

On and beyond Entropy Production: the Case of Markov Jump Processes

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Abstract. How is it that entropy derivatives almost in their own are characterizing the state of a system close to equilibrium, and what happens further away from it? We explain within the framework of Markov jump processes why fluctuation theory can be based on considerations involving entropy production alone when perturbing around the detailed balance condition. Variational principles such as that of minimum entropy production are understood in that way. Yet, further away from equilibrium, dynamical fluctuations reveal a structure where the time-symmetric sector crucially enters. The fluctuations of densities and currents get coupled and a time-symmetric notion of dynamical activity becomes the counterpart and equal player to the entropy production. The results are summarized in an extended Onsager–Machlup Lagrangian, which in its quadratic approximation is expected to be quite general in governing the small fluctuations of nonequilibrium systems whose macroscopic behavior can be written in terms of a Master equation autonomously describing the time-dependence of densities and currents.

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1. Scope

The breaking of time-reversal symmetry is certainly an important feature of nonequilibrium systems. While the underlying microscopic dynamics is (under usual circumstances) time-reversal symmetric, the plausibility of the time-reversed history of mesoscopic or even more macroscopic conditions can greatly differ from that of the original history. These considerations are very much

linked with the concept of entropy and its production. As written by Max Planck in 1926 [20]: “. . . *there is no other general measure for the irreversibility of a process than the amount of increase of entropy.*” As an example, the by now well-known fluctuation symmetries of the entropy production, be it transient or in the steady state, are on a formal level nothing but expressions of that relation between entropy production and time-reversal breaking. That point was especially emphasized in [3, 12, 13].

In particular and even if not always explicit, much emphasis in the study of nonequilibrium phenomena has gone to the study of the entropy production, or to some nonequilibrium extension and generalization of thermodynamic potentials. Nevertheless there are reasons to doubt the unique relevance of the entropy production concept, as traditionally understood, in far from equilibrium set-ups. Similar thoughts have already been expressed longer time ago in [11]. The characterization of nonequilibrium could very well require to consider observations that are somewhat foreign to equilibrium thermodynamics. Entropy production governs equilibrium fluctuations and remains useful for close-to-equilibrium processes via the concept of heterogeneous equilibrium. Yet, that *hydrodynamic experience* is mostly related to the problem of return to equilibrium. For all we can imagine, perhaps other quantities must complement the entropy production to account for other relevant nonequilibrium features that have to do not only with dissipation but perhaps also with more constructive aspects of the nonequilibrium kinetics and its dynamical activity.

We are interested here in the fluctuation functionals for the nonequilibrium statistics of state-occupations and of state-transitions. Much research into this has already been done, and for a review of this we refer to [1]. Our emphasis lies on the *joint* fluctuations of these time-symmetric and time-antisymmetric sectors of the dynamical fluctuations. The most important observations will be, that these sectors are coupled and that they are not solely determined by the stationary entropy production. A similar emphasis was put already in the treatment of the steady state statistics of diffusions, [16], and in the elucidation of a canonical structure of the steady fluctuations [15]. Here we are adding the discussion on the transient regime and for the steady state we concentrate fully on the small fluctuations around the stationary values. That suffices to appreciate the appearance of a new quantity, that we have called *traffic* and that measures the dynamical activity in a time-symmetric way.

The type of nonequilibrium systems to which we believe our analysis applies almost literally are composed of weakly interacting particles, as in a driven dilute gas for which a Boltzmann–Grad limit can be taken, or as in a driven Lorentz gas, or consist of a multi-level system in contact with particle or heat reservoirs. The latter are frequently encountered in quantum transport systems on the nanoscale. When dealing with interacting particles, we would think that the line of analysis can be kept but interesting new behavior, including phase

transitions, can result and be accompanied by a less trivial application of the theory of large deviations. We refer to [7] and references therein for an update.

The text is not a fully mathematical treatment. Our excuse is that we think more important today to put the physical concepts in place and to suggest a fruitful line of physical reasoning. Moreover, the mathematical formulation and proofs are expected to be rather straightforward, and not adding substantially to the interest of the paper. Nevertheless, we realize that the paper can still appear a bit heavy on the formal side; no standard examples are included from the recent nonequilibrium literature. We hope that future contributions will remedy that.

The next section presents the set-up; the particular framework is that of Markov jump processes to be used as start for a nonequilibrium thermodynamics of free particles. Section 3 reminds us of the notion of entropy production. A separate Section 4 is devoted to the notion of dynamical fluctuations. The rest of the paper analyzes the resulting generalized Onsager – Machlup Lagrangian, first around equilibrium in Section 5.1 and then for more arbitrary nonequilibrium conditions in Section 5.2.

2. Set-up

Imagine a large number N of degrees of freedom (x_t^1, \dots, x_t^N) evolving in continuous time t . The case we consider is that of a collection of jump processes, with a common state space Ω . On the level of a macroscopic description, two types of empirical averages present themselves: first,

$$p_t^N(x) = \frac{1}{N} \sum_{k=1}^N \chi[x_t^k = x], \quad x \in \Omega \quad (2.1)$$

where the indicator function χ , possibly understood in a distributional sense, gives the state-occupation. Secondly, there is the empirical distribution of jumps $x \rightarrow y$ for all pairs of states $x, y \in \Omega$, that we write in the form

$$\frac{1}{N} \#\{\text{jumps } x \rightarrow y \text{ within } [t, t + \tau]\} = \tau p_t^N(x) k_t^N(x, y) + o(\tau) \quad (2.2)$$

that adds the empirical jump rates. The p^N and k^N are not completely arbitrary but they have to satisfy the consistency (or balance) relation, per trajectory,

$$\frac{dp_t^N(x)}{dt} = \sum_{y \neq x} \{p_t^N(y) k_t^N(y, x) - p_t^N(x) k_t^N(x, y)\}. \quad (2.3)$$

One recognizes in (2.3) the form of a Master equation. It is one of the challenges of nonequilibrium statistical mechanics to actually derive such Master equations from more microscopic evolution laws. Here we ignore that problem

and we actually *start* from the x_t^i as a collection of independent and identical Markov jump processes. To make it even simpler, we assume that Ω is finite and that the process (x_t^i) is ergodic with unique stationary law ρ . All that is believed not to be extremely important, as we have in mind the $N \uparrow +\infty$ limit. The possibility of phase-transitions or of *non-smooth* behavior is not considered here in exchange for a thorough look at the fluctuation theory of the $(p_t^N, k_t^N)_t$ and derived quantities.

2.1. Macroscopic limit

We consider a collection of identical independent ergodic continuous time Markov processes x_t^i , each taking values in the finite state space Ω and with rates $\lambda(x, y) \geq 0$ for jumps between the states $x \rightarrow y$. We interpret the process $(x_t^i)_t$ as the random trajectory of the i th particle, where randomness refers to some reduced description where further degrees of freedom are integrated out possibly in combination with some particular limiting procedure. On the macroscopic level, we deal with the trajectory $(p_t^N, k_t^N)_t$ from (2.1) and (2.2). It defines the whole empirical process which is (time-inhomogeneous) Markov even for finite N by construction; note that we do not include three- and higher-time empirical correlations into our macroscopic description. From the law of large numbers, the random occupations $(p_t^N)_t$ concentrate in the limit $N \rightarrow \infty$ on the unique solution of the Master equation

$$\frac{dp_t(x)}{dt} = \sum_{y \neq x} \{p_t(y)\lambda(y, x) - p_t(x)\lambda(x, y)\}. \quad (2.4)$$

2.2. Path distribution

The trajectories of the particle do not all have the same probability. And the same trajectory has different probabilities depending on the rates of the process. All that can be studied via standard tools for comparing probability densities, in particular via the so called Girsanov formula for Markov processes. For our context, the density of one path-space measure \mathcal{P}_μ over a time T , starting at probability law μ , with respect to another one $\bar{\mathcal{P}}_{\bar{\mu}}$ is given by

$$\frac{d\mathcal{P}_\mu(\omega)}{d\bar{\mathcal{P}}_{\bar{\mu}}} = \frac{\mu(x_0)}{\bar{\mu}(x_0)} \exp \left\{ - \int_0^T (\xi(x_t) - \bar{\xi}(x_t)) dt + \sum_{0 < t < T} \log \frac{\lambda(x_{t-}, x_t)}{\bar{\lambda}(x_{t-}, x_t)} \right\} \quad (2.5)$$

where $\omega = (x_t)_0^T$, $x_t \in \Omega$, is a piecewise constant trajectory (or path) and, in the first integral of the exponent, the $\xi(x) = \sum_{y \neq x} \lambda(x, y)$ are escape rates; the last sum in the exponent is over the jump times t where the path takes x_t to x_{t+} . As a convention, we always take right-continuous versions of the trajectories. As usual with probability densities, there is the assumption of absolute continuity

making the indefiniteness not worse than giving weight zero to terms of the form $0/0$. Mathematical details and derivation can be found in e.g. Appendix 2 of [9].

That is useful for our fluctuation theory as we can obtain the probability of an event as the density of the original process with respect to a new process which makes the event typical, conditioned on that event. That is sometimes referred to as the Cramer-trick in the theory of large deviations; a gentle introduction is contained in [22].

Remark that the exponent in (2.5) contains two terms, the first one (with the escape rate) is time-symmetric, the second one is time-antisymmetric. In fact, soon we will see (in Sections 3.1 and 3.4) that the time-antisymmetric part in the action governing the path-space distribution is exactly the entropy production.

2.3. Relation to thermodynamics: local detailed balance

Up to here, we have only statistically defined our model. To get a physical (measurable) interpretation we should associate thermodynamics to it. In the case of equilibrium, this is well-known: by equilibrium we mean that case where

$$\rho(x)\lambda(x, y) = \rho(y)\lambda(y, x) \quad (2.6)$$

where $\rho(x) \propto \exp\{-\beta U(x)\}$ is a Gibbs-distribution. This relation expresses a reversal symmetry for each of the transitions $x \rightleftharpoons y$, which finally amounts to the time-reversal symmetry of the stationary process. We restrict us here to state spaces for which the kinematical time-reversal is trivial (no velocities). That is a serious restriction, which is typical for chemical reaction networks or for overdamped motion but one should understand that it greatly influences the relation between time-reversal, equilibrium and entropy production.

For models of nonequilibrium systems, a thermodynamic interpretation becomes difficult because one expects that the condition of detailed balance (2.6) is broken. What replaces it, is either derived from more microscopic models or is assumed. What guides that procedure is known as the condition of local detailed balance. For our purposes we can write it in terms of an energy function $U(x)$ and a work function (or driving) $F(x, y) = -F(y, x)$ to assign rates to the transitions between each x and y , satisfying

$$\frac{\lambda(x, y)}{\lambda(y, x)} = e^{\beta(F(x, y) + U(x) - U(y))} \quad (2.7)$$

where $\beta \geq 0$ is a parameter that stands for the inverse temperature of a reference reservoir.

The fundamental reason for local detailed balance is the time-reversibility of an underlying microscopic dynamics over which our effective stochastic model

is presumably built. Hence, violating such a condition reduces the physical interpretation of our stochastic model. As further explained in the next section, condition (2.7) is also intimately related to the symmetries of nonequilibrium fluctuations and to the role of entropy production in there. At any event, (2.7) allows to write

$$\rho_o(x)\lambda(x, y) = \gamma(x, y)e^{(\beta/2)F(x, y)} \quad (2.8)$$

with $\rho_o(x) = \exp\{-\beta U(x)\}/Z$ a reference equilibrium probability distribution and some symmetric $\gamma(x, y) = \gamma(y, x)$, which is left unspecified. To reveal the meaning of γ , notice that in equilibrium, i.e. for $\rho = \rho_o$ and $F = 0$, one has $2\gamma(x, y) = \rho(x)\lambda(x, y) + \rho(y)\lambda(y, x)$. The right-hand side is the expectation of the empirical observable

$$\tau_t^N(x, y) = p_t^N(x)k_t^N(x, y) + p_t^N(y)k_t^N(y, x) \quad (2.9)$$

that measures the time-symmetric dynamical activity (the total number of jumps across the bond (x, y)) and we call it *traffic*. As we will see in Sections 6 and 5, the traffic is a crucial quantity to characterize the nonequilibrium fluctuations far from equilibrium.

3. Entropy production

The notion of entropy production should not be fully re-invented when dealing with Markov jump processes. It must match with the thermodynamic or hydrodynamic interpretations. We start however with a view that goes beyond model-specifics and that emphasizes the relation with time-reversal.

3.1. Statistical interpretation: time-irreversibility

Dynamical time-reversal plays on the level of single trajectories $\omega = (x_t)_0^T$. We define the time-reversal as $\theta\omega = (x_{T-t})_0^T$, not indicating the trivial modifications at the jump times for restoring the right-continuity of paths. If we denote the original Markov process started at distribution μ by \mathcal{P}_μ , then there is a time-reversed process $\mathcal{P}_{\mu_T}\theta$ starting at the (time-evolved) distribution μ_T . There is a density of one with respect to the other, and that we call the (variable, fluctuating) entropy production

$$S_\mu^T(\omega) = \log \frac{d\mathcal{P}_\mu}{d\mathcal{P}_{\mu_T}\theta}(\omega). \quad (3.1)$$

We can use the Girsanov formula (2.5) for its computation, see the details in [17]. That formula (3.1) captures the idea of the entropy production as measuring the amount of time-reversal breaking. The so called fluctuation theorem, steady or transient, time-dependent or not, very much rests on that unifying idea [13].

For a foundation starting from the Hamiltonian dynamics and microcanonical ensemble, see [12]. We come back to fluctuation relations in Section 3.4.

It is interesting to note that by convexity $\langle S_\mu^T \rangle_\mu \geq 0$ as it should for an entropy production, where the brackets take the average with respect to \mathcal{P}_μ , the path-space measure starting at μ .

Being interested in an instantaneous (average) entropy production rate when the distribution is μ , we define

$$\sigma(\mu) = \lim_{T \downarrow 0} \frac{1}{T} \left\langle \log \frac{d\mathcal{P}_\mu}{d\mathcal{P}_{\mu_T} \theta} \right\rangle_\mu \quad (3.2)$$

so that, by the Markov property, $\langle S_\mu^T \rangle_\mu = \int_0^T \sigma(\mu_t) dt$ with $(\mu_t)_0^T$ the time-evolved measures. The instantaneous entropy production rate $\sigma(\mu)$ can easily be computed for our Markov process:

$$\sigma(\mu) = \sum_{(xy)} [\mu(x)\lambda(x, y) - \mu(y)\lambda(y, x)] \log \left(\frac{\mu(x)\lambda(x, y)}{\mu(y)\lambda(y, x)} \right). \quad (3.3)$$

The notation (xy) under the sum will from now on be used to mean that we sum over unordered pairs of states.

The previous expressions do make physical sense even for a single Markov process defining a dynamics for a small finite number of degrees of freedom, thinking of an open system effectively coupled to and/or driven by large external reservoirs. It becomes however more physically transparent when formulated in terms of the empirical distribution as explained next.

3.2. Thermodynamic interpretation

An open system dissipates heat that results in a change of entropy in the environment. Assuming a large environment we can compute it as the reversible heat. From the first law of thermodynamics that dissipated heat is identical to the work plus the change in internal energy. So, again in our ensemble-interpretation, the rate of change of energy is $-\sum_{x,y} j_t^N(x, y) U(x)$ with, see (2.3),

$$j_t^N(x, y) = p_t^N(x) k_t^N(x, y) - p_t^N(y) k_t^N(y, x) \quad (3.4)$$

being the empirical currents, and the power is $\sum_{(xy)} j_t^N(x, y) F(x, y)$. If therefore the empirical currents at time t equal $j_t^N(x, y) = j(x, y)$, then the dissipated heat is

$$\mathcal{Q}(j) = \sum_{(xy)} j(x, y) (U(x) - U(y) + F(x, y)) \quad (3.5)$$

and the entropy current is $\beta \mathcal{Q}(j)$ (setting Boltzmann's constant equal to one) for an environment at temperature β^{-1} .

Secondly, there is the change of the entropy of the system itself. Here we only have the densities (2.1) as macroscopic variable and

$$S_{\text{sys}}(p) = - \sum_x p(x) \log p(x) \quad (3.6)$$

is the (static) fluctuation functional in the probability law for observing the empirical density p when sampling the particles from the flat distribution. Its change in time is the internal entropy production: $\dot{S}_{\text{sys}}(p, j) = \sum_{(xy)} j(x, y) \times (\log p(x) - \log p(y))$. Summing it up, we get the total (macroscopic) entropy production rate

$$\dot{S}(p, j) \equiv \dot{S}_{\text{sys}}(p, j) + \beta \mathcal{Q}(j) \quad (3.7)$$

for the empirical values p and j for densities (2.1) and currents (3.4), respectively.

3.3. Relating the two interpretations

Using the local detailed balance condition (2.7), the macroscopic entropy production rate in (3.7) is

$$\dot{S}(p, j) = \sum_{(xy)} j(x, y) \log \frac{p(x)\lambda(x, y)}{p(y)\lambda(y, x)} \quad (3.8)$$

in terms of the instantaneous densities $p(x)$ and currents $j(x, y) = p(x)k(x, y) - p(y)k(y, x)$. Remember that $p(x)k(x, y)$ is the fraction of particles that actually make the transition $x \rightarrow y$, and $j(x, y)$ is the (net) current of particles. By the law of large numbers, the typical value of these currents at given densities $p(x)$ is $p(x)\lambda(x, y) - p(y)\lambda(y, x)$ and hence the typical entropy production rate (3.8) just coincides with $\sigma(p)$, see (3.3). This not only justifies our form of the local detailed balance assumption, it also explains the relation between the single Markov process formalism of Section 3.1 and the empirical description for an ensemble of the processes, cf. (2.1)–(2.2); this duality is exploited throughout the whole text.

A different decomposition that is equally useful (and used in the following subsection) writes

$$\dot{S}(p, j) = \dot{S}_{\text{ext}}(j) + \dot{S}_{\text{int}}(p, j) \quad (3.9)$$

for the entropy current

$$\dot{S}_{\text{ext}}(j) = \sum_{(xy)} j(x, y) F(x, y) \quad (3.10)$$

in excess with respect to the equilibrium reference, and

$$\dot{S}_{\text{int}}(p, j) = \sum_{(xy)} j(x, y) \left(\log \frac{p(x)}{\rho_o(x)} - \log \frac{p(y)}{\rho_o(y)} \right)$$

is now the rate of change of the system's *relative* entropy (always summing over pairs). Note that this decomposition differs only from the former (3.7) in the use of another reference. In (3.7) the reference is the flat distribution. To end this section we review two simple applications of the single process formalism of Section 3.1.

3.4. Fluctuation relations

The decomposition (3.9) into the internal and external change of the entropy can equivalently be done pathwise for a single process, starting from (3.1). Note that it depends on the choice of the reference equilibrium process; using the notation $\mathcal{P}_{\rho_o}^o$ for such a reference started from a reversible measure ρ_o . Then (2.5) can be written in the form $d\mathcal{P}_\mu(\omega) = d\mathcal{P}_{\rho_o}^o(\omega) [\mu(x_0)/\rho_o(x_0)] \exp\{-A(\omega)\}$ with the action A that can be read from (2.5). Since the reference process is time-reversal invariant, we can now rewrite (3.1) as

$$S_\mu^T(\omega) = \log \frac{\mu(x_0)\rho_o(x_T)}{\mu_T(x_T)\rho_o(x_0)} + A(\theta\omega) - A(\omega).$$

That corresponds to the decomposition (3.9) and we call $S_{\text{ext}}(\omega) = A(\theta\omega) - A(\omega)$ the (variable) entropy flux for a single chain, in excess with respect to the reference equilibrium process. Obviously, we also have

$$S_{\text{ext}}(\omega) = \log \frac{d\mathcal{P}_{\rho_o}}{d\mathcal{P}_{\rho_o}\theta}(\omega)$$

and hence, for all path-dependent observables f ,

$$\langle f \rangle_{\rho_o} = \langle f\theta \exp(-S_{\text{ext}}) \rangle_{\rho_o} \quad (3.11)$$

which gives an exact (for all finite times T) symmetry in the distribution of the (excess) entropy flux $S_{\text{ext}} = -S_{\text{ext}}\theta$ at least when started from the reference equilibrium. Steady fluctuation symmetries are then obtained as the asymptotics for $T \uparrow +\infty$. Note that in general one needs to deal with the temporal boundary term. However, in the present framework of ergodic Markov processes over a finite state space the dependence on the initial condition is irrelevant.

The fluctuation symmetry (3.11) also has a formulation in terms of the macroscopic fluctuation theory within the ensemble formalism of the previous subsection; this will be discussed at the end of Section 4.1.

3.5. Stationary measure

One may wonder how the above considerations are reflected on the level of the stationary distribution ρ itself. That in fact is the subject of earlier work by Zubarev and by McLennan [18]: what is a first order correction around a

reference equilibrium/detailed balance, and is there a systematic perturbation theory? There are a number of ways to discuss that question. One possible direction is to try to formulate a variational principle for the ρ ; this approach will be discussed later. Another, more direct approach is to compute the asymptotics $T \uparrow +\infty$ of the time-evolved measure μ_T or, equivalently, to project the path-space distribution \mathcal{P}_μ^T on the time T , again asymptotically. As explained in a recent preprint [10], the latter approach can be conveniently started from the fluctuation symmetry (3.11). Indeed, by taking $f(\omega) = \chi[x_T = x] \exp(-S_{\text{ext}}(\omega)/2)$, one has $f\theta(\omega) = \chi[x_0 = x] \exp(S_{\text{ext}}(\omega)/2)$ and therefore

$$\left\langle \chi[x_T = x] \exp\left(-\frac{S_{\text{ext}}}{2}\right) \right\rangle_{\rho_o} = \left\langle \chi[x_0 = x] \exp\left(-\frac{S_{\text{ext}}}{2}\right) \right\rangle_{\rho_o}.$$

As a consequence, the probability to see x at time T when started from reference equilibrium ρ_o is

$$\mathbf{P}_{\rho_o}^T(x_T = x) = \rho_o(x) \frac{\langle \exp(-S_{\text{ext}}/2) \rangle_{x_0=x}}{\langle \exp(-S_{\text{ext}}/2) \rangle_{x_T=x, \rho_o}} \quad (3.12)$$

where we have to condition on the final-time event $x_T = x$ in the denominator. The ratio is one for the equilibrium dynamics, and the nonequilibrium correction is made by the time-asymmetry in the fluctuations of the entropy production. An advantage of this representation of the evolved measure lies in the cancellation of various nontransient (i.e. unbounded upon T growing) terms when expanding the exponents, so that the limit $T \uparrow +\infty$ can be controlled, see [10] for more details.

4. Dynamical fluctuations

In the ensemble picture, trajectories $\omega^N = (x_t^1, \dots, x_t^N)_t$ have their coarse-grained counterparts in the empirical distributions $(p_t^N)_t$ and the empirical rates $(k_t^N)_t$. They fluctuate around their typical values ρ and λ , the typicality being in the sense of a law of large numbers with N as the large parameter. Computing the probability of the event $p_t^N = p_t, k_t^N = k_t$ for all $0 \leq t \leq T$ is done via the Girsanov formula (2.5), by comparing the system with modified dynamics (such that p_t and k_t are typical) with the original system. Clearly, a macroscopic trajectory $(p_t, k_t)_t$ satisfying the consistency condition (2.3) becomes typical under the modified (time-inhomogeneous) Markov dynamics with the rates $(k_t)_t$ and the initial measure p_0 . Its probability with respect to the original i.i.d. Markov processes x^1, \dots, x^N started each from the distribution μ has the large deviation form

$$\mathbf{P}_\mu^N \{ (p_t^N = p_t, k_t^N = k_t)_{0 \leq t \leq T} \} \doteq \exp \left\{ -N \left[S(p_0 | \mu) + \int_0^T dt \mathbb{L}(p_t, k_t) \right] \right\} \quad (4.1)$$

where \doteq refers to the logarithmic equivalence as $N \rightarrow \infty$, and the relative entropy \mathcal{S} and the Lagrangian \mathbb{L} are

$$\mathcal{S}(p_0 | \mu) = \sum_x p_0(x) \log \frac{p_0(x)}{\mu(x)}, \tag{4.2}$$

$$\mathbb{L}(p, k) = \sum_{x, y \neq x} p(x) \left[k(x, y) \log \frac{k(x, y)}{\lambda(x, y)} - k(x, y) + \lambda(x, y) \right]. \tag{4.3}$$

In particular, the Lagrangian on this level of description has a simple explicit form, irrespective of any detailed balance or stationarity assumptions. It is therefore a natural point of departure for the investigation of also more coarse-grained dynamical fluctuations.

Next we consider one step of such a conceivable hierarchy.

4.1. Lagrangian for currents

A quantity of special interest is the collection of empirical currents $j_t^N(x, y) = -j_t^N(y, x)$ given in (3.4). The Lagrangian $\mathcal{L}(p, j)$ that governs the joint occupation-current dynamical fluctuations is

$$\mathcal{L}(p, j) = \inf_k \{ \mathbb{L}(p, k) \mid p(x)k(x, y) - p(y)k(y, x) = j(x, y); \forall x, y \} \tag{4.4}$$

where j is an arbitrary antisymmetric current matrix. The distribution of empirical trajectories $(p_t^N, j_t^N)_t$ follows from the large deviation law (4.1) via the contraction principle:

$$\mathbf{P}_\mu^N \{ (p_t^N = p_t, j_t^N = j_t)_{0 \leq t \leq T} \} \doteq \exp \left\{ -N \left[\mathcal{S}(p_0 | \mu) + \int_0^T dt \mathcal{L}(p_t, j_t) \right] \right\}$$

whenever the consistency constraint (2.3), $\dot{p}_t(x) + \sum_{y \neq x} j_t(x, y) = 0$, is satisfied. It can be made explicit by the method of Lagrange multipliers. One finds $\mathcal{L}(p, j) = \mathbb{L}(p, k^j)$ where the k^j solve the equations

$$k^j(x, y) = \lambda(x, y) \exp\{(\beta/2)\psi^j(x, y)\}, \tag{4.5}$$

$$j(x, y) = p(x)k^j(x, y) - p(y)k^j(y, x), \tag{4.6}$$

for some specific $\psi^j(x, y) = -\psi^j(y, x)$, or explicitly,

$$k^j(x, y) = \frac{1}{2p(x)} \{ j(x, y) + [j^2(x, y) + 4p(x)p(y)\lambda(x, y)\lambda(y, x)]^{1/2} \}. \tag{4.7}$$

Hence, the typical macroevolution constrained by fixing the currents to some j becomes *unrestrainedly typical* by modifying correspondingly the antisymmetric part of the transition rates, as in (4.5).

From (4.7) we check that $p(x)k^j(x, y) = p(y)k^{-j}(y, x)$. It is then straightforward to derive that

$$\mathcal{L}(p, -j) - \mathcal{L}(p, j) = \dot{S}(p, j) \tag{4.8}$$

so that the entropy production rate (3.8) is (indeed) the time-antisymmetric part of the Lagrangian. As a consequence, always in the logarithmic sense and in the limit $N \uparrow +\infty$,

$$\begin{aligned} & \frac{\mathbf{P}_\mu^N \{ (p_t^N = \bar{p}_t, j_t^N = \bar{j}_t)_{0 \leq t \leq T} \}}{\mathbf{P}_{\mu_T}^N \{ (p_t^N = p_{T-t}, j_t^N = -j_{T-t})_{0 \leq t \leq T} \}} \\ & \doteq \exp \left\{ N \left[S(p_T | \mu_T) - S(p_0 | \mu) + \int_0^T dt \dot{S}(p_t, j_t) \right] \right\} \end{aligned} \tag{4.9}$$

which is a macroscopic variant of the fluctuation relations described in Section 3.4.

4.2. Fluctuations of empirical time averages

Instead of looking at the probabilities of specific macroscopic trajectories of the system, we now consider empirical time averages:

$$\bar{p}_T^N = \frac{1}{T} \int_0^T p_t^N dt, \quad \bar{j}_T^N = \frac{1}{T} \int_0^T j_t^N dt. \tag{4.10}$$

For a fixed initial distribution μ , the asymptotic ($T \uparrow +\infty$) probability that the empirical time averages are equal to some density p and current j is

$$\mathbf{P}_\mu^{N,T} \{ \bar{p}_T^N = p, \bar{j}_T^N = j \} \doteq e^{-N \mathcal{A}_T(p,j)} \tag{4.11}$$

with the rate \mathcal{A}_T given by

$$\mathcal{A}_T(p, j) = \inf_{p_t, j_t} \left\{ S(p_0 | \mu) + \int_0^T dt \mathcal{L}(p_t, j_t) \Big| \bar{p}_T = p, \bar{j}_T = j \right\} \tag{4.12}$$

where the infimum is over all macrotrajectories $(p_t, j_t)_{0 \leq t \leq T}$ such that $\dot{p}_t(x) + \sum_{y \neq x} j_t(x, y) = 0$. The infimum in (4.12) is easy to compute in the limit of infinite time span, $T \uparrow +\infty$, in which the minimizing trajectory becomes essentially constant, $(p_t, j_t)_t \equiv (p, j)$, and the initial distribution loses its relevance. One obtains $\mathcal{A}_T(p, j) = T \mathcal{L}(p, j) + o(T)$, yielding

$$\mathbf{P}^{N,T} \{ \bar{p}_T^N = p, \bar{j}_T^N = j \} \doteq e^{-NT \mathcal{L}(p,j)} \tag{4.13}$$

whenever the currents are stationary, $\sum_{y \neq x} j(x, y) = 0$. The equality is meant in the logarithmic sense and after taking first the limit $N \uparrow +\infty$ and then the limit $T \uparrow +\infty$. For details on these manipulations and techniques from the theory of large deviations, we refer to [4–6, 8, 22].

It thus appears that the Lagrangian \mathcal{L} of (4.4)–(4.7) governs the joint steady statistics of time-averaged occupations and currents. Its study has already been started in [15, 16], emphasizing a canonical structure. As it is fully explicit, one can use it as variational functional to characterize the steady state, and for further contractions to obtain variational functionals for the occupations and currents separately. That is however not the subject of the present paper. What comes in the sequel is an analysis of the structure of the above Lagrangians in the quadratic approximation and its physical consequences.

5. Structure of normal fluctuations

The most accessible fluctuations are small — both mathematically and practically. The Lagrangians can be expanded in both the densities and currents around their typical (= steady) values and the strictly positive quadratic form obtained in the leading order describes normal fluctuations. From a physical point of view, the structure of these normal fluctuations have been first analyzed by Onsager and Machlup, [19], for the case of relaxation to equilibrium. Here we show a natural extension of the original Onsager–Machlup formalism to nonequilibrium systems by starting from the above macroscopic fluctuation theory.

We distinguish the two following scaling regimes: first, we analyze the nonequilibrium fluctuations in the immediate vicinity of a reference equilibrium through the scaled Lagrangian

$$L^0(u, j; F) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \mathcal{L}(\rho_o + \varepsilon \rho_o u, \varepsilon j; \varepsilon F) \quad (5.1)$$

where we have explicitly denoted here the dependence on the work function F as it quantifies the distance from equilibrium and participates in the scaling. The Lagrangian corresponds to the dynamics (2.8) with some γ , ρ_0 , and β fixed.

Second, we consider a steady state arbitrarily far from equilibrium and examine the structure of small deviations through the function

$$L(v, j; F) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \mathcal{L}(\rho + \varepsilon \rho v, \bar{j} + \varepsilon j; F). \quad (5.2)$$

Note that the work function is kept fixed here and both the density and current are expanded around the stationary values ρ respectively \bar{j} . We will see below how the structure of fluctuations remarkably changes in this regime.

5.1. Close to equilibrium

Starting from the dynamics with transition rates parameterized as in (2.8), $\lambda(x, y) = \rho_0^{-1}(x)\gamma(x, y) \exp\{(\beta/2)F(x, y)\}$, the scaled Lagrangian (5.1) is easily computed from (4.4)–(4.7):

$$\begin{aligned} L^0(u, j; F) &= \sum_{(xy)} \frac{1}{4\gamma(x, y)} \{j(x, y) - \gamma(x, y)[u(x) - u(y) + \beta F(x, y)]\}^2 \\ &= \frac{1}{2} \left[\frac{1}{2} \mathcal{D}(j) + \frac{1}{2} \mathcal{E}(u) - \dot{s}(u, j) \right] \end{aligned} \quad (5.3)$$

where we have introduced the scaled entropy production, cf. (3.8),

$$\dot{s}(u, j) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \dot{S}(\rho_o + \varepsilon \rho_o u, \varepsilon j; \varepsilon F) = \sum_{(xy)} j(x, y)[u(x) - u(y) + \beta F(x, y)] \quad (5.4)$$

and the pair of (variant Onsager–Machlup) dissipation functions

$$\mathcal{D}(j) = \sum_{(xy)} \frac{j^2(x, y)}{\gamma(x, y)}, \quad \mathcal{E}(u) = \sum_{(xy)} \gamma(x, y)[u(x) - u(y) + \beta F(x, y)]^2. \quad (5.5)$$

In contrast with the equilibrium Onsager–Machlup theory [19], we keep here the currents j as the variables of the Lagrangian.

Fixing some u , the typical current j^u minimizes the Lagrangian, which has the immediate solution

$$j^u(x, y) = \gamma(x, y)[u(x) - u(y) + \beta F(x, y)]. \quad (5.6)$$

That variational problem for j^u , i.e. $(1/2)\mathcal{D}(j) - \dot{s}(u, j) = \min$, is known as the Onsager *least dissipation principle*. Equivalently, it is sometimes formulated as a *transient maximum entropy production principle*: the j^u solves $\dot{s}(u, j) = \max$ under the constraint $\mathcal{D}(j) = \dot{s}(u, j)$.

One checks that the dissipation function $\mathcal{E}(u)$ is a scaled version of the mean entropy production rate (3.3):

$$\mathcal{E}(u) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \sigma(\rho_o + \varepsilon \rho_o u; \varepsilon F) \quad (5.7)$$

where F again enters via the parametrization of the rates (2.8). This is in accord with the equality

$$\dot{s}(u, j^u) = \mathcal{E}(u) = \mathcal{D}(j^u) \quad (5.8)$$

following from (5.3).

The consistency condition (2.3) determines the typical macroscopic trajectory as a solution of the equation

$$\rho_o(x) \frac{du_t(x)}{dt} + \sum_{y \neq x} j^{u_t}(x, y) = 0 \tag{5.9}$$

for all x , with j^u the typical current (5.6). In particular, the stationary distribution $\rho = \rho_o + \varepsilon \bar{u} + o(\varepsilon)$ is in this scaling limit found from the (linearized version of the) Master equation

$$\sum_{y \neq x} \gamma(x, y) [\bar{u}(x) - \bar{u}(y) + \beta F(x, y)] = 0 \tag{5.10}$$

that has to be solved under the normalization constraint $\sum_x \rho_o(x) \bar{u}(x) = 0$. Alternatively, the (linearized) stationary density \bar{u} and the corresponding steady current $\bar{j} = j^{\bar{u}}$ can be found by minimizing the Lagrangian (5.3) subject to the stationary constraint $\sum_{y \neq x} j(x, y) = 0$. Note a remarkable simplification: because of this constraint, the entropy production (5.4) equals $\dot{s}(u, j) = \beta \sum_{(xy)} j(x, y) F(x, y)$ and hence it is independent of u . As a consequence, the density and current become decoupled in the Lagrangian (5.3). By the arguments of Section 4.2 this means that the time-averages \bar{p}_T and \bar{j}_T are uncorrelated in the close-to-equilibrium regime and within the quadratic approximation. An immediate consequence of this observation is a simple structure of the marginal distributions of u respectively j that provides a fluctuation-based justification of the two familiar stationary variational principles — the minimum and the maximum entropy production principles — as we explain next. See [2, 14–16] for some more details and illustrations.

MinEP principle. We consider the marginal distribution of the empirical time-average \bar{p}_T^N defined in (4.10). By (4.13) and in the present scaling limit the asymptotic law of \bar{p}_T^N reads

$$\begin{aligned} & - \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \lim_{T \uparrow +\infty} \lim_{N \uparrow +\infty} \log \mathbf{P}^{N, T} \{ \bar{p}_T^N = \rho_o + \varepsilon \rho_o u \} \\ & = \inf_j \left\{ L^0(u, j; F) \mid \sum_{y \neq x} j(x, y) = 0 \right\} \\ & = \frac{1}{4} [\mathcal{E}(u) - \mathcal{E}(\bar{u})] \end{aligned} \tag{5.11}$$

where the last equality follows from (5.8) by using the decoupling between u and j under the stationary constraint. The minimum entropy production principle immediately follows: $\mathcal{E}(u) \geq \mathcal{E}(\bar{u})$ with the equality only if $u = \bar{u}$, hence, the stationary measure minimizes the entropy production rate, cf. (5.7).

MaxEP principle. We proceed analogously for the time-averaged empirical current (4.10). For any j satisfying the stationary condition $\sum_{x,y} j(x,y) = 0$ we have by the contraction principle from (4.13):

$$\begin{aligned} & -\lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \lim_{T \uparrow +\infty} \lim_{N \uparrow +\infty} \log \mathbf{P}_\mu^{N,T} \{ \bar{j}_T^N = \varepsilon j \} \\ & = \inf_u L^0(u, j; F) \\ & = (1/2)[(1/2)\mathcal{D}(j) + (1/2)\mathcal{E}(\bar{u}) - \dot{s}(j)]. \end{aligned} \quad (5.12)$$

(Remember that \dot{s} is independent of u under the stationary condition.) Restricting the set of currents even further by imposing the condition $\mathcal{D}(j) = \dot{s}(j)$, the above equals

$$(5.12) = (1/4)[\mathcal{D}(\bar{j}) - \mathcal{D}(j)] = (1/4)[\dot{s}(\bar{j}) - \dot{s}(j)]. \quad (5.13)$$

This in particular yields that the stationary current maximizes the entropy production rate under the above two constraints, which is an instance of the stationary maximum entropy production principle.

5.2. Far from equilibrium

As we have seen the most remarkable feature of small fluctuations in the close-to-equilibrium regime is that the empirical distributions of occupations and of currents become uncorrelated. This appears to be the fundamental reason for the entropy production principles discussed in the previous section to be valid. An important novel feature of the nonequilibrium statistics beyond the close-to-equilibrium regime is that both empirical observables get coupled as we can demonstrate via the other scaling limit introduced in (5.2). There we do an expansion up to leading order around the stationary density ρ and the corresponding stationary current $\bar{j}(x,y) = \rho(x)\lambda(x,y) - \rho(y)\lambda(y,x)$; recall that F and hence the rates $\lambda(x,y)$ remain fixed now. Using also the notation

$$\bar{\tau}(x,y) = \rho(x)\lambda(x,y) + \rho(y)\lambda(y,x) \quad (5.14)$$

for the steady traffic, the scaled Lagrangian (5.2) obtains the form:

$$L(v, j; F) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \mathcal{L}(\rho + \varepsilon \rho v, \bar{j} + \varepsilon j; F) = \sum_{(xy)} \frac{1}{4\bar{\tau}} [j - \bar{\tau} \nabla^- v - \bar{j} \nabla^+ v]^2(x,y) \quad (5.15)$$

with the notation $\nabla^\pm v(x,y) = (1/2)[v(x) \pm v(y)]$. This is the Lagrangian describing normal fluctuations around the typical evolution, with $\bar{\tau} \nabla^- v + \bar{j} \nabla^+ v$ being the typical (or expected) first-order deviation from the steady current \bar{j} . We see that the steady traffic $\bar{\tau}$ plays the role of a variance in this fluctuation law.

In the stationary regime, i.e. under the constraint $\sum_{y \neq x} j(x, y) = 0$, the Lagrangian (5.15) yields the rate function for the joint distribution of time-average occupations and currents, cf. (4.13). We can write it in the form

$$L(v, j; F) = \frac{1}{2} \sum_{(xy)} \left[\frac{1}{2\bar{\tau}} j^2 + \frac{\bar{\tau}}{2} (\nabla^- v)^2 - \frac{\bar{j}}{\bar{\tau}} j \nabla^+ v + \frac{\bar{j}^2}{2\bar{\tau}} (\nabla^+ v)^2 \right] (x, y) \quad (5.16)$$

which demonstrates that the emerged occupation-current coupling is proportional to the stationary current and indeed vanishes only close to equilibrium when moreover $\bar{j} = O(\varepsilon)$.

6. Towards a more general theory

Adding a nonequilibrium driving not only generates nonzero steady currents but it also modifies the steady averages of *time-symmetric* observables and their fluctuation statistics. For a long time, the latter has not been of primary interest in transport considerations, partially because of the success of linear response theories in which only the currents and the entropy production play a fundamental role. The origin of that has been discussed in Section 5.1 on the structure of close-to-equilibrium normal fluctuations in which the time-symmetric and the time-antisymmetric sectors become totally decoupled. Their coupling away from equilibrium, cf. Section 5.2, suggests that some systematic and robust description of nonequilibrium fluctuations might be achieved by analyzing the time-antisymmetric (currents) and the time-symmetric (e.g. the occupation times) observables simultaneously; this is exactly the strategy brought up in the present paper.

6.1. Traffic

An important drawback of the transport theories based on stochastic models is that we only have a direct thermodynamic interpretation for the time-antisymmetric part of the transition rates, cf. the local detailed balance condition (2.7), whereas rather little can *generally* be said about the symmetric part and its dependence on the nonequilibrium driving. Yet, the fluctuation theory can help also here: instead of giving an interpretation to the symmetric part of the rates, one can try to understand the role of the traffic (5.14) as a time-symmetric dynamical observable and a counterpart to the current. We have already seen that in equilibrium the (mean) traffic coincides with $2\gamma(x, y)$, and away from equilibrium it enters, according to (5.16), as a variance for normal dynamical fluctuations. More generally and even beyond the regime of normal fluctuations, it can be shown that the dependence of the traffic on the driving fully determines the structure of nonequilibrium fluctuations in the time-symmetric sector, and also specifies the symmetric-antisymmetric coupling in a canonical way [15, 16]. We give here a brief review of this approach.

The Lagrangian \mathcal{L} introduced in (4.4)–(4.7) has the form

$$\mathcal{L}(p, j) = \sum_{x,y} p(x) \left[k^j(x, y) \frac{\beta}{2} \psi^j(x, y) - k^j(x, y) + \lambda(x, y) \right] \tag{6.1}$$

with the modified rates k^j given by (4.5): they can be thought of as the original rates but with the modified work function $F \rightarrow F + \psi^j$ fixed so that the current j becomes typical,

$$j(x, y) = p(x)k^j(x, y) - p(y)k^j(y, x). \tag{6.2}$$

Therefore,

$$\mathcal{L}(p, j) = \sum_{(xy)} [j(x, y) \psi^j(x, y) - \tau_{p, F+\psi^j}(x, y) + \tau_{p, F}(x, y)] \tag{6.3}$$

with the (mean) traffic $\tau_{p, G} = p(x)\lambda_G(x, y) + p(y)\lambda_G(y, x)$, $\lambda_G(x, y) = \lambda_0(x, y) \times \exp\{(\beta/2)G(x, y)\}$ considered here as a function of the work matrix G ; the $\lambda_0(x, y) = \rho_o^{-1}(x)\gamma(x, y)$ being the rates corresponding to the reversible reference dynamics $G = 0$, cf. (2.8). Note that $\lambda_{F+\psi^j} = k^j$ under the relation (6.2); in fact the driving and the current appear to be conjugated variables in the sense of a canonical formalism, see [15] for details.

Clearly, (6.3) splits in two parts: the first term is to be understood as an *excess* of entropy production and the second one is an excess of overall traffic. Starting from this general scheme, we can calculate the fluctuation rate functions of arbitrary more coarse-grained dynamical observables. If that observable is purely time-symmetric, e.g. only depends on the occupations, then the excess ψ^j is in fact a gradient, $\psi^j(x, y) = V(y) - V(x)$ for state function V , and the first entropy production term in (6.3) vanishes for currents satisfying the stationarity condition $\sum_{y \neq x} j(x, y) = 0$. Then, what remains is the excess traffic as variational functional.

6.2. Why does the entropy production govern close-to-equilibrium?

By construction the traffic functionals are quite different from the entropy production functionals. Yet, close to equilibrium the excess of traffic and the excess of entropy production are related to each other in a simple way: analogously to the scaled mean entropy production (5.7),

$$\mathcal{E}_F(u) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \sigma(\rho_o + \varepsilon \rho_o u; \varepsilon F) \tag{6.4}$$

we consider the scaled overall traffic

$$\mathcal{T}_F(u) = \lim_{\varepsilon \downarrow 0} \varepsilon^{-2} \sum_{(xy)} [\tau_{\rho_o + \varepsilon \rho_o u; \varepsilon F}(x, y) - \tau_{\rho_o + \varepsilon \rho_o u; 0}(x, y)] \tag{6.5}$$

(relatively with respect to the equilibrium reference so that the limit is well defined), and we find the relation

$$\mathcal{T}_F(u) = (1/2)[\mathcal{E}_F(u) - \mathcal{E}_0(u)]. \quad (6.6)$$

Hence, close to equilibrium the mean overall traffic is determined by the mean entropy production. This observation remarkably simplifies the structure of Lagrangian (6.1), and it finally leads to the (generalized) Onsager–Machlup theory of Section 5.1 in which the entropy production and derived quantities are the only players.

To summarize, we understand the dominance of entropy production and the simple structure of the close-to-equilibrium regime as a consequence (1) of the decoupling of the time-symmetric and the time-antisymmetric fluctuations, (2) of the relation between the mean traffic and the mean entropy production in this regime.

6.3. Beyond entropy production far from equilibrium

In a far-from-equilibrium regime the time-symmetric and the time-antisymmetric fluctuations get coupled and the relation (6.6) is no longer valid. This gives a motivation why it is natural to study both dynamical sectors jointly. As we have seen, the entropy production alone is not sufficient to describe fluctuations in either of these sectors, and the traffic functional enters as a new important player in the nonequilibrium fluctuation theory. We hope that more theoretical investigation and also experimental evidence will support this line of research.

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