon)}_{ij}(t) + c_i^{(\epsilon)}(t)\mathsf{Q}_{ij}\Big]\,dt\\ &\quad\leq\int_0^T\Big[\dot{\mathsf{w}}^{(\epsilon)}_{ij}(t)\log\frac{\dot{\mathsf{w}}^{(\epsilon)}_{ij}(t)}{\hat{c}_i^{(\epsilon)}(t)\mathsf{Q}_{ij}} - \dot{\mathsf{w}}^{(\epsilon)}_{ij}(t) + \hat{c}_i^{(\epsilon)}(t)\mathsf{Q}_{ij}\Big]\,dt + \underbrace{\frac{T\epsilon}{(|I|-1)}\theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)}\mathsf{Q}_{ij}}_{\to 0}.\end{aligned}$$

On the other hand, for any pair $\hat{i}\neq j$,

$$\begin{aligned}&\int_0^T\Big[\dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t)\log\frac{\dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t)}{c_{\hat{i}}^{(\epsilon)}(t)\mathsf{Q}_{\hat{i}j}} - \dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t) + c_{\hat{i}}^{(\epsilon)}(t)\mathsf{Q}_{\hat{i}j}\Big]\,dt\\ &\quad=\int_0^T\Big[\dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t)\log\frac{\dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t)}{\hat{c}_{\hat{i}}^{(\epsilon)}(t)\mathsf{Q}_{\hat{i}j}} - \dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t) + \hat{c}_{\hat{i}}^{(\epsilon)}(t)\mathsf{Q}_{\hat{i}j}\\ &\qquad\qquad - \epsilon\theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)}\mathsf{Q}_{\hat{i}j} - \dot{\mathsf{w}}^{(\epsilon)}_{\hat{i}j}(t)\log\Big(1-\frac{\epsilon\theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)}}{\underbrace{\hat{c}_{\hat{i}}^{(\epsilon)}(t)}_{\geq\theta^{(\epsilon)}(T)\|c_{\hat{i}}\|_{L^1(0,T)}}}\Big)\Big]\,dt.\end{aligned}$$

Putting both bounds together yields

$$\limsup_{\epsilon\to0}\mathcal{J}_{(0,T)}(c^{(\epsilon)},\mathsf{w}^{(\epsilon)}) \leq \limsup_{\epsilon\to0}\mathcal{J}_{(0,T)}(\hat{c}^{(\epsilon)},\mathsf{w}^{(\epsilon)}) = \mathcal{J}_{(0,T)}(c,\mathsf{w}),$$

which was to be proven. □

In particular, as a consequence of Theorem 4.2 and Sanov's Theorem, we have:

Corollary 4.16. *Let $C^{(n)}(0) = \frac{1}{n}\sum_{k=1}^n \mathbb{1}_{X_k}$ where $X_1, \dots, X_n$ are i.i.d. according to $\pi \in \mathcal{P}(I)$, the unique invariant measure for the Markov chain with generator Q. Then $(C^{(n)}, \mathsf{W}^{(n)})$ satisfies a large-deviation principle in $\mathrm{BV}\big(0,T;\mathcal{P}(I)\times \mathfrak{l}^1(I\times I)\big)$ with the hybrid topology, with good rate functional $\mathcal{S}\big(c(0)|\pi\big) + \mathcal{J}_{(0,T)}(c,\mathsf{w})$.*

Remark 4.17. The dynamic large deviations could also be proven by first proving the large deviations of the empirical measure on the path space, and then contracting to the path of the empirical measure-integrated flux pair. This is the strategy used in [Fen94, Léo95] to prove large deviations for the empirical measure. □

4.6 Large deviations for the reacting particle model

We now describe how a similar large-deviation principle as Theorem 4.2 can be derived for the more general reacting particle system. The arguments are purely formal and so we will not be concerned too much with the mathematical details. We use the framework as set out by [FK06, § 8.6.1.2], starting from the generator (3).

To this aim we first define the following operator on test functions $\phi \in C_b^1\big(\mathfrak{l}^1(I)\times\mathfrak{l}^1(\mathcal{R})\big)$,

$$(\mathcal{H}^{(V)}\phi)(c,\mathsf{w}) := \frac{1}{V}e^{-V\phi(c,\mathsf{w})}\big(\mathcal{Q}^{(V)}e^{V\phi}\big)(c,\mathsf{w}).$$

It is easily calculated that $\frac{d}{dt}U^{(V)}(t)\phi = \mathcal{H}^{(V)}\big(U^{(V)}(t)\phi\big)$, where

$$\big(U^{(V)}(t)\phi\big)(c,\mathsf{w}) := \frac{1}{V}\log\mathbb{E}\big[e^{V\phi(C^{(V)}(t),\mathsf{W}^{(V)}(t))}|(C^{(V)}(0),\mathsf{W}^{(V)}(0)) = (c,\mathsf{w})\big].$$

In this sense $\mathcal{H}^{(V)}$ is called the *non-linear generator* of the semigroup $U^{(V)}(t)$. This semigroup plays an important role in the theory of large deviations of Markov processes, as the time-dependent version of the functionals that appear in the Varadhan-Bryc Theorem [DZ87, Th. 4.4.13]. We can study the asymptotics of this semigroup via its non-linear generator, formally (recall that $V^{-1}k^{(V,r)} \to \bar{k}^{(r)}$ as $V\to\infty$),

$$\begin{aligned}(\mathcal{H}^{(V)}\phi)(c,\mathsf{w}) &= \frac{1}{V}\sum_{r\in\mathcal{R}} k^{(V,r)}(c)\big[e^{V\phi(c+V^{-1}\gamma^{(r)},\mathsf{w}+V^{-1}\mathbb{1}_r)-V\phi(c,\mathsf{w})}-1\big]\\ &\xrightarrow[V\to\infty]{} \sum_{r\in\mathcal{R}}\bar{k}^{(r)}(c)\big[e^{\nabla_c\phi(c,\mathsf{w})\cdot\gamma^{(r)}+\nabla_{\mathsf{w}}\phi(c,\mathsf{w})\cdot\mathbb{1}_r}-1\big].\end{aligned}$$

Here $\cdot$ now denotes the dual pairing between $\mathfrak{l}^\infty(\mathcal{R})$ and $\mathfrak{l}^1(\mathcal{R})$. To remain consistent with the notation and results in Section 4, we ignore the fluctuations in c (there are none) and focus on the fluctuations in w. This means that can restrict to test functions that depend on w only, as long as we implicitly set $c(t) = c(0) + \Gamma\mathsf{w}(t)$. Then, the last calculation shows that the limit non-linear generator depends on ϕ through $\zeta := \nabla\phi(c,\mathsf{w})$ only; if this would not be true then the many-particle limit would not hold. The limit non-linear generator thus defines the operator that will play the role of a Hamiltonian

$$\mathcal{H}(c,\mathsf{w},\zeta) = \sum_{r\in\mathcal{R}}\bar{k}^{(r)}(c)\big(e^{\zeta_r}-1\big). \tag{29}$$

Naturally, this is the reacting particle equivalent of the operator (17). Then we have the following:

Formal Theorem 4.18 ([FK06, Th. 8.14])**.** *Let the random variables $C^{(V)}(0)$ satisfy a large-deviation principle in $\mathfrak{l}^1(I)$ with rate functional $\mathcal{I}_0$. Then the pair $(C^{(V)}, \mathsf{W}^{(V)})$ satisfies a large-deviation principle with rate functional $(c,\mathsf{w}) \mapsto \mathcal{I}_0\big(c(0)\big) + \mathcal{J}_{(0,T)}(c,\mathsf{w})$, where*

$$\begin{aligned}\mathcal{I}(c,\mathsf{w}) &= \sup_{\zeta\in C_b^2((0,T);\mathfrak{l}^\infty(\mathcal{R}))} \int_0^T \zeta(t)\cdot \dot{\mathsf{w}}(dt) - \int_0^T \mathcal{H}\big(c(t),\mathsf{w}(t),\zeta(t)\big)\,dt \\ &= \int_0^T \mathcal{S}(\dot{\mathsf{w}}(t) \mid \bar{k}(c(t)))\big)\,dt. \end{aligned} \tag{30}$$

and we implicitly set $\mathcal{J}_{(0,T)}(c,\mathsf{w}) = \infty$ whenever $\lim_{t\searrow 0}\mathsf{w} = 0$ or $c(t) = c(0) + \Gamma \mathsf{w}(t)$ is violated.

By a direct calculation analogous to Proposition 4.4, it follows that the dynamic rate is indeed the same as (8).

Remark 4.19. It is very difficult to turn Theorem 4.18 into a rigorous statement. Using the approach from [FK06], one would need to show that the limit non-linear generator also generates a semigroup, which is usually done via viscosity solutions.

Another approach would be to try to generalise the arguments of Section 4. However, the central step, Lemma 4.11, is based on a two-argument Jensen inequality, but the functional $\mathcal{S}\big(\,\cdot\,|\bar{k}(\cdot)\big)$ is now only jointly convex if and only $\bar{k}$ is concave. But even for concave rates $\bar{k}$ it is not at all clear how to mollify such that the test function $\zeta_r(t) := \log \frac{\dot{\mathsf{w}}_r^{(\epsilon)}(t)}{\bar{k}^{(r)}(c^{(\epsilon)}(t))}$ lies in C_b^2, as $\bar{k}$ could be zero, as well as its derivatives along the boundary. □

5 Implications for Macroscopic Fluctuation Theory

Using the large-deviation principle from the previous section, we now explore which parts of MFT can be applied to one-way fluxes on discrete spaces. We will work with the general reacting particle system with generator (3), implicitly assuming that the formal large-deviation Theorem 4.18 holds true. To adopt the usual language of MFT, we consider the integrand of (30) as a Lagrangian:

$$\mathcal{L}(c,\mathsf{j}) := \mathcal{S}\big(\mathsf{j} \mid k(c)\big). \tag{31}$$

This section will be less rigorous; for example we always implicitly assume that curves (c,w) are sufficiently regular so that the rate functional is finite, as in (5). We stress that although the dynamic large-deviation rate $\mathcal{J}_{(0,T)}$ is known and completely specified by $\mathcal{L}$, the quasipotential, i.e. the large-deviation rate $\mathcal{I}_0$ for the invariant measure, is often not known, and we will make no such claims.

The key concept in MFT is time-reversal and corresponding symmetries. In Subsection 5.1 we show how for finite V the Markov process can be reversed in time. From this we derive a time-reversal symmetry for the flux Lagrangian $\mathcal{L}$ in Subsection 5.2. We then derive a similar time-reversal symmetry for the concentration large-deviations in Subsection 5.3. In Subsection 5.4 we discuss possible contractions and decompositions of the rate functionals, which in particular leads to the net flux Lagrangian; from there on we shall need to work with net fluxes. In Subsection 5.5 we introduce the concept of a dissipative structure, and its relation to generalised gradient systems in an abstract setting. Finally, in Subsection 5.6 we study how these concepts apply to the reacting particle model studied in this paper.

5.1 The time-reversed process

An essential role in MFT is played by the time-reversed process. In this subsection we construct this process, which will we exploited in the next subsections.

We will assume that the effective stoichiometric vectors $\gamma^{(r)}$ are $\mathbb{N}_0^I$-valued, that the process $C^{(V)}$ is non-explosive on $(0,T)$, irreducible and positive recurrent, so that there exists a unique, coordinate-wise positive invariant measure $0 < \Pi^{(V)} \in \mathcal{P}(\frac{1}{V}\mathbb{N}_0^I)$. Note that this is not a trivial assumption, for the following reasons. The whole space $\frac{1}{V}\mathbb{N}_0^I$ may be decomposed into separate so-called stoichiometric compatibility classes $\big(C^{(V)}(0) + span\{\gamma^{(r)}\}_{r\in\mathcal{R}}\big) \cap \mathbb{N}_0^I$, depending on the initial condition $C^{(V)}(0)$, see for example [AK11]. In this case we will implicitly assume that $\Pi^{(V)}$ is supported inside one such class, so that $C^{(V)}(0) \sim \Pi^{(V)}$ remains within that class. This forces the irreducibility of the process, apart from possible effects on the boundary of the compatibility class. Such boundaries correspond to the case where some species is no longer present in the system. If that happens it could become impossible for a reaction to take place, and hence the process might still be reducible. However, for now we are only concerned with finite V, so we can always add a very small reaction rate away from the boundary such that it vanishes in the limit $\frac{1}{V}k^{(V,r)} \to \bar{k}^{(r)}$ as $V \to \infty$.

Usually in MFT, one defines the time-reversed path by simply replacing time t by $T - t$. However, this would turn non-decreasing integrated fluxes into non-increasing ones, which are non-feasible as we consider one-way rather than net fluxes. The solution is evident: if for the forward process a reaction r takes place at time t, then for the reversed process a reverse reaction $\overleftarrow{r}$ should take place at time $T - t$, where $\overleftarrow{r}$ denotes the with effective stoichiometric vector $\gamma^{(\overleftarrow{r})} := -\gamma^{(r)}$. More precisely, let $\overleftarrow{\mathcal{R}} := \{\overleftarrow{r} : r \in \mathcal{R}\}$, which may not be disjoint from $\mathcal{R}$. For any given path $(c, \mathsf{w}) \in \mathrm{BV}(0,T;\mathfrak{l}^1(I)\times\mathfrak{l}^1(\mathcal{R}))$, we then define the time-reversed path $(\overleftarrow{c}, \overleftarrow{\mathsf{w}}) \in \mathrm{BV}(0,T;\mathfrak{l}^1(I)\times\mathfrak{l}^1(\overleftarrow{\mathcal{R}}))$ by[1]:

$$\overleftarrow{c}(t) := c(T-t) \qquad \text{and} \qquad \overleftarrow{\mathsf{w}}(t) := \mathsf{w}^{\mathsf{T}}(T) - \mathsf{w}^{\mathsf{T}}(T-t),$$

using the notation $w_r^{\mathsf{T}} := w_{\overleftarrow{r}}$. Observe that the reversed integrated fluxes $\overleftarrow{\mathsf{w}}(t)$ are now indeed non-decreasing, and the initial condition $\overleftarrow{\mathsf{w}}(0) = 0$ also holds. Moreover, if we extend the continuity operator by setting $\Gamma\mathsf{w} := \sum_{r\in\mathcal{R}}\gamma^{(r)}\mathsf{w}_r + \sum_{\overleftarrow{r}\in\overleftarrow{\mathcal{R}}}\gamma^{(\overleftarrow{r})}\mathsf{w}_{\overleftarrow{r}}$ for $w \in \mathbb{R}^{\mathcal{R}\cup\overleftarrow{\mathcal{R}}}$, then the continuity equation is invariant under time-reversal:

$$\partial_t(\overleftarrow{c})(t) - \Gamma\partial_t(\overleftarrow{\mathsf{w}})(t) = -\dot{c}(T-t) + \Gamma\dot{\mathsf{w}}(T-t) = 0.$$

We can now characterise the time-reversed process as follows:

Proposition 5.1. *Fix $V > 0$. Assume that all $\gamma^{(r)} \in \mathbb{N}_0^I$, that the process $C^{(V)}(t)$ is non-explosive on $(0,T)$, irreducible and positive recurrent, with unique, coordinate-wise positive invariant measure $0 < \Pi^{(V)} \in \mathcal{P}(\frac{1}{V}\mathbb{N}_0^I)$. Let $(C^{(V)}(0), \mathsf{W}^{(V)}(0))$ be distributed according to $\Pi^{(V)}(I) \times \delta_0$. Then the process $\big(\overleftarrow{C}^{(V)}(t), \overleftarrow{\mathsf{W}}^{(V)}(t)\big)$ is again Markov, where $\big(\overleftarrow{C}^{(V)}(0), \overleftarrow{\mathsf{W}}^{(V)}(0)\big)$ is distributed according to $\Pi^{(V)} \times \delta_0$, and its generator is:*

$$(\overleftarrow{\mathcal{Q}}\phi)(c,\mathsf{w}) = \sum_{\overleftarrow{r}\in\overleftarrow{\mathcal{R}}} \overleftarrow{k}^{(V,\overleftarrow{r})}(c)\big[\phi(c + \tfrac{1}{V}\gamma^{(\overleftarrow{r})}, \mathsf{w} + \tfrac{1}{V}\mathbb{1}_{\overleftarrow{r}}) - \phi(c,\mathsf{w})\big], \tag{32}$$

[1] In fact, a more precise definition would involve the left limit $(T-t)^-$ so that a time-reversed càdlàg path is again càdlàg; for brevity we will ignore this issue.

where

$$\overleftarrow{k}^{(V,\overleftarrow{r})}(c) := \frac{\Pi^{(V)}(c+\frac{1}{V}\gamma^{(\overleftarrow{r})})}{\Pi^{(V)}(c)} k^{(V,r)}(c+\tfrac{1}{V}\gamma^{(\overleftarrow{r})}). \tag{33}$$

Moreover, for any hybrid-measurable $\mathcal{A} \subset \mathrm{BV}\big(0,T;\mathbb{N}_0^I \times \mathfrak{l}^1(\mathcal{R})\big)$*,*

$$\mathrm{Prob}\big((C^{(V)},\mathsf{W}^{(V)}) \in \mathcal{A}\big) = \mathrm{Prob}\big((\overleftarrow{C}^{(V)},\overleftarrow{\mathsf{W}}^{(V)}) \in \mathcal{A}\big). \tag{34}$$

Proof. For brevity we shall omit the superscripts V in this proof. Since the concentrations $C(t)$ can only take values in $\frac{1}{V}\mathbb{N}_0^I$ we can use discrete Markov chain theory. It should be noted that $\Pi \times \delta_0$ is not an invariant measure - in fact, $W(t)$ does not have any. Nevertheless, the time-reversal can be proven along the lines of the standard Markov time-reversal result [Nor97, Th. 3.7.1]; we only outline how the proof needs to be adapted to account for the lack of an invariant measure on the integrated fluxes.

Since the state space is discrete we can rewrite the operators $\mathcal{Q}$, $\overleftarrow{\mathcal{Q}}$ in terms of matrices, i.e.

$$Q(c,\mathsf{w}\to\hat{c},\hat{\mathsf{w}}) = \begin{cases} k^{(r)}(c), & \hat{c}=c+\frac{1}{V}\gamma^{(r)}, \hat{\mathsf{w}}=\mathsf{w}+\frac{1}{V}\mathbb{1}_r,\\ -\sum_{r\in\mathcal{R}} k^{(r)}(c), & \hat{c}=c, \hat{\mathsf{w}}=\mathsf{w},\\ 0, & \text{otherwise,}\end{cases}$$

$$\overleftarrow{Q}(\hat{c},\hat{\mathsf{w}}\to c,\mathsf{w}) = \begin{cases} \overleftarrow{k}^{(\overleftarrow{r})}(\hat{c}), & c=\hat{c}+\frac{1}{V}\gamma^{(\overleftarrow{r})}, \mathsf{w}=\hat{\mathsf{w}}+\frac{1}{V}\mathbb{1}_{\overleftarrow{r}},\\ -\sum_{\overleftarrow{r}\in\overleftarrow{\mathcal{R}}} \overleftarrow{k}^{(\overleftarrow{r})}(c), & c=\hat{c}, \mathsf{w}=\hat{\mathsf{w}},\\ 0, & \text{otherwise.}\end{cases}$$

Hence, by translation invariance in the integrated fluxes, we have

$$\begin{aligned} Q(c,\mathsf{w}\to\hat{c},\hat{\mathsf{w}}) &= \frac{\Pi(\hat{c})}{\Pi(c)}\overleftarrow{Q}(\hat{c},\mathsf{w}+(\hat{w}-w)^{\mathsf{T}}\to c,\mathsf{w})\\ &= \frac{\Pi(\hat{c})}{\Pi(c)}\overleftarrow{Q}(\hat{c},\hat{\mathsf{w}}^{\mathsf{T}}\to c,\mathsf{w}^{\mathsf{T}}). \end{aligned} \tag{35}$$

Let $p_t(c,\mathsf{w}\to\hat{c},\hat{\mathsf{w}})$ be the transition probability corresponding to the generator matrix Q. We claim that the transition probability for the time-reversed generator matrix $\overleftarrow{Q}$ is

$$\overleftarrow{p}_t(\hat{c},\hat{\mathsf{w}}^{\mathsf{T}}\to c,\mathsf{w}^{\mathsf{T}}) = \frac{\Pi(c)}{\Pi(\hat{c})}p_t(c,\mathsf{w}\to\hat{c},\hat{\mathsf{w}}).$$

This can be seen by using the Kolmogorov backward equation for p_t to derive the Kolmogorov forward equation for $\overleftarrow{p}_t$:

$$\begin{aligned} \frac{d}{dt}\overleftarrow{p}_t(\hat{c},\hat{\mathsf{w}}^{\mathsf{T}}\to c,\mathsf{w}^{\mathsf{T}}) &= \frac{\Pi(c)}{\Pi(\hat{c})}\sum_{\tilde{c},\tilde{\mathsf{w}}} Q(c,\mathsf{w}\to\tilde{c},\tilde{\mathsf{w}})p_t(\tilde{c},\tilde{\mathsf{w}}\to\hat{c},\hat{\mathsf{w}})\\ &\overset{(35)}{=} \sum_{\tilde{c},\tilde{\mathsf{w}}} \overleftarrow{p}_t(\hat{c},\hat{\mathsf{w}}^{\mathsf{T}}\to\tilde{c},\tilde{\mathsf{w}}^{\mathsf{T}})\overleftarrow{Q}(\tilde{c},\tilde{\mathsf{w}}^{\mathsf{T}}\to c,\mathsf{w}^{\mathsf{T}}). \end{aligned}$$

It is then easily checked (through arbitrary finite-dimensional distributions) that $\overleftarrow{p}_t$ defines a Markov process, and that (34) holds. By construction $(\overleftarrow{C}(0),\overleftarrow{\mathsf{W}}(0))$ is distributed according to $\Pi\times\delta_0$. □

Remark 5.2. The process $C^{(V)}(t)$ is in detailed balance with respect to $\Pi^{(V)}$ precisely when $\overleftarrow{k}^{(V,r)} = k^{(V,r)}$ for all $r \in \mathcal{R}$. In that case the limit rates also satisfy

$$\overleftarrow{\bar{k}}^{(r)} = \bar{k}^{(r)}; \tag{36}$$

we call this symmetry *chemical detailed balance* to distinguish it from detailed balance of a Markov process. □

Remark 5.3. In the case of independent particles we can calculate the reversed jump rates explicitly. Set $k^{(n,(i,j))}(c) := n\mathsf{Q}_{ij}c_i$, and let $\pi \in \mathcal{P}(I)$ be the invariant measure for one particle. The invariant measure for $C^{(n)}$ is the multinomial distribution $\Pi^{(n)}(c) = \frac{n!}{\kappa_1!\dots\kappa_I!}\prod_{i=1}^I \pi_i^{nc_i}$. Then (33) becomes $\overleftarrow{k}^{(n,\overleftarrow{(i,j)})}(c) = n\frac{\pi_i}{\pi_j}\mathsf{Q}_{ij}c_j$, which is not surprising as $\frac{\pi_j}{\pi_i}\mathsf{Q}_{ji}$ is the reversed one-particle generator. Hence the average jump rates are simply

$$\bar{k}^{(i,j)}(c) = \mathsf{Q}_{ij}c_i \qquad \text{and} \qquad \overleftarrow{\bar{k}}^{(\overleftarrow{i,j})}(c) = \overleftarrow{\bar{k}}^{(j,i)}(c) = \frac{\pi_i}{\pi_j}\mathsf{Q}_{ij}c_j. \tag{37}$$

□

5.2 Time-reversal symmetry for the fluxes

Let $\overleftarrow{\mathcal{J}}_{(0,T)}(c,w) := \int_0^T \overleftarrow{\mathcal{L}}\big(c(t),\mathsf{w}(t)\big)\,dt$ be the large-deviation rate functional corresponding to the time-reversed process (32), defined as in (8) where the sum now ranges over $\overleftarrow{\mathcal{R}}$ and the average jump rates $\bar{k}^{(r)}$ are replaced by $\overleftarrow{\bar{k}}^{(\overleftarrow{r})} := \lim_{V\to\infty}\frac{1}{V}\overleftarrow{k}^{(V,\overleftarrow{r})}$. Moreover, let $\mathcal{I}_0$ be the *quasi-potential*, that is, the large-deviation rate corresponding to $\Pi^{(V)}$.

If we consider the forward and the backward processes initially distributed according to $\Pi^{(V)} \times \delta_0$ as in Proposition 5.1, then using the time-reversal (34) property together with Corollary 4.16, we find the following time-reversal symmetry:

$$\mathcal{I}_0(c(0)) + \mathcal{J}_{(0,T)}(c,\mathsf{w}) = \mathcal{I}_0(c(T)) + \overleftarrow{\mathcal{J}}_{(0,T)}(\overleftarrow{c},\overleftarrow{\mathsf{w}}). \tag{38}$$

Following [BDSG$^+$15, Sec. II.C], we replace the time-interval by $(t-\epsilon, t+\epsilon)$ for some $t \in (0,T)$, divide (38) by 2ϵ and then let $\epsilon \to 0$, yielding the local time-reversal relation:

$$\mathcal{L}(c,\mathsf{j}) - \overleftarrow{\mathcal{L}}(c,\mathsf{j}^\mathsf{T}) = D\mathcal{I}_0(c)\cdot s, \qquad s + \Gamma\mathsf{j} = 0 \tag{39}$$

for all $(c,\mathsf{j}) \in \mathfrak{l}^1_+(I)\times\mathfrak{l}^1(\mathcal{R})$.

For the independent particles model, the reversed average jump rates are known a priori by (37), and relation (39) can also be derived directly:

$$\begin{aligned}
&\mathcal{L}(c,\mathsf{j}) - \overleftarrow{\mathcal{L}}(c,\mathsf{j}^\mathsf{T}) \\
&\quad= \mathcal{S}(\mathsf{j}|c\otimes\mathsf{Q}) - \mathcal{S}\big(\mathsf{j}^\mathsf{T}\big|\tfrac{c}{\pi}\otimes\mathsf{Q}^\mathsf{T}\otimes\pi\big) \\
&\quad= \sum_i \Big[\underbrace{\sum_{j\neq i}(\mathsf{j}_{ij}-\mathsf{j}_{ji})}_{=-\operatorname{div}_i\mathsf{j}=s}\log\frac{c_i}{\pi_i} + c_i\underbrace{\sum_{j\neq i}(\mathsf{Q}_{ij}-\frac{1}{\pi_i}\mathsf{Q}_{ji}\pi_j)}_{=0}\Big] \\
&\quad= D\mathcal{S}(c|\pi)\cdot s.
\end{aligned}$$

For the more general reacting particle model, the exact expressions for the backward rates may not be known, and we can exploit (39) to derive a relation with the forward rates:

$$\begin{aligned}\Gamma^{\mathsf{T}} D\mathcal{I}_0(c)\cdot \mathsf{j} = D\mathcal{I}_0(c)\cdot \Gamma \mathsf{j} = D\mathcal{I}_0(c)\cdot s &= \mathcal{S}\big(\mathsf{j}|\bar{k}(c)\big) - \mathcal{S}\big(\mathsf{j}^{\mathsf{T}}|\overleftarrow{\bar{k}}(c)\big)\\ &= \sum_{r\in\mathcal{R}} \mathsf{j}_r \log\frac{\overleftarrow{\bar{k}}^{(\overleftarrow{r})}(c)}{\bar{k}^{(r)}(c)} - \bar{k}^{(r)}(c) + \overleftarrow{\bar{k}}^{(\overleftarrow{r})}(c),\end{aligned} \tag{40}$$

where $(\Gamma^{\mathsf{T}}\xi)_r := \xi\cdot\gamma^{(r)}$ and $\mathsf{j}^{\mathsf{T}}_{\overleftarrow{r}} = \mathsf{j}_r$. Since (40) has to hold for all c, j, the reversed rates have to satisfy the following two conditions for all c:

$$\begin{aligned}\big(\Gamma^{\mathsf{T}} D\mathcal{I}_0(c)\big)_r = D\mathcal{I}_0(c)\cdot\gamma^{(r)} = \log\frac{\overleftarrow{\bar{k}}^{(\overleftarrow{r})}(c)}{\bar{k}^{(r)}(c)} \quad \forall r\in\mathcal{R},\\ \sum_{r\in\mathcal{R}} \bar{k}^{(r)}(c) = \sum_{\overleftarrow{r}\in\overleftarrow{\mathcal{R}}} \overleftarrow{\bar{k}}^{(\overleftarrow{r})}(c).\end{aligned} \tag{41}$$

The first condition is sometimes interpreted as a fluctuation-dissipation equation, see [BDSG$^+$15, Sec. II.C] and [MPR14, Sec. 3.5]. It also shows that there is a strong relation between mass-action kinetics and relative entropy, see [MPPR, Sec. 3.5].

Remark 5.4. At least formally, the time-averaged flux

$$\mathsf{J}^{(V,T)} := T^{-1}\int_0^T \dot{\mathsf{W}}^{(V)}(dt) = T^{-1}\mathsf{W}^{(V)}(T)$$

satisfies a large-deviation principle as $V\to\infty$ and subsequently $T\to\infty$ with rate functional (or the lower semicontinuous regularisation thereof):

$$\mathsf{j}\mapsto \lim_{T\to\infty}\inf_{\substack{(c,\mathsf{w})\in W^{1,1}(0,T;\mathfrak{l}^1(I)\times\mathfrak{l}^1(\mathcal{R})):\\ \dot{c}=\Gamma\dot{\mathsf{w}},\\ \mathsf{w}(T)/T=\mathsf{j}}} \frac{1}{T}\mathcal{J}_{(0,T)}(c,\mathsf{w}),$$

and so-called a Galavotti-Cohen time-reversal symmetry also holds for this functional. This is beyond the scope of the current paper. □

5.3 Time-reversal symmetry for the densities

As briefly mentioned in the introduction, by a simple contraction principle we retrieve the dynamic large deviations of the empirical measure $C^{(V)}$ with rate functional

$$\mathcal{I}_{(0,T)}(c) = \inf_{\substack{\mathsf{w}\in \mathrm{BV}(0,T;\mathfrak{l}^1_+(I\times I)):\\ \dot{c}=\Gamma\dot{\mathsf{w}}}} \mathcal{J}_{(0,T)}(c,\mathsf{w}) = \int_0^T \underbrace{\inf_{\substack{\mathsf{j}\in\mathfrak{l}^1_+(\mathcal{R}):\\ \dot{c}(t)=\Gamma j}} \mathcal{L}(c(t),\mathsf{j})}_{\hat{\mathcal{L}}(c(t),\dot{c}(t))}\, dt, \tag{42}$$

see for example [Fen94, Léo95, SW95, PR16]. Let $\overleftarrow{\hat{\mathcal{L}}}$ be the same functional, but for the time-reversed process $\overleftarrow{C}^{(V)}$, i.e. the rates $\bar{k}$ in $\mathcal{L}$ are replaced by the backward rates $\overleftarrow{\bar{k}}$. Taking the infimum over feasible fluxes $s=\Gamma\mathsf{j}$ of (39) yields the time-reversal symmetry for the densities:

$$\hat{\mathcal{L}}(c,s) - \overleftarrow{\hat{\mathcal{L}}}(c,-s) = D\mathcal{I}_0(c)\cdot s. \tag{43}$$

Moreover, from this relation the quasi-potential can be retrieved via (again, see [BDSG+15, Sec. IV.A])

$$\mathcal{I}_0(c) = \inf_{\substack{\hat{c}\in \mathrm{BV}(-\infty,0;\mathfrak{l}^1_+(I)):\\ \hat{c}(0)=c}} \mathcal{J}_{(-\infty,0]}(\hat{c}).$$

5.4 Net fluxes and other decompositions

The advantage of working with one-way fluxes is that the rate functional $\mathcal{J}_{(0,T)}$ has a nice explicit formula, from which we can deduce additional structure as in (38). Of course the one-way fluxes encode more information, and by a straight-forward contraction one finds the large-deviation rate for the net integrated fluxes $\overline{\mathsf{W}}^{(V)}(t) := \mathsf{W}^{(V)}(t) - \mathsf{W}^{(V)\mathsf{T}}(t)$:

$$\overline{\mathcal{J}}_{(0,T)}(c,\overline{\mathsf{w}}) := \inf_{\substack{\mathsf{w}\in W^{1,1}(0,T;\mathfrak{l}^1(\mathcal{R})):\\ \mathsf{w}(t)-\mathsf{w}^{\mathsf{T}}(t)=\overline{\mathsf{w}}(t)}} \mathcal{J}_{(0,T)}(c,w) = \int_0^T \underbrace{\inf_{\substack{\mathsf{j}\in\mathfrak{l}^1(\mathcal{R}):\\ \mathsf{j}-\mathsf{j}^{\mathsf{T}}=\dot{\overline{\mathsf{w}}}(t)}} \mathcal{L}\big(c(t),\mathsf{j}\big)}_{=:\overline{\mathcal{L}}(c(t),\dot{\overline{\mathsf{w}}}(t))} \, dt. \tag{44}$$

Observe that net (integrated) fluxes are always anti-symmetric, and so we can rewrite the continuity operator:

$$\overline{\Gamma\mathsf{j}} := \sum_{r\in\mathcal{R}_{\mathrm{fw}}} \bar{\mathsf{j}}_r\gamma^{(r)} = \sum_{r\in\mathcal{R}_{\mathrm{fw}}} (\mathsf{j}_r - \mathsf{j}_{\overleftarrow{r}})\gamma^{(r)} = \sum_{r\in\mathcal{R}_{\mathrm{fw}}\cup\mathcal{R}_{\mathrm{bw}}} \mathsf{j}_r\gamma^{(r)} = \Gamma\mathsf{j}. \tag{45}$$

Taking the infimum of (39) over $\mathsf{j}\in\mathfrak{l}^1(\mathcal{R}) : \mathsf{j}-\mathsf{j}^{\mathsf{T}} = \bar{j}$, which does not affect the velocity field s, we find the time-reversal symmetry for the net fluxes:

$$\overline{\mathcal{L}}(c,\bar{\mathsf{j}}) - \overleftarrow{\overline{\mathcal{L}}}(c,-\bar{\mathsf{j}}) = DI_0(c)\cdot s, \qquad s = \overline{\Gamma\mathsf{j}}, \tag{46}$$

where $\overleftarrow{\overline{\mathcal{L}}}$ is again the same as $\overline{\mathcal{L}}$ with the reversed reaction rates $\overleftarrow{k}$. The net flux Lagrangian $\overline{\mathcal{L}}$ will play a central in the derivation of dissipative structures in Subsections 5.5 and 5.6. For more information, analysis and thermodynamic interpretation we refer to [BMN09].

Alternatively, if the non-negativity of the net fluxes is of importance, one could also split $\mathsf{W}^{(V)}_r = [\mathsf{W}^{(V)}_r \wedge \mathsf{W}^{(V)}_{\overleftarrow{r}}] + [\mathsf{W}^{(V)}_r - \mathsf{W}^{(V)}_r \wedge \mathsf{W}^{(V)}_{\overleftarrow{r}}]$, and apply contractions to find the large deviations for the cancelling fluxes and the net fluxes respectively.

The net fluxes can be decomposed even further. It is however unclear how to do this in a meaningful way. As common in MFT, we could decompose any flux into a gradient and a solenoidal part $\mathsf{w} = \mathsf{w}_\nabla + \mathsf{w}_{\mathrm{sol}}$, where $\mathsf{w}_\nabla = \nabla\xi$ for some $\xi\in\mathbb{R}^I$, and $\operatorname{div}\mathsf{w}_{\mathrm{sol}} = 0$. However, one usually exploits the quadratic structure of the rate functional to devise an *orthogonal* decomposition such that the rate functional split into two parts. In the discrete setting of this paper however, the rate functional $\mathcal{J}_{(0,T)}$ is entropic rather than quadratic, and so it lacks a natural orthogonality relation. Therefore it remains an open question how a meaningful decomposition of the fluxes should look like, if there is any.

5.5 General theory of dissipative structures and gradient structures

In an ideal setting, the concentration large deviations satisfy a time-reversal symmetry of the form (39) where $\overleftarrow{\hat{\mathcal{L}}} = \hat{\mathcal{L}}$; in that case the theory of [MPR14] shows that $\hat{\mathcal{L}}$ actually induces a natural generalised gradient structure, as we explain below. A gradient structure holds precise physical information about the dissipation mechanism and force that drives the system towards its equilibrium. In a much less

ideal setting, the flux large deviations often induces a similar *dissipative structure*. For the setting of this paper, it turns out to be impossible to induce such structure unless we consider the net flux Lagrangian $\overline{\mathcal{L}}$. Therefore we shall from now on consider net fluxes rather than one-way fluxes.

To shed light on the key properties behind the arguments, we shall introduce the concepts and their relation in a much more general setting. We nevertheless use the same symbols $\overline{\mathcal{L}}, \hat{\mathcal{L}}, \Gamma, c, s$ and $\bar{\jmath}$, etc. In the next subsection we discuss how this general theory applies to the reacting particles model.

Definition 5.5. *Let X be a Banach space[2]. A function $\hat{\mathcal{L}} : X \times X \to \mathbb{R}$ is called an* L-function *whenever $\hat{\mathcal{L}}$ is non-negative, convex in the second argument, and for any $c \in X$ there exists a unique $s \in X$ for which $\hat{\mathcal{L}}(c,s) = 0$.*

A generalised gradient structure is a triple $(X, \mathcal{F}, \Psi)$, where X is a Banach space, $\mathcal{F} : X \to \mathbb{R}$ is a differentiable functional, and $\Psi : X \times X \to \mathbb{R}$ is a dissipation potential*, meaning that*

1. *$\Psi(c,\cdot)$ is convex,*
2. *$\min \Psi(c,\cdot) = \Psi(c,0) = 0$.*

We say that an L-function $\hat{\mathcal{L}}$ induces the generalised gradient structure *$(X, \mathcal{F}, \Psi)$ if one can write*

$$\hat{\mathcal{L}}(c,s) = \Psi(c,s) + \Psi^*\big(c, -D\mathcal{F}(c)\big) + \langle D\mathcal{F}(c), s\rangle, \tag{47}$$

where

$$\Psi^*(c,\xi) := \sup_{s\in X} \langle \xi, s\rangle - \Psi(c,s) \qquad \text{for } \xi \in X^*.$$

A few comments about these definitions are in place. Firstly, observe that $\hat{\mathcal{L}}(c(t), \dot{c}(t)) = 0$ uniquely characterises an evolution equation; for the reacting particles model this would be the Reaction Rate Equation (7). Hence if $\hat{\mathcal{L}}$ induces a generalised gradient structure then by Legendre duality that evolution equation can be written as $\dot{c}(t) = D_\xi \Psi^*\big(c(t), -D\mathcal{F}(c(t))\big)$. This can be interpreted as a gradient flow or response theory, generalised to allow for nonlinear relations between the driving force $-D\mathcal{F}(c)$ and velocities $\dot{c}$. Moreover, the conditions on Ψ imply that both Ψ and Ψ^* are non-negative, therefore implying thermodynamic consistency: $\frac{d}{dt}\mathcal{F}(c(t)) = \langle D\mathcal{F}(c(t)), \dot{c}(t)\rangle \le 0$. Finally, relation (47) says that the energetic cost to pay to deviate from the evolution equation is exactly the cost function $\hat{\mathcal{L}}$.

Inspired by [MN08, Mae17, KJZ17], we study a more general dissipative structure, induced by a (net flux) Lagrangian. In this definition we will need a second Banach space for the fluxes.

Definition 5.6. *Let X, Y be Banach spaces. A dissipative structure is a quadruple (X, Y, F, Φ), where $F : X \to Y^*$, and $\Phi : X \times Y \to \mathbb{R}$ is a dissipation potential in the same sense as in Definition 5.5.*

Moreover, we say that the L-function (in the same sense as in Definition 5.5) $\overline{\mathcal{L}}$ induces the dissipative structure *(X, Y, F, Φ) if one can write*

$$\overline{\mathcal{L}}(c,\bar{\jmath}) = \Phi(c,\bar{\jmath}) + \Phi^*\big(c, F(c)\big) - \langle F(c), \bar{\jmath}\rangle, \tag{48}$$

[2]We introduce the theory on a general Banach space X rather than on a manifolds to shorten notation. Possible issues that can arise near boundaries of the manifold, e.g. where concentrations can become negative, can be easily dealt with by setting the appropriate functionals to ∞ whenever such restrictions are violated.

where

$$\Phi^*(c,\zeta) := \sup_{\bar{\jmath}\in Y} \langle \zeta, \bar{\jmath}\rangle - \Phi(c,\bar{\jmath}) \qquad \textit{for } \zeta \in Y^*.$$

As in the case of a gradient structure, the optimal net fluxes can be rewritten as $\bar{\jmath} = D_F\Phi^*\big(c, F(c)\big)$, which now describes a relation between the force $F(c)$ and the net flux $\bar{\jmath}$. For more background and physical interpretation of this structure we refer to [MN08, Mae17].

We now study the relation between dissipative structures and generalised gradient structures. For this we need to link generalised fluxes to velocities via a continuity operator $\overline{\Gamma}$. To shorten notation we restrict to L-functions that are differentiable in the second argument; the induced dissipation potentials are then also differentiable. All arguments below can easily be generalised by using subdifferentials rather than derivatives.

Proposition 5.7. *Let X, Y be Banach spaces, let $\overline{\mathcal{L}} : X \times Y \to \mathbb{R}_+$ be a given L-function, differentiable in the second argument, such that it induces a disipative structure (X, Y, F, Φ), and let $\overline{\Gamma} : Y \to X$ be a bounded linear operator. If there is a differentiable $\mathcal{F} : X \to \mathbb{R}$ such that*

$$F(c) = -\overline{\Gamma}^{\mathsf{T}} D\mathcal{F}(c), \tag{49}$$

then $\hat{\mathcal{L}}(c,s) = \inf\big\{\overline{\mathcal{L}}(c,\bar{\jmath}) : \bar{\jmath}\in Y, s = \overline{\Gamma}\bar{\jmath}\big\}$ induces the generalised gradient structure $(X, \mathcal{F}, \Psi)$, where $\mathcal{F}$ is unique up to constants, and

$$\Psi(c,s) := \inf_{\bar{\jmath}\in Y: s=\overline{\Gamma}\bar{\jmath}} \Phi(c,\bar{\jmath}), \qquad \textit{and} \qquad \Psi^*(c,\xi) = \Phi^*(c,\overline{\Gamma}^{\mathsf{T}}\xi). \tag{50}$$

On the other hand, if $\hat{\mathcal{L}}$ induces a generalised gradient structure $(X, \mathcal{F}, \Psi)$ then

$$\overline{\Gamma} D\Phi^*\big(c, \overline{\Gamma}^{\mathsf{T}} D\mathcal{F}(c) + F(c)\big) = 0. \tag{51}$$

Naturally property (51) is a weaker statement than (49); since Φ is a dissipation potential there holds $D\Phi^*(c,0) = 0$.

Proof. It is easily calculated that the potentials (50) are indeed dual to each other, and that they are dissipation potentials.

Assume that (49) holds. We can then calculate:

$$\begin{aligned}
\hat{\mathcal{L}}(c,s) &= \inf_{\bar{\jmath}\in Y: s=\overline{\Gamma}\bar{\jmath}} \Phi(c,\bar{\jmath}) + \Phi^*\big(c, F(c)\big) - \langle F(c), \bar{\jmath}\rangle \\
&= \inf_{\bar{\jmath}\in Y: s=\overline{\Gamma}\bar{\jmath}} \Phi(c,\bar{\jmath}) + \Phi^*\big(c, -\overline{\Gamma}^{\mathsf{T}} D\mathcal{F}(c)\big) + \langle D\mathcal{F}(c), s\rangle \\
&= \Psi(c,s) + \Psi^*\big(c, -D\mathcal{F}(c)\big) + \langle D\mathcal{F}(c), s\rangle.
\end{aligned}$$

This shows this $\hat{\mathcal{L}}$ induces the gradient structure $(X, \mathcal{F}, \Psi)$ as claimed; uniqueness of $\mathcal{F}$ up to constants follows from [MPR14, Th. 2.1(ii)].

For the other direction, assume that $\hat{\mathcal{L}}$ induces a gradient structure $(X, \mathcal{F}, \Psi)$. Then for any $\xi \in X^*$,

$$\begin{aligned}
&\Psi^*\big(c, \xi - D\mathcal{F}(c)\big) - \Psi^*\big(c, -D\mathcal{F}(c)\big) \\
&\qquad = \sup_{s\in X} \langle \xi, s\rangle - \Psi(c,s) - \Psi^*\big(c, -D\mathcal{F}(c)\big) - \langle D\mathcal{F}(c), s\rangle \\
&\qquad = \hat{\mathcal{H}}(c,\xi) \\
&\qquad = \sup_{s\in X} \langle \xi, s\rangle - \inf_{\bar{\mathrm{j}}\in Y : s = \overline{\Gamma}\bar{\mathrm{j}}} \overline{\mathcal{L}}(c,\bar{\mathrm{j}}) \\
&\qquad = \overline{\mathcal{H}}(c, \overline{\Gamma}^{\mathsf{T}}\xi) \\
&\qquad = \sup_{\bar{\mathrm{j}}\in Y} \langle \overline{\Gamma}^{\mathsf{T}}\xi, \bar{\mathrm{j}}\rangle - \Phi(c,\bar{\mathrm{j}}) - \Phi^*\big(c, F(c)\big) + \langle F(c), \bar{\mathrm{j}}\rangle \\
&\qquad = \Phi^*\big(c, \overline{\Gamma}^{\mathsf{T}}\xi + F(c)\big) - \Phi^*\big(c, F(c)\big).
\end{aligned}$$

Differentiating both sides in $\xi = D\mathcal{F}(c)$ yields (51). □

In fact, whenever condition (49) holds, then $\overline{\mathcal{L}}(c,\bar{j})$ also induces a gradient structure in flux space, as follows. By a change of variables we can redefine the energy functional in terms of the integrated net flux $\overline{\mathrm{w}}(t) := \int_0^{\hat{t}} \bar{\mathrm{j}}(\hat{t})\, d\hat{t}$:

$$\tilde{\mathcal{F}}(\overline{\mathrm{w}}) := \mathcal{F}\big(c(0) + \overline{\Gamma}\overline{\mathrm{w}}\big).$$

Then clearly $D\tilde{\mathcal{F}}(\overline{\mathrm{w}}) = \overline{\Gamma}^{\mathsf{T}} D\mathcal{F}\big(c(0) + \overline{\Gamma}\overline{\mathrm{w}}\big)$, and hence (48) can be written as

$$\tilde{\mathcal{L}}(\overline{\mathrm{w}}, \bar{\mathrm{j}}) := \overline{\mathcal{L}}\big(c(0) + \overline{\Gamma}\overline{\mathrm{w}}, \bar{j}\big) = \tilde{\Phi}(\overline{\mathrm{w}}, \bar{\mathrm{j}}) + \tilde{\Phi}^*\big(\overline{\mathrm{w}}, -D\tilde{\mathcal{F}}(\overline{\mathrm{w}})\big) + \langle D\tilde{\mathcal{F}}(\overline{\mathrm{w}}), \bar{\mathrm{j}}\rangle,$$

where $\tilde{\Phi}(\overline{\mathrm{w}}, \bar{\mathrm{j}}) := \Phi\big(c(0) + \overline{\Gamma}\overline{\mathrm{w}}, \bar{j}\big)$.

5.6 Dissipative structure and gradient structure for the reacting particle model

We now study how the theory of the previous subsection can be applied to the reacting particle model. We take $X = \mathfrak{l}^1(I), Y = \mathfrak{l}^1(\mathcal{R}_{\mathrm{fw}})$, and so $\langle \zeta, \bar{\mathrm{j}}\rangle = \zeta \cdot_{\mathcal{R}_{\mathrm{fw}}} \bar{\mathrm{j}} := \sum_{r\in\mathcal{R}_{\mathrm{fw}}} \zeta_r \bar{\mathrm{j}}_r$. Moreover, $\overline{\mathcal{L}}$ and $\hat{\mathcal{L}}$ are now again given by (44) and (42) where $\mathcal{L}$ is (31), and $\overline{\Gamma}$ is given by (45). We will see that a necessary and sufficient condition for the net flux Lagrangian to induce a dissipative structure is a condition which is sometimes called *weak detailed balance*, and a sufficient condition for the concentration Lagrangian to induce a generalised gradient system is chemical detailed balance.

Proposition 5.8. *The net flux Lagrangian $\overline{\mathcal{L}}$ from* (44) *induces a dissipative structure $(\mathfrak{l}^1(I), \mathfrak{l}^1(\mathcal{R}_{\mathrm{fw}}), F, \Phi)$ precisely when*

$$\bar{k}^{(r)}(c) = 0 \iff \bar{k}^{(\overleftarrow{r})}(c) = 0 \qquad\qquad \forall c \in \mathfrak{l}^1_+(I), r \in \mathcal{R}_{\mathrm{fw}}. \tag{52}$$

In that case, the dissipative structure is unique on the support of $\bar{k}^{(r)}(c)$, and given by

$$F_r(c) = \frac{1}{2}\log\frac{\bar{k}^{(r)}(c)}{\bar{k}^{(\overleftarrow{r})}(c)} \qquad \textit{and} \qquad \Phi^*(c,\zeta) = \sum_{r\in\mathcal{R}_{\mathrm{fw}}} 2\sqrt{\bar{k}^{(r)}(c)\bar{k}^{(\overleftarrow{r})}(c)}\big(\cosh(\zeta_r) - 1\big). \tag{53}$$

Proof. Fix an arbitrary $c \in \mathfrak{l}^1_+(I)$. We apply [MPR14, Prop. 2.1] to the net flux Lagrangian; the existence of the dissipative structure is equivalent to the existence of the derivative $-F(c) = D_{\dot{\mathsf{j}}}\overline{\mathcal{L}}(c,0)$. Since $\overline{\mathcal{L}}$ is only implicitly defined through an infimum, it is easier to check the equivalent statement that $F(c)$ minimises $\overline{\mathcal{H}}(c,\cdot)$, the convex dual of $\overline{\mathcal{L}}(c,\cdot)$. In order to calculate this dual, we need will need, for any $\zeta \in \mathfrak{l}^\infty(\mathcal{R}_{\rm fw})$ its antisymmetric extension

$$\overline{\zeta}_r := \begin{cases} \zeta_r, & r \in \mathcal{R}_{\rm fw}, \\ -\zeta_{\overleftarrow{r}}, & r \in \mathcal{R}_{\rm bw}. \end{cases}$$

With this notation,

$$\overline{\mathcal{H}}(c,\zeta) = \sup_{\mathsf{j}\in\mathfrak{l}^1(\mathcal{R})} \zeta\cdot(\mathsf{j}-\mathsf{j}^{\mathsf{T}})\mathbb{1}_{\mathcal{R}_{\rm fw}} - \mathcal{L}(c,\mathsf{j}) = \sup_{\mathsf{j}\in\mathfrak{l}^1(\mathcal{R})} \overline{\zeta}\cdot\mathsf{j} - \mathcal{L}(c,\mathsf{j}) = \mathcal{H}(c,\overline{\zeta}).$$

where $\mathcal{H}$ is the Hamiltonian (29) corresponding to $\mathcal{L}$. By straightforward differentiation with respect to ζ_r (which occurs twice in $\mathcal{H}(c,\overline{\zeta})$ because of the antisymmetry), we find that $\mathcal{H}$ has the antisymmetric minimiser $-F_r(c)$ for all c, r if and only if (52) holds, and $F(c)$ is unique up to c, r for which $\bar{k}^{(r)}(c) = 0$.

Finally, by [MPR14, Lem. 2.1], the desired dissipation potential can be calculated explicitly through the formula $\Phi^*(c,\zeta) = \overline{\mathcal{H}}(c,\zeta - F(c)) - \overline{\mathcal{H}}(c,-F(c))$. □

Remark 5.9. By the same arguments, it is easily seen that the (one-way) flux Lagrangian $\mathcal{L}$ never induces a dissipative structure. Indeed, a minimiser ζ of $\mathcal{H}(c,\cdot)$ should satisfy for all $r \in \mathcal{R}$,

$$0 = \partial_{\zeta_r}\mathcal{H}(c,\zeta) = \bar{k}^{(r)}(c)e^{\zeta_r},$$

which can only hold if $\bar{k}^{(r)}(c) = 0$. □

If weak detailed balance (52) holds, then the backwards net flux Lagrangian $\overleftarrow{\mathcal{L}}$ also induces a dissipative structure, where $\overleftarrow{F}_r(c) = \frac{1}{2}\log\frac{\overleftarrow{\bar{k}}^{(r)}(c)}{\overleftarrow{\bar{k}}^{(\overleftarrow{r})}(c)}$. The forward and backward force fields characterise two interesting quantities:

$$F_r(c) + \overleftarrow{F}_{\overleftarrow{r}}(c) = \frac{1}{2}\log\frac{\bar{k}^{(r)}(c)\,\overleftarrow{\bar{k}}^{(\overleftarrow{r})}(c)}{\overleftarrow{\bar{k}}^{(r)}(c)\bar{k}^{(\overleftarrow{r})}(c)}, \tag{54}$$

$$F_r(c) - \overleftarrow{F}_{\overleftarrow{r}}(c) = \big(\Gamma^{\mathsf{T}} D\mathcal{I}_0(c)\big)_r. \tag{55}$$

The first quantity can be seen as a measure for how far the system is from chemical detailed balance, see Remark 5.2. The second equation, which follows from (41), relates the force field induced by the net flux Lagrangian to the quasi-potential. It is then not difficult to show that chemical detailed balance is a sufficient condition for the concentration Lagrangian $\hat{\mathcal{L}}$ to induce a generalised gradient flow:

Proposition 5.10. *Assume chemical detailed balance* (36) *holds. Then the concentration Lagrangian* $\hat{\mathcal{L}}$ *induces a generalised gradient system with* $\mathcal{F}(c) = \frac{1}{2}\mathcal{I}_0(c)$ *and* $\Psi^*(c,\xi) = \Phi^*(c,-\overline{\Gamma}^{\mathsf{T}}\xi)$ *and* Φ^* *is given by* (53)*.*

Proof. Since chemical detailed balance holds, weak detailed balance also holds, and so by Proposition 5.8 the net flux Lagrangian $\overline{\mathcal{L}}$ induces the dissipative structure $(\mathfrak{l}^1(I), \mathfrak{l}^1(\mathcal{R}_{\rm fw}), F, \Phi)$. Moreover, because of chemical detailed balance, we have $F(c) = \overleftarrow{F}(c)$, and so by the assymetry of $F(c)$, (55) becomes $F(c) = \frac{1}{2}\Gamma^{\mathsf{T}} D\mathcal{I}_0(c)$. The claim then follows from Proposition 5.7. □

The generalised gradient structure induced by the concentration Lagrangian $\hat{\mathcal{L}}$ was already found in [MPPR, Subsec. 3.4]; the arguments in this paper explain how the dissipation potential Ψ is related to the dissipative structure $(\mathfrak{l}^1(I), \mathfrak{l}^1(\mathcal{R}_{\rm fw}), F, \Phi)$ and the continuity operator $\overline{\Gamma}$.

References

[AFP06] L. Ambrosio, N. Fusco, and D. Pallara. *Functions of bounded variation and free discontinuity problems*. The Clarendon Press, Oxford University Press, New York, NY, USA, 2006.

[AK11] D.F. Anderson and T.G. Kurtz. Continuous time Markov chain models for chemical reaction networks. In Heinz Koeppl, Gianluca Setti, Mario di Bernardo, and Douglas Densmore, editors, *Design and Analysis of Biomolecular Circuits*, pages 3–42. Springer, New York, N.Y. U.S.A, 2011.

[BDSG$^+$02] L. Bertini, A. De Sole, D. Gabrielli, G. Jona-Lasinio, and C. Landim. Macroscopic fluctuation theory for stationary non-equilibrium states. *Journal of Statistical Physics*, 107(3-4):635–675, 2002.

[BDSG$^+$03] L. Bertini, A. De Sole, D. Gabrielli, G. Jona-Lasinio, and C. Landim. Large deviations for the boundary driven symmetric simple exclusion process. *Mathematical Physics, Analysis and Geometry*, 6:231–267, 2003.

[BDSG$^+$07] L. Bertini, A. De Sole, D. Gabrielli, G. Jona-Lasinio, and C. Landim. Large deviations of the empirical current in interacting particle systems. *Theory of Probability & Its Applications*, 51(1):2–27, 2007.

[BDSG$^+$15] L. Bertini, A. De Sole, D. Gabrielli, G. Jona-Lasinio, and C. Landim. Macroscopic fluctuation theory. *Reviews of Modern Physics*, 87(2), 2015.

[BFG15] L. Bertini, A. Faggionato, and D. Gabrielli. Large deviations of the empirical flow for continuous time Markov chains. *Annales de l'Institut Henri Poincaré*, 51(3):867–900, 2015.

[Big04] J.D. Biggings. Large deviations for mixtures. *Electronic Communications in Probability*, 9:60–71, 2004.

[BMN09] M. Baiesi, C. Maes, and K. Netočný. Computation of current cumulants for small nonequilibrium systems. *Journal of Statistical Physics*, 135(1):57–75, 2009.

[Bré83] H. Brézis. *Analyse fonctionelle, Théorie et applications (In French, English translation available)*. Mason, Paris, France, 1983.

[DLE03] B. Derrida, J.L. Lebowitz, and Speer E.R. Exact large deviation functional of a stationary open driven diffusive system: The asymmetric exclusion process. *Journal of Statistical Physics*, 110(3–6):775–809, 2003.

[Dud89] R.M. Dudley. *Real analysis and probability*. Wadsworth & Brooks/Cole, Pacific Grove, CA, USA, 1989.

[DZ87] A. Dembo and O. Zeitouni. *Large deviations techniques and applications*, volume 38 of *Stochastic modelling and applied probability*. Springer, New York, NY, USA, 2nd edition, 1987.

[EK05] S.N. Ethier and T.G Kurtz. *Markov processes – characterization and convergence*. John Wiley & sons, Inc., Hoboken, NJ, USA, 2005.

[Eva98] L.C. Evans. *Partial differential equations*. American Mathematical Society, Providence, RI, USA, 1998.

[Fen94] S. Feng. Large deviations for empirical process of mean-field interacting particle system with unbounded jumps. *The Annals of Probability*, 22(4):1679–2274, 1994.

[FK06] J. Feng and T.G. Kurtz. *Large deviations for stochastic processes*, volume 131. American Mathematical Society, Providence, RI, USA, 2006.

[HPR16] M. Heida, R.I.A. Patterson, and D.R.M. Renger. The space of bounded variation with infinite-dimensional codomain. WIAS Preprint No. 2353, 2016.

[Jak97] A. Jakubowski. A non-Skorohod topology on the Skorohod space. *Electronic Journal of Probability*, 2(4):1–21, 1997.

[Kal97] O. Kallenberg. *Foundations of modern probability*. Springer, New York, NY, USA, 1997.

[KJZ17] M. Kaiser, R.L. Jack, and J. Zimmer. Canonical structure and orthogonality of forces and currents in irreversible markov chains. *ArXiv Preprint*, 1708.01453, 2017.

[KL99] C. Kipnis and C. Landim. *Scaling limits of interacting particle systems*. Springer, Berlin-Heidelberg, Germany, 1999.

[Kur70] T.G. Kurtz. Solutions of ordinary differential equations as limits of pure jump processes. *Journal of Applied Probability*, 7(1):49–58, 1970.

[Kur72] T. G. Kurtz. The relationship between stochastic and deterministic models for chemical reactions. *The Journal of Chemical Physics*, 57(7):2976–2978, 1972.

[Léo95] C. Léonard. Large deviations for long range interacting particle systems with jumps. *Annales de l'Institut Henri Poincaré, section B*, 31(2):289–323, 1995.

[Mae17] C. Maes. Frenetic bounds on the entropy production. *ArXiv Preprint*, 1705.07412, 2017.

[MN08] C. Maes and K. Netočný. Canonical structure of dynamical fluctuations in mesoscopic nonequilibrium steady states. *EPL*, 82(3):30003, 2008.

[MPPR] A. Mielke, I.A. Patterson, M.A. Peletier, and D.R.M. Renger. Non-equilibrium thermodynamical principles for chemical reactions with mass-action kinetics (to appear in SIAM Journal on Applied Mathematics). WIAS preprint nr. 2165.

[MPR14] A. Mielke, M.A. Peletier, and D.R.M. Renger. On the relation between gradient flows and the large-deviation principle, with applications to Markov chains and diffusion. *Potential Analysis*, 41(4), 2014.

[Nor97] J.R. Norris. *Markov Chains*. Cambridge University Press, Cambridge, England, 1997.

[Ons31] L. Onsager. Reciprocal relations in irreversible processes I. *Phys. Rev.*, 37(4):405–426, Feb 1931.

[PR16] R.I.A. Patterson and D.R.M. Renger. Dynamical large deviations of countable reaction networks under a weak reversibility condition (submitted). WIAS Preprint nr. 2273, 2016.

[SW95] A. Shwartz and A. Weiss. *Large deviations for performance analysis: queues, communications, and computing*. Chapman & Hall, London, UK, 1995.