Flux cycles as building blocks of non-equilibrium steady states

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Abstract. - Detailed balance and the resulting constraints on equilibrium steady states constitute corner stones of statistical physics. No principles of comparable significance are known for non-equilibrium steady states. Here we introduce a representation of non-equilibrium steady-states (that violate detailed balance) in terms of cycle fluxes. We show that on the new space where the states are the original flux cycles, there is a natural dynamics that satisfies detailed balance. The non-equilibrium steady-state occupation numbers of cycles hence follow a Boltzmann distribution, and expectation values of arbitrary observables for the stochastic systems can be expressed as cycle averages, resembling the representation of expectation values in dynamical systems by cycle expansions.

Introduction. – A major challenge of statistical physics is to identify principles organizing the structure of steady states [1,2]. Equilibrium systems are singled out by detailed balance, a symmetry in the transition rates between different states which can be used to explicitly calculate their free energies [3,4]. This symmetry provides a thermodynamic potential which yields all thermodynamic properties of the system. In non-equilibrium steady states (NESS) detailed balance is broken. Consequently, there is no reason to expect that there is a potential function for these systems that allows one to calculate the steady-state properties of the system.

Here we demonstrate that this expectation is premature. In our argument we follow Penrose [5], and idealize observable processes as irreducible, positively recurrent Markov processes on a finite state space. Irreducible means that the system can reach any state i from any other state j with a finite number of transitions. Positive recurrence means that in an infinite time span the system visits every state infinitely often. Together, these assumptions imply ergodicity, and hence ensure the existence of a steady state [6].

Below, we present a principle to map the non-equilibrium steady state of such a system to a Markov process on a dual space where detailed balance is restored. Steady-state averages take the form of equilibrium averages on that dual space.

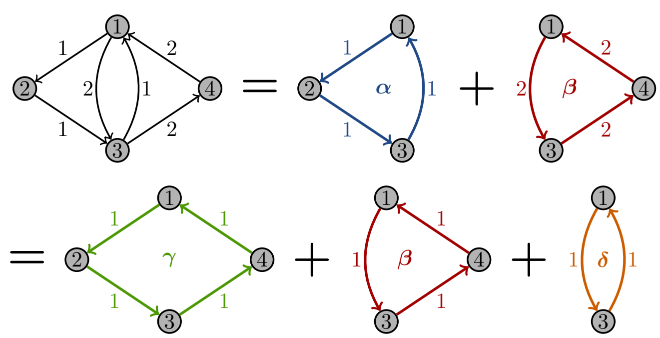

Fig. 1: Representation of a non-equilibrium steady state in terms of linear superpositions of cycle fluxes. The numbers on the arrows representing the directed edges are the values of the fluxes. The steady-state fluxes between the states 1, 2, 3 and 4 can be decomposed into cycle fluxes with positive weights. Two different decompositions are indicated and cycles are labeled by greek letters.

The essence of this approach is best viewed in the ensemble picture. Consider a large number of identical physical systems with a finite number of states. Each system entering a certain state i stays there for an average time $\langle\tau_i\rangle$, and then proceeds to another state j according to a fixed transition rate. Up to normalization the flux may be seen as the number of systems proceeding from one state to another per unit time. In fig. 1 we present an elementary four-state example. A cycle representation of the fluxes means to write them as a linear superposition of cycle fluxes with a non-negative weight assigned to each cycle. Below we show that such representations exist for every NESS. To follow the line of arguments it is helpful to consider a socio-physical analogy: the cycles may be interpreted as the lines of a mass transit system with the peculiarity that the lines are running one-way on closed loops. The fluxes are proportional to the total amount of passengers travelling from one station to another; *i.e.,* from a state i to state j of the Markov process. The lines are represented in different colors in fig. 1. We imagine each passenger to carry a (correspondingly colored) ticket indicating the line he is currently using. Passengers can change lines in the stations. To remain in a steady state this involves exchange of tickets between passengers at stations.

The aim of this work is to formally describe the representation of NESS by cyclic fluxes, that represent the number of passengers on the lines, and to explore consequences of this point of view on NESS. Natural questions concern the existence and uniqueness of such a representation. Fig. 1 shows by example that a representation in terms of a linear superposition of cycles is not unique. The question concerning existence will be answered positively in the following.

Markov Processes Revisited. – We start by briefly reviewing Markov processes on a finite state space [3,4,6]. We represent the process as a random walk on a graph $G = (V, E)$ with $N = |V|$ vertices v_i, $i \in \{1, \dots, N\}$ and directed edges $(i, j) \in E$. The vertices represent the states of the non-equilibrium system, and are shown as grey circles displaying the vertex indices $1, \dots, 4$ in fig. 1. A system entering vertex v_i will jump to another vertex v_j with probability a^i_j after having stayed in state i for an exponentially distributed waiting time τ_i. Consequently, the transition rates per unit time are $w^i_j := a^i_j / \langle \tau_i \rangle$. A system trajectory is the realization of a random walk of one of the passengers through the transit system. In terms of the transition matrix

$$W^i_j := \begin{cases} w^i_j & i \neq j \\ -\sum_{k \neq i} w^i_k = -\langle \tau_i \rangle^{-1} & i = j \end{cases} \tag{1}$$

or for the fluxes from i to $j \neq i$

$$\phi^i_j = p_i w^i_j \tag{2}$$

the equation for the evolution of the probability $p_i(t)$ to find the system in a state i at time t becomes the compact form

$$\frac{\mathrm{d}p_i}{\mathrm{d}t} = \sum_j W^j_i p_j = \sum_{j \neq i} \left(\phi^i_j - \phi^j_i \right). \tag{3}$$

Here and in the following we suppress the explicit time-dependence and write, e.g., p_i instead of $p_i(t)$. The first equality in eq. (3) stresses the linearity of the problem and is useful for algebraic considerations. The second emphasizes the physical concept of a master or continuity equation: in a steady state the net influx must equal the net outflux, $\sum_{j \neq i} {\phi^i_j}^* = \sum_{j \neq i} {\phi^j_i}^*$. In terms of the currents, $I^i_j := \phi^i_j - \phi^j_i$, this node condition,

$$0 \stackrel{!}{=} \sum_{j \neq i} \left({\phi^i_j}^* - {\phi^j_i}^* \right) = \sum_{j \neq i} {I^i_j}^*, \tag{4}$$

amounts to Kirchhoff's current law which expresses particle (or probability) conservation at each vertex [10]. Here and in the following the * marks steady-state quantities.

Due to the continuity equation (3) every normalized initial distribution remains normalized at all times, and it relaxes to a steady state p^*_i [3].

Algebraically the steady-state probability distribution p^*_i is a left eigenvector of W with eigenvalue zero. Ergodicity ascertains the existence of a path $i_0 \dots i_n$ with a positive $\omega_{i_0, \dots, i_n} := \prod_{j=1}^n w^{i_{j-1}}_{i_j}$ for every pair of vertices i_0 and i_n. This ensures existence and uniqueness of the normalized distribution obeying

$$\sum_i p^*_i = 1. \tag{5}$$

In the physics literature a steady state is called an equilibrium if it obeys *detailed balance*, *i.e.,* if the individual fluxes between any two vertices i and j cancel

$${I^i_j}^* = {\phi^i_j}^* - {\phi^j_i}^* = 0. \tag{6}$$

A necessary yet not sufficient condition for detailed balance is that ${\phi^i_j}^* \neq 0$ implies ${\phi^j_i}^* \neq 0$. As we will argue below, this is a symmetry property obeyed by all physical systems. For an equilibrium system the ratio of $\omega_{i_0, \dots, i_n}$ and the one for the reverse path $\omega_{i_n, \dots, i_0}$ only depends on the initial and final point irrespective of the chosen path [3,4]. Examining the above relation for paths starting from a fixed reference vertex j one obtains an explicit representation of the steady-state probability density

$$p^*_i = p^*_j \; \frac{\omega_{j, \dots, i}}{\omega_{i, \dots, j}} =: p^*_j \; \exp\left(-U^{(j)}_i \right). \tag{7}$$

Hence one can always write $U^{(j)}_i = U_i + c_j$, where U_i is a universal function and c_j depends on the chosen reference site. Consequently,

$$p^*_i = Z^{-1} \exp(-U_i) \tag{8a}$$

where the partition function

$$Z = \sum_k \exp(-U_k) \tag{8b}$$

secures normalization eq. (5).

Cycle Representation and Transform. – The cycle transform is based on the idea that fluxes in a steady state may be represented as superpositions of cycle fluxes (cf. fig. 1). A cycle α of length s_α is an equivalence class of ordered sets of s_α vertices which form a self-avoiding closed path, where paths differing only by a cyclic permutation of vertices are identified. We quantify the number of systems traversing each edge of α by the weight m^*_α. There can be several cycles traversing an edge (i,j), the flux ϕ^i_j quantifies the total number of states traversing that edge per unit time. In the remainder of this section we work out how the steady-state fluxes can be represented by different cycles α with positive weights m^*_α assigned to each of them.

To express the geometrical structure of the cycles we define the indicator functions $\chi^i_{j,\alpha}$ and $\chi_{i,\alpha}$ as

$$\chi^i_{j,\alpha} = \begin{cases} 1 & \text{if } \alpha \text{ passes through the directed edge } (i,j) \\ 0 & \text{otherwise} \end{cases} \tag{9a}$$

$$\chi_{i,\alpha} = \begin{cases} 1 & \text{if } \alpha \text{ passes through vertex } i \\ 0 & \text{otherwise} \end{cases} \tag{9b}$$

In the language of graph theory $\chi^i_{j,\alpha}$ is the adjacency matrix of a cycle. The following identities hold:

$$\sum_j \chi^i_{j,\alpha} = \sum_j \chi^j_{i,\alpha} = \chi_{i,\alpha}, \tag{10}$$

$$\sum_i \chi_{i,\alpha} = s_\alpha, \tag{11}$$

where s_α is the length of the cycle α. With their help we formulate the ideas of the previous paragraph mathematically. As we show below, there is a set of cycles $\{\alpha_k\}$ with non-negative flux densities $m^*_\alpha \geq 0$ such that

$$\phi^{i\,*}_j = \sum_\alpha m^*_\alpha \chi^i_{j,\alpha} \tag{12}$$

for all pairs of vertices (i,j).

To obtain a decomposition we choose an arbitrary enumeration of all M possible cycles $\alpha_1, \alpha_2, \ldots, \alpha_M$ on G. The ambiguity in choosing the order of this enumeration leads to different decompositions constructed by the following algorithm:

Start the iteration for cycle α_1 with a flux field $\phi^{i\,(1)}_j = \phi^{i\,*}_j$ that contains the steady-state fluxes of the original system:

- Initialization:

$$\phi^{i\,(1)}_j := \phi^{i\,*}_j, \text{ for all } i,j. \tag{13}$$

Successively subtract the fluxes along different cycles. In the kth step set $m^*_{\alpha_k}$ to be the minimum of the values of the flux $\phi^{i\,(k)}_j$ along the edges contained in α_k. The new flux field in iteration $k+1$ is the current one with $m^*_{\alpha_k}$ subtracted at the edges traversed by cycle α_k:

- Iteration:

$$m^*_{\alpha_k} := \min_{i,j}\{\phi^{i\,(k)}_j : \chi^i_{j,\alpha_k} > 0\}, \tag{14a}$$

$$\phi^{i\,(k+1)}_j := \phi^{i\,(k)}_j - m^*_{\alpha_k}\chi^i_{j\alpha_k}. \tag{14b}$$

The algorithm terminates after all possible cycles have been considered:

- Termination condition:

$$k = M \tag{15}$$

We claim that at this point all edge fluxes have been assigned to a cycle, and the remaining flux field is zero along all edges,

$$\phi^{i\,(M+1)}_j = 0, \text{ for all } i,j. \tag{16}$$

Existence of a valid decomposition. – To show existence of such a decomposition we demonstrate that for every flux field satisfying the steady-state condition, eq. (4), the algorithm terminates with zero fluxes along all edges, eq. (16), and provides non-negative weights which fulfill the defining equation (12).

The algorithm always terminates in finite time because M is finite.

Since the weight assigned to a cycle, eq. (14a), is the minimum of all $\phi^{i\,(k)}_j$ among the edges of cycle α_k, the new fluxes $\phi^{i\,(k+1)}_j$ assigned by eq. (14b) remain non-negative. Consequently, the steady-state weights $m^*_{\alpha_k}$ are non-negative.

We prove eq. (16) by contradiction. Suppose there is a flux $\phi^{i\,(M+1)}_j \neq 0$. If this flux fulfills the node condition there is a cycle which could have been assigned a larger weight $m^*_{\alpha_k}$, contradicting eq. (14a). Hence, the remaining fluxes obey

$$\sum_j \left(\phi^{i\,(M+1)}_j - \phi^{j\,(M+1)}_i\right) \neq 0. \tag{17}$$

In contrast, for every steady state the intial flux field eq. (13) fulfills the node condition (4). Iterating the initial flux field we find

$$\begin{aligned} 0 &= \sum_j \left(\phi^{i\,(k)}_j - \phi^{j\,(k)}_i\right) \\ &= \sum_j \left(\phi^{i\,(k+1)}_j - \phi^{j\,(k+1)}_i\right) + m^*_{\alpha_k} \sum_j \left(\chi^i_{j,\alpha_k} - \chi^j_{i,\alpha_k}\right) \\ &= \sum_j \left(\phi^{i\,(k+1)}_j - \phi^{j\,(k+1)}_i\right) \end{aligned}$$

where we used eq. (10) in the last line. In contradiction to eq. (17) this holds for every $k \leq M$, and we hence proved eq. (16).

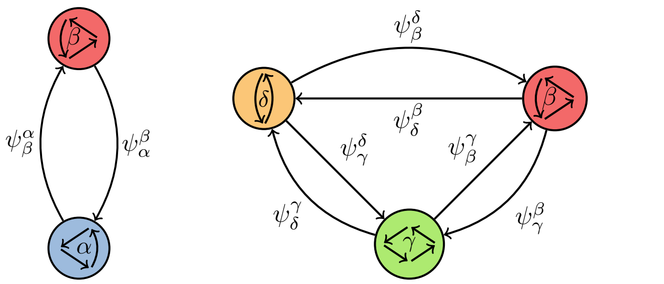

Fig. 2: Transformed graph H obtained for the original graph G for the two decomposition of the flux field introduced in fig. 1.

By construction the cycle-fluxes obtained in this way fulfill eq. (12). We use eq. (14b) and a telescope sum argument to obtain

$$\sum_{k=1}^{M} m^*_{\alpha_k} \chi^i_{j,\alpha_k} = \phi^{i\,(1)}_j - \phi^{i\,(M+1)}_j = \phi^{i\,*}_j$$

where in the last equation we used the algorithm initialization eq. (13) and eq. (16).

Although one cannot specify in advance which cycles are used in the general case, one can (by using the freedom of choice in the enumeration) specify a set of disjoint cycles to be part of the cycles used in the decomposition, *i.e.,* cycles α with non-vanishing weights m^*_α. A possible choice is to include the set of 2-cycles. The result is a splitting of the fluxes in a detailed-balance part (the set represented by the 2-cycles), and the remaining current part. This resembles the approach in [4], but is conceptionally different because the decomposition here does not discard the information stored in the 2-cycles.

Transitions on cycle space. – The set of weights $\{m^*_{\alpha_k}\}$ can be interpreted as a mapping that transforms the original graph $G = (V, E)$ into a new one $H = (C, E_C)$, see fig. 2. For instance, the vertex $\alpha \in C$ represents the cycle α in G with the non-zero weight m^*_α as identified by the algorithm. A directed edge $(\alpha, \beta) \in E_C$ indicates that two cycles share at least one vertex of G, *i.e.,* one state of the original system. Each edge (α, β) of the transformed graph is associated with a transition rate b^α_β. In the analogy of the mass transit system $\psi^\alpha_\beta := m^*_\alpha b^\alpha_\beta$ characterizes the number of passengers changing from line α to line β in the stationary system, see fig. 2.

We shall call the operation $G \to H$ the *cycle transform*. By virtue of eq. (12), the inverse of the cycle transform exists and is unique.

To find the rate constants b^α_β we realize that in the steady state at each vertex v_i (*i.e.,* station, in the sociophysical picture) a constant number of passengers arrives per unit time. This number is proportional to the overall influx $\sum_\gamma \chi_{i,\gamma} m^*_\gamma = \sum_{j\neq i} \phi^{j\,*}_i$. After all, the passengers carry tickets indicating which line they are running on. Upon arrival at the station, the passengers randomly exchange their tickets with other passengers, and board the line for which their new ticket holds. We adopt a random exchange where all passengers arriving at a station put their tickets into an urn and subsquently draw a new one from the urn. The probability for a passenger to continue on line β after arriving at station i amounts to the ratio of the number of tickets for line β to the overall number of tickets.

Thus at station i the probability of continuing with line β is

$$b^{(i)}_\beta = \frac{m^*_\beta}{\sum_\gamma \chi_{i,\gamma} m^*_\gamma}. \tag{18}$$

The total flux ψ^α_β from line α to line β is obtained by summing the local exchange flux $m^*_\alpha b^{(i)}_\beta$ over all mutual stations where $\chi_{\beta,i}\chi_{\alpha,i} = 1$

$$\psi^\alpha_\beta = \sum_i \chi_{\beta,i}\chi_{\alpha,i} m^*_\alpha b^{(i)}_\beta = m^*_\alpha \sum_i \frac{\chi_{\beta,i}\chi_{\alpha,i}}{\sum_\gamma \chi_{i,\gamma} m^*_\gamma} m^*_\beta = \psi^\beta_\alpha. \tag{19}$$

One of the key features of the new formulation is that these cycle-space fluxes fulfill detailed balance ($\psi^\alpha_\beta = \psi^\beta_\alpha$ for all α, β). This is due to the formulation of the exchange process as a microscopically balanced ticket exchange.

Because of detailed balance in H we can proceed along the line indicated by eq. (7). Replacing w^i_j by b^α_β, one obtains a potential $\mathcal{H}_\alpha$, such that the occupation numbers m^*_α are given by Boltzmann weights,

$$m^*_\alpha = \mathcal{Z}^{-1} \exp(-\mathcal{H}_\alpha). \tag{20}$$

Here the partition function

$$\mathcal{Z} = \sum_\alpha \tau_\alpha \exp\left(-\mathcal{H}_\alpha\right), \tag{21}$$

includes the average cycle period

$$\tau_\alpha = \sum_i \chi_{\alpha,i} \langle \tau_i \rangle. \tag{22}$$

After all, the weights m^*_α are no probabilities. According to eqs. (1) and (2) they rather fulfill

$$\sum_\alpha m^*_\alpha \tau_\alpha = \sum_\alpha m^*_\alpha \sum_i \chi_{\alpha,i} \langle \tau_i \rangle = \sum_i \chi^*_i = 1$$

In summary, the potential $\mathcal{H}_\alpha$ is obtained from the rate constants w^i_j, by determining the population density m^*_α of the cycles, followed by eqs. (18), (19) and finally (7).

Physical applications. – Markov processes often possess a symmetry property, sometimes denoted as *dynamical reversibility* [11]: if for any two states i and j the transition $i \to j$ is possible, then the reverse transition $j \to i$ must also be possible, *i.e.,* $w^i_j \neq 0 \Leftrightarrow w^j_i \neq 0$. Still, one can have $w^i_j \ll w^j_i$. This symmetry always holds for physical systems with reversible microscopic laws because for every microscopic "forward" trajectory leading the system from state i to j the time-reversed "backward" trajectory from j to i is also a valid trajectory. In contrast, the example of fig. 1 does not fulfill dynamic reversibility along its outer edges. Yet the transformed graphs (fig. 2) always do.

The connection of Markov process to thermodynamics [3, 5, 12–14] allows us to explore the importance of fluxes in two fundamental ways:

1. in non-equilibrium thermodynamics the central quantities are the non-zero currents I which are driven by affinities A. Though such quantities are usually defined for macroscopic transport, one can consistently define them for stochastic transitions if dynamic reversibility holds:

$$I^i_j := \phi^i_j - \phi^j_i, \tag{23a}$$

$$A^i_j := \log \phi^i_j - \log \phi^j_i. \tag{23b}$$

Observe that $\mathrm{sgn}(I^i_j) = \mathrm{sgn}(A^i_j)$. Consequently, the positive total entropy production can always be expressed [3] as

$$P_{tot} = \frac{1}{2} \sum_{i,j} A^i_j I^i_j; \tag{24}$$

2. also the well-known fluctuation relations for the entropy production along (a set of) individual random trajectories crucially rely on distinguishing transition $i \to j$ from $j \to i$, as they compare probabilities of trajectories and their reverse counter-parts [11, 13–16].

An electric and thermodynamic analogy. – In this section we introduce an analogy relating Markov processes, thermodynamics and electrical circuits. Different electric analogies have been presented in the literature that are suitable for different purposes (see e.g. [4,6]). The appropriate analogies are summarized in table 1. In this analogy the quantities defined above have the properties of their electrical counterparts:
U, I, A and $\mathcal{E}$ are antisymmetric and the resistance R is symmetric and positive.
The definition of the fluxes, eq. (2), obeys Kirchhoff's equation [10],

$$U^i_j + \mathcal{E}^i_j = R^i_j I^i_j, \tag{25}$$

which states that if no current is flowing between two nodes with a battery-like element connecting them, a voltage difference U is created. This voltage is the negative of the electromotance $\mathcal{E}$ of the battery. However, if a current is running over a resistor R, it obeys an Ohmic law and the voltage drops by $R \cdot I$.
Kirchhoff's current law ("node rule") amounts to eq. (4). Kirchhoff's voltage law ("mesh rule") states that integrating the voltage differences around a closed cycle is zero. This also holds in our analogy. It is the basis for the identification of U with a total differential in thermodynamics.

symbol	analogy	thermodynamic	electric
V_i	$-\log p_i$	potential	potential
U^i_j	$\log[p_i/p_j]$	tot. differential	voltage
I^i_j	$\phi^i_j - \phi^j_i$	current	current
A^i_j	$\log[\phi^i_j/\phi^j_i]$	affinity	-
$\mathcal{E}^i_j$	$\log[w^i_j/w^j_i]$	-	electromotance
R^i_j	U^j_i/I^j_i	-	resistance
P_{sys}	$\frac{1}{2}\sum_{i,j} U^i_j I^i_j$	entropy change	power

Table 1: Electrical and thermodynamical analogies for Markov processes.

Finally, the quantity P_{sys} describes the change of the system's Gibbs entropy $S_{sys} := -\sum_i p_i \log p_i$ as the systems undergoes its dynamics,

$$P_{sys} = \frac{\mathrm{d}}{\mathrm{d}t} S_{sys}. \tag{26}$$

It vanishes in the steady state, and can be related to the irreversible entropy production P_{tot} by defining an entropy flux to the medium [3, 4]

$$P_{med} = \frac{1}{2} \sum_{i,j} \left(\phi^i_j - \phi^j_i\right) \log \frac{w^i_j}{w^j_i}. \tag{27}$$

One then finds $P_{tot} = P_{sys} + P_{med}$. Introducing thermodynamic analogues one obtains $P_{med} = \frac{1}{2}\sum_{i,j} I^i_j \mathcal{E}^i_j$ such that $P_{tot} = \frac{1}{2}\sum_{i,j} I^i_j (U^i_j + \mathcal{E}^i_j)$. Hence, the defintions of table 1 are consistent with the definitions made earlier, and $A^i_j = U^i_j + \mathcal{E}^i_j$.

The analogy is not perfect, however. Consider a simple cycle with the same current flowing through all nodes. Then the potential difference between two non-adjacent nodes i and j cannot be obtained from an effective resistance (or electromotance) which is the sum of the individual resistances (or electromotances) of the edges connecting i to j as it would be the case in an electrical network.

Averages on cycle space. – For every well-defined mapping $F : \alpha \mapsto F_\alpha$ from the set of cycles to the real numbers we define the cycle average

$$\langle F \rangle_C := \sum_\alpha m^*_\alpha F_\alpha. \tag{28}$$

For instance, for the geometric matrices $\chi^i_{j,\alpha}$ we have $\langle \chi^i_j \rangle_C = {\phi^i_j}^*$ by the definitions eqs. (12, 28). On the other hand, $\langle 1 \rangle_C \neq 1$, because the edge fluxes are not normalized weights.

Now let us consider cycle-space observables related to physical quantities. Consider some matrix $F \in \mathbb{R}^{N\times N}$. We can interpret this quantity as the change of some physical observable due to the transitions between different states. We define

$$J_F(t) = \sum_{i,j} F^i_j \phi^i_j(t) =: \langle F \rangle_{2,t} \tag{29}$$

as the average flux of quantity F at time t. The last equivalence is the definition of the average as the two-point probability-density function at time t. For antisymmetric F one has $J_F = 1/2 \sum_{i,j} F^i_j I^i_j$.

To connect this with the cycle transform we define an observable

$$F_\alpha = \sum_{i,j} \chi^i_{j,\alpha} F^i_j \tag{30}$$

which is the integrated contribution of F along cycle α. It is straightforward to show that

$$J^*_F = \lim_{t\to\infty} \langle F \rangle_{2,t} = \langle F \rangle_C. \tag{31}$$

For $F^i_j = A^i_j = \log\left(\phi^i_j/\phi^j_i\right)$ we hence generalize the result of [3] for the entropy production in the steady state

$$P^*_{tot} = J^*_A = \langle A \rangle_C = \sum_\alpha m^*_\alpha A_\alpha \tag{32}$$

to cycles that are constructed in a completely different way than in [3]. Here, to obtain the first equality we used the antisymmetry of A^i_j.

Conclusion & Outlook. – In this work we presented a mapping, the cycle transform, that generally applies to steady states of discrete Markov processes. It can be used to transform a non-equilibrium steady state represented by a graph G into an equilibrium steady state on a graph H whose vertices are appropriatly chosen cycles in G. For physical systems, a natural symmetry on G, called dynamical reversibility, allows us to relate our method to thermodynamics. The presented mapping supports the paradigm of focussing on fluxes rather than currents. The non-uniqueness of the decomposition can be used to seperate detailed balance contributions (2-cycles) from non-equilibrium currents (non-trivial cycles). Also, the connection between averages defined on the space of cycles to steady-state averages was made.

In forthcoming work the cycle transform might serve as another perspective on thermodynamical machines where different cycles represent the different operation modes. A well-studied example is the steady-state dynamics of the molecular motor kinesin (see eg. [17]) in the framework of a Markov process. For such small machines thermal fluctuations play a crucial role. The cycle-transform representation of the entropy production, eq. (32), is a novel perspective to this problem, which can also provide a deeper understanding of (steady-state) fluctuation relations [15, 18, 19].

The suggested approach also has interesting parallels to the theory of dynamical systems, especially chaos theory [7–9]. In chaos theory cycles, *i.e.,* unstable periodic orbits of the dynamical system, play a crucial role. They lie dense in phase space such that trajectories can be seen as a realization of a random-walk dynamics between cycles, similar to the dynamics in cycle space considered in the present study. Also expectation values in such systems can be calculated using cycle expansions.

Finally, though this paradigm might be useful to capture non-equilibrium thermodynamics it still relies crucially on the existence of a steady state. For many interesting non-equilibrium systems, like the Belousov-Zhabotinsky reaction in a flow reactor [2,20] this paradigm is not applicable. Instead of having a steady state with constant macroscopic quantities such systems show persistent regular oscillations on a macroscopic scale which prevent the existence of a steady-state probability density. To describe such systems we propose to explore the generalization of the present results to Markov processes with sources and sinks of probability (and thus drop the assumption of irreducibility) in order to account mass flux through the system.

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