



PAPER

Linear stochastic thermodynamics

OPEN ACCESS

RECEIVED
1 June 2022REVISED
11 July 2022ACCEPTED FOR PUBLICATION
22 July 2022PUBLISHED
19 August 2022

Original content from
this work may be used
under the terms of the
[Creative Commons
Attribution 4.0 licence](#).

Any further distribution
of this work must
maintain attribution to
the author(s) and the
title of the work, journal
citation and DOI.

Danilo Forastiere^{1,*} , Riccardo Rao^{2,*} and Massimiliano Esposito^{1,*}¹ Department of Physics and Materials Science, Complex Systems and Statistical Mechanics, University of Luxembourg, L-1511, Luxembourg² Simons Center for Systems Biology, School of Natural Sciences, Institute for Advanced Study, Princeton, NJ 08540, United States of America

* Authors to whom any correspondence should be addressed.

E-mail: danilo.forastiere@uni.lu, riccardorao@ias.edu and massimiliano.esposito@uni.lu**Keywords:** linear response theory, stochastic thermodynamics, nonequilibrium statistical mechanics

Abstract

We study the thermodynamics of open systems weakly driven out-of-equilibrium by nonconservative and time-dependent forces using the linear regime of stochastic thermodynamics. We make use of conservation laws to identify the potential and nonconservative components of the forces. This allows us to formulate a unified near-equilibrium thermodynamics. For nonequilibrium steady states, we obtain an Onsager theory ensuring nonsingular response matrices that is consistent with phenomenological linear irreversible thermodynamics. For time-dependent driving protocols that do not produce nonconservative forces, we identify the equilibrium ensemble from which Green–Kubo relations are recovered. For arbitrary periodic drivings, the averaged entropy production (EP) is expressed as an independent sum over each driving frequency of non-negative contributions. These contributions are bilinear in the nonconservative and conservative forces and involve a novel generalized Onsager matrix that is symmetric. In the most general case of arbitrary time-dependent drivings, we advance a novel decomposition of the EP rate into two non-negative contributions—one solely due to nonconservative forces and the other solely due to deviation from the instantaneous steady-state—directly implying a minimum EP principle close to equilibrium. This setting reveals the geometric structure of near-equilibrium thermodynamics and generalizes previous approaches to cases with nonconservative forces.

1. Introduction

The development of phenomenological irreversible thermodynamics in the first half of the twentieth century primarily relies on the concept of local equilibrium. Since macroscopic bulk systems are considered, equilibrium thermodynamics is assumed to hold within each volume element of the system. Exchange processes of globally conserved quantities (e.g. energy and mass) between nearby volumes cause an irreversible entropy production (EP). This fundamental quantity determines the amount of entropy which is irreversibly dissipated over time, and it can be evaluated by relying on a second assumption: the gradients of intensive quantities (e.g. temperature and chemical potential) are locally small enough to justify linearizing the currents of the conjugated extensive conserved quantities (e.g. energy flow for temperature gradients and particle flow for chemical potential gradients) [1–6]. A third important assumption is that the Onsager matrix resulting from that linearization is symmetric. Justifications of this fact rely on microreversibility and detailed balance, which holds at equilibrium. They follow two main directions. In the first, the internal dynamics of the system is not modeled but linear stochastic equations are assumed to describe the fluxes of extensive quantities [7–12]. The second approach is based on linear response theories, where the internal dynamics of a system initially described by an equilibrium distribution is weakly perturbed from the outside. The dynamical response of the system near equilibrium can then be expressed in terms of equilibrium correlators [13–18].

Stochastic thermodynamics (ST) is a more modern endeavor which formulates nonequilibrium thermodynamics for systems far from equilibrium in contact with reservoirs and described by stochastic dynamics [19–30], and has therefore become the standard setting to model far from equilibrium mesoscopic systems. The scope of ST broadened with time: it started from describing the dynamics of the averages of thermodynamic observables [17, 19, 21, 22] and arrived at a precise identification of their fluctuating analogs. The central assumption of local detailed balance (LDB) relates features of the noise induced by the reservoirs with the entropy change in the reservoir [17, 31–33]. This assumption can be justified microscopically by assuming that the reservoirs are only weakly displaced from equilibrium by the system [34, 35], and it is a key ingredient to study systems driven arbitrarily far from equilibrium. In fact, the LDB property allows to translate the fluctuation theorems [36–39]—which are major breakthroughs in nonequilibrium statistical physics—in the language of ST. These theorems provide a refinement of the second law [24] and enable to extend thermodynamics at the trajectory level [25, 40–42]. By now, many experimental validations of ST are available [43, 44] (and references therein).

To fix the terminology, let us consider a system in contact with two thermostats at temperatures $T_1(t)$ and $T_2(t)$, which may depend on time. For $T_1(t) \neq T_2(t)$, the system is pushed away from equilibrium by the nonconservative thermodynamic force $\mathcal{F}(t) = \frac{1}{T_1(t)} - \frac{1}{T_2(t)}$. If $\mathcal{F}(t)$ changes in time because of a driving protocol, this driving is said to be *nonconservative*. In contrast, if $\mathcal{F}(t) = 0$ at all times, both the system and the driving protocol are said to be *detailed balanced*. It may happen that a system does not feature any nonconservative force, for example if a single thermal reservoir constitutes the whole environment. In this case, the driving is detailed balanced by construction, and we speak of an *unconditionally detailed balance* driving and system. Linear response theory provides a natural tool to explore the close-to-equilibrium regime of ST and to establish connections with phenomenological irreversible thermodynamics. Many such studies have been carried out in the past, but most consider unconditionally detailed balance drivings (arbitrary [45–47] or periodic [48–53]) or nonequilibrium steady states [41, 54]. Few also described systems driven by time-dependent nonconservative forces [53, 55]. However, these descriptions lack a systematic procedure to decompose the driving into its conservative and nonconservative contributions, which is important since these two contributions generate very different kinds of responses. A procedure to achieve this separation based on conservation laws is a quite recent achievement [29, 56]. In this paper, we build on it to formulate a linear response theory of ST for arbitrary drivings that makes contact with classic results of irreversible thermodynamics. We develop our theory using Markov jump processes at the ensemble averaged level. For nonequilibrium steady states, macroscopic theory for steady-state transport is recovered, and a symmetric nonsingular Onsager matrix ensues by construction. For unconditionally detailed-balanced drivings, we show how to recover Green–Kubo relations for the response of state observables. For periodically driven systems, we introduce a novel frequency-resolved generalized Onsager matrix (75) that—in contrast to previous descriptions—is symmetric and independent on the driving protocol. For arbitrary protocols, we show that the EPR decomposes into two positive-definite quadratic forms, equation (118). The first reduces to the total EPR in a NESS and contains the infinite-time response matrix of physical currents, i.e. the Onsager matrix computed at steady state. The second gives the dissipation due to the lag of the dynamics with respect to the instantaneous steady state identified by the forces. The minimum EP principle follows immediately.

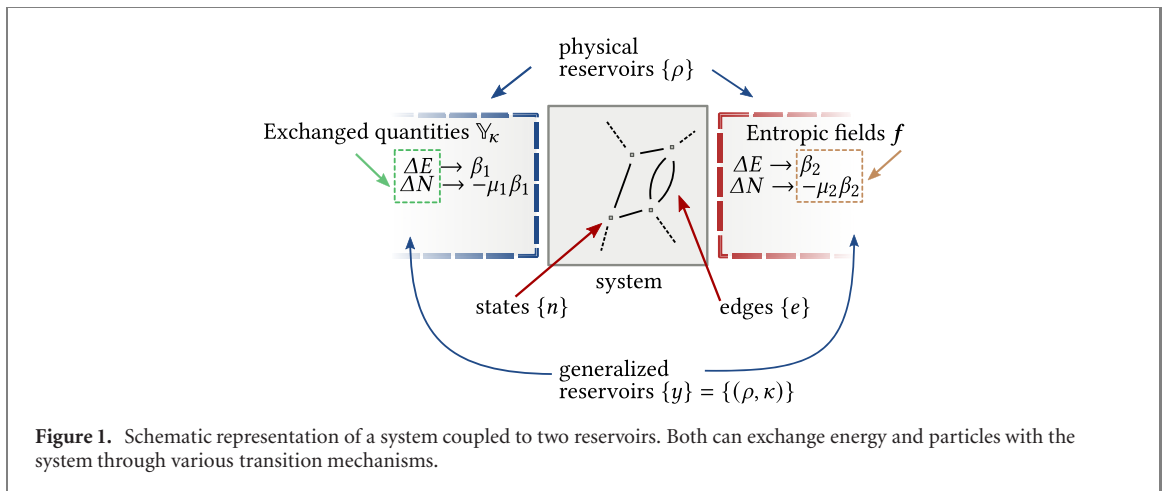
Outline. We start by reviewing ST of Markov jump processes using conservation laws, section 2. In section 3, we then formulate a linear response theory for perturbations only acting on the intensive field that characterize the reservoirs. The generalisation to perturbations that also act on system quantities is presented in section 4. The theory is illustrated on simple systems in section 5 and conclusions are drawn in section 6.

2. Stochastic thermodynamics and conservation laws

In this section we revisit the formulation of ST that makes use of conservation laws to discriminate between conservative and nonconservative driving forces.

2.1. Thermodynamics for Markov jump processes

We consider a system composed of a set $\mathcal{V} = \{n\}$ of mesoscopic states to which one assigns a set $\mathcal{K} = \{\kappa\}$ of different extensive quantities (e.g. energy, particle numbers), denoted $\mathbb{Y}_{\kappa n}$, see figure 1. If the system was isolated, these quantities would be conserved. But the system is in contact with a set $\mathcal{P} = \{\rho\}$ of equilibrium *reservoirs*. Each reservoir ρ is characterized by a number of intensive entropic fields [6] conjugated with the extensive quantities exchanged with the system, e.g. inverse temperature with energy, chemical potential divided by temperature with particle number, etc. These fields are denoted by f_y with $y = (\rho, \kappa)$ and the set of all entropic fields by $\mathcal{Y} = \{(\rho, \kappa)\}$. The reservoirs can trigger a set $\mathcal{E}^+ = \{e\}$ of



(directed) transitions between the mesoscopic states of the system $n \xrightarrow{e} n'$, where n, n' are two different states, and e is the transition mechanism. Mesoscopic states and transition mechanisms define the nodes and edges of a directed graph $(\mathcal{V}, \mathcal{E}^+)$, which allows for multiple edges between pairs of nodes. Micro-reversibility requires that each oriented edge e has a corresponding inverse transition denoted $-e$. Using the notation $o(e)$ to select the origin node of the edge e , the components of the *incidence matrix* that identifies the multigraph are $\mathbb{D}_{ne} = \delta_{no(-e)} - \delta_{no(e)}$. Since the extensive quantities are conserved in the union of system plus reservoirs, their changes along each transition e satisfy a balance equation

$$\mathbb{Y}_{o(-e)\kappa} - \mathbb{Y}_{o(e)\kappa} = \sum_m \mathbb{Y}_{\kappa m} \mathbb{D}_{me} = \sum_{\rho} \mathbb{X}_{(\rho,\kappa)e}, \quad (1)$$

where the matrix $\mathbb{X} = (\mathbb{X}_{ye})$ encodes the amount of extensive quantities exchanged with each such reservoir y along e . Micro-reversibility is formalized as the requirement that $\mathbb{X}_{y,-e} := -\mathbb{X}_{ye}$. In addition to the $|\mathcal{K}|$ *trivial* conservation laws leading to the balance equation (1), additional *non-trivial* conservation laws may arise from constraints in the internal structure of the system.

The dynamics of the system obeys a continuous-time Markov process over the set of mesoscopic states. This implies that the probability vector $\mathbf{p}(t) = (p_n(t))$ describing the probabilities to find the system in each mesoscopic state at time t is the solution of the master equation generated by the $|\mathcal{V}| \times |\mathcal{V}|$ matrix of transition rates $\mathbb{W}(t)$:

$$d_t \mathbf{p}(t) = \mathbb{W}(t) \mathbf{p}(t) = \mathbb{D} \mathbf{J}(t), \quad (2)$$

where the component of the vector of probability currents $\mathbf{J}(t)$ read

$$J_e(t) := w_e(t) p_{o(e)}(t) - w_{-e}(t) p_{o(-e)}(t) = -J_{-e}(t). \quad (3)$$

The explicit time dependence will often be omitted in the rest of the paper.

Dynamically, the balance equations for the extensive quantities (1) give rise to the continuity equations

$$d_t \langle \mathbb{Y}_{\kappa} \rangle := d_t \sum_n \mathbb{Y}_{\kappa n} p_n = \langle d_t \mathbb{Y}_{\kappa} \rangle + \sum_{\rho} \mathbf{I}_{(\rho,\kappa)}, \quad (4)$$

where the last term represents the currents of extensive quantities exchanged with the reservoirs

$$\mathbf{I} = \mathbb{X} \mathbf{J}. \quad (5)$$

The thermodynamic consistency is ensured by the LDB which links the stochastic dynamics and thermodynamics (the Boltzmann constant $k_B = 1$) [17, 31–33]:

$$\ln \frac{w_e}{w_{-e}} = (\mathbb{D}^T \mathbf{S} - \mathbb{X}^T \mathbf{f})_e. \quad (6)$$

Here \mathbf{S} denotes the internal entropy of the mesoscopic states. As a result, the first term in the rhs of (6) denotes the internal entropy change arising in the system along the transition e , while the second one denotes the entropy changes in the reservoirs caused by the exchanges of extensive quantities.

By rewriting the probability currents (3) as

$$J_e = w_e p_{o(e)} (1 - e^{-A_e}), \quad (7)$$

we introduce the edge affinities

$$A_e := \ln \frac{w_e p_{o(e)}}{w_{-e} p_{o(-e)}} \quad (8)$$

which, using the LDB condition (6), take the form

$$\mathbf{A} = -\mathbb{X}^T \mathbf{f} + \mathbb{D}^T (\mathbf{S} - \ln \mathbf{p}). \quad (9)$$

Now and in the following, component-wise operations over vectors will be used: $\mathbf{ab} = (a_i b_i)$, $\ln \mathbf{a} = (\ln a_i)$ and $\mathbf{ab}^T = (a_i b_i)$. The affinities (9) represent the entropy changes in the reservoirs and in the system (internal and self-information) induced by the transition e . The EPR along each transition can therefore be expressed as the flux–force expression

$$\dot{\sigma} = \mathbf{J} \mathbf{A} \geq 0, \quad (10)$$

and the total EPR by

$$\dot{\Sigma}(t) = \sum_e \dot{\sigma}_e(t) = \sum_e J_e(t) A_e(t) = \mathbf{J}^T \mathbf{A} \geq 0. \quad (11)$$

Here and in the rest of the paper the sum runs over the directed edges $e \in \mathcal{E}^+$. Indeed, using (9) and (6), and introducing the average entropy of the system $S_{\text{sys}} := \sum_n (S_n - \ln p_n) p_n$, we find as expected [31] that

$$d_t S_{\text{sys}} = \dot{\Sigma} + \underbrace{\sum_n p_n d_t S_n - \sum_{e,y} f_y \mathbb{X}_{ye} J_e}_{\text{Entropy flow}}. \quad (12)$$

When the system is at equilibrium, the affinities, the currents and the EPR along every edge e vanish, which coincides with the condition of detailed balance

$$A_e = 0 \Leftrightarrow w_e p_{o(e)}^{\text{eq}} = w_{-e} p_{o(-e)}^{\text{eq}} \Leftrightarrow J_e = 0 \Leftrightarrow \dot{\sigma} = 0. \quad (13)$$

Master equations of the type (2) with (6) are widely used, for instance to model chemical and biological systems [57–59] or electric circuits [43, 60, 61].

2.2. Topology and conservation laws

Introducing the incidence matrix \mathbb{D} in equation (2) enables us to leverage the topological properties of the systems. Indeed, Schnakenberg proved that cyclic sets of transitions form bases for the space of the steady-state probability currents [19]. In reference [29] these sets were used to identify all the conservation laws at work in the system, as well as characterizing their flowing through the system. Here, we briefly review these results. The construction of all the quantities that we introduce is illustrated using a minimal model in section 5.2.

The cycle decomposition of the Markov process is obtained by finding the kernel of the incidence matrix \mathbb{D} , i.e. the space of vectors satisfying $\mathbb{D} \mathbf{C}_\alpha = \mathbf{0}$. These vectors are called cycles since they identify sequences of transitions such that one returns back to the initial state after their completion. The kernel $\mathcal{A} \equiv \ker \mathbb{D}$ is spanned by $|\mathcal{A}|$ vectors \mathbf{C}_α which are the column vectors of the matrix $\mathbb{C} = (\mathbf{C}_{e\alpha})$. They are such that $\mathbf{C}_{e\alpha} = 0$ if the cycle α does not contain the edge e and $\mathbf{C}_{e\alpha} = \pm 1$ otherwise. The sign defines the orientation needed to complete the cycle (see the networks in the examples in section 5).

While cycles are enough to obtain the steady-state distribution of a Markov process [19], the extensive quantities exchanged along each cycle are of relevance for thermodynamics. They are encoded in the *physical topology* matrix

$$\mathbb{M} := \mathbb{X} \mathbb{C}. \quad (14)$$

Each entry represents the amount of extensive quantity κ exchanged with physical reservoir ρ after performing a cycle α . The constraints that the conservation laws impose on the cycle affinities are encoded in the left null space \mathbb{L} . This vector space is spanned by $|\mathbb{L}| = |\text{coker } \mathbb{M}|$ conservation law vectors $\boldsymbol{\ell}_\lambda$ which are the columns vectors of the matrix $\mathbb{I} := (\mathbb{I}_{y\lambda})$ that satisfies

$$\mathbb{I}^T \mathbb{M} = \mathbf{0}. \quad (15)$$

For each conservation law $\boldsymbol{\ell}_\lambda$ we can identify a corresponding conserved quantity L_λ , column vector of the matrix $\mathbb{L} = (\mathbb{L}_{n\lambda})$, that satisfies the balance equation

$$\mathbb{D}^T \mathbf{L}_\lambda = \mathbb{X}^T \boldsymbol{\ell}_\lambda, \quad (16)$$

or equivalently in matrix form

$$\mathbb{D}^T \mathbb{L} = \mathbb{X}^T \mathbb{I}. \quad (17)$$

This result is obtained realizing that the definition (15) is equivalent to finding the subspace orthogonal to $\ker \mathbb{D} = \text{span}\{\mathbf{C}_\alpha\}$ because of (14). This means that the vectors $\ell_\lambda^\top \mathbb{X}$ are in the cokernel of the incidence matrix \mathbb{D} , i.e. $\ell_\lambda^\top \mathbb{X} \in \text{coker } \mathbb{D} = (\ker \mathbb{D})^\perp$. It follows that, since $(\ker \mathbb{D})^\perp$ is isomorphic to the image of \mathbb{D}^\top (the coimage of \mathbb{D} , see e.g. [62]), there exist $|\Lambda|$ basis vectors \mathbf{L}_λ that are mapped into the transposed of $\ell_\lambda^\top \mathbb{X}$ by \mathbb{D}^\top . Physically, equations (16) and (17) are balance equations. The lhs quantify the changes of the conserved quantities in the system when transitions occur. The rhs identifies the contributions to these variations due to the different reservoirs. Notice that each \mathbf{L}_λ is defined up to an additive constant, as only their differences enter the balance equation. Importantly, these conserved quantities \mathbb{L} encompass the $|\mathcal{K}|$ *trivial* conservation laws, but also contain $|\Lambda| - |\mathcal{K}| \geq 0$ *non-trivial* additional ones, which are system specific (see examples 5.2 and 5.3).

In the rest of the article, we will assume that the topology is left invariant by the driving protocol, so that the conservation laws and conserved quantities can be determined by solving equations (15) and (17) once. If this is not the case, all results still apply, with the caveat that (15) will now have a solution $\mathbb{l}(t)$ that is piece-wise constant in time, and all the objects that derive from it will inherit this property.

2.3. Fundamental forces

The balance equation (16) can be used to split the vector of entropic fields \mathbf{f} into one block of potential fields \mathbf{f}_p and one of fundamental nonconservative fields \mathbf{f}_f , $\mathbf{f} = (\mathbf{f}_f, \mathbf{f}_p)^\top$. Physically, this corresponds to selecting a minimal subset \mathbf{f}_p of entropic fields that defines a reference equilibrium for the system, and the subset of remaining fields \mathbf{f}_f , whose presence may independently prevent the system from reaching such equilibrium. The potential fields \mathbf{f}_p are obtained by selecting the maximal invertible submatrix of \mathbb{l} , denoted \mathbb{l}_p . This matrix is always square, has dimension $|\Lambda|$, and is full rank, since from the definition equation (15) it follows that $\text{rk } \mathbb{l} = |\Lambda|$. We call \mathbb{l}_f the remaining rectangular block of the matrix \mathbb{l} . Equation (17) can be solved for the exchanged quantities corresponding to \mathbf{f}_p , denoted \mathbb{X}_p , in terms of \mathbb{l}_p , \mathbb{l}_f , and the remaining $|\mathcal{Y}| - |\Lambda|$ exchanged quantities, \mathbb{X}_f , with $\mathbb{X} = (\mathbb{X}_f, \mathbb{X}_p)$:

$$\mathbb{X}_p^\top = -\mathbb{X}_f^\top \mathbb{l}_f \mathbb{l}_p^{-1} + \mathbb{D}^\top \mathbb{L} \mathbb{l}_p^{-1}. \quad (18)$$

This procedure leads to the following rewriting of the exchange contribution appearing in the LDB (6)

$$\mathbb{X}^\top \mathbf{f} = -\mathbb{X}_f^\top \mathcal{F} + \mathbb{D}^\top \mathbb{L} (\mathbb{l}_p^{-1}) \mathbf{f}_p. \quad (19)$$

Equation (19) features the *nonconservative forces* defined by

$$\mathcal{F} := -\mathbf{f}_f + \mathbb{l}_f \mathbb{l}_p^{-1} \mathbf{f}_p, \quad (20)$$

which we refer to as *fundamental* because they are a minimal set of independent mechanisms that break detailed balance, i.e. prevent the system from reaching equilibrium. For example, in a system with two reservoirs exchanging energy, the vector \mathcal{F} contains the difference between the inverse temperatures of the reservoirs (see also example 5.2).

Note that, because of the rank-nullity theorem for the $|\mathcal{Y}| \times |\mathcal{A}|$ physical topology matrix \mathbb{M} , we have that

$$|\text{Fund. forces}| = |\mathcal{Y}| - |\Lambda| = \text{rk } \mathbb{M} = |\mathcal{A}| - |\ker \mathbb{M}|, \quad (21)$$

i.e. the number of fundamental forces is given by the rank of \mathbb{M} . When $\text{rk } \mathbb{M} = 0$: $|\mathcal{Y}| = |\Lambda|$; there are not enough reservoirs for nonconservative forces to be generated; and the system is unconditionally detailed balance. This further emphasizes how \mathbb{M} encapsulates the thermodynamic properties of the system.

Finally, we make explicit that equation (20) defines a linear transformation from the entropic intensive fields \mathbf{f} to the fundamental forces and potential fields:

$$\mathbb{T} \mathbf{f} = \begin{pmatrix} \mathcal{F} \\ \mathbf{f}_p \end{pmatrix}, \quad (22)$$

with its block form being

$$\mathbb{T} = \begin{pmatrix} -\mathbb{1} & \mathbb{l}_f \mathbb{l}_p^{-1} \\ \mathbf{0} & \mathbb{1} \end{pmatrix}. \quad (23)$$

Notice that the transformation matrix \mathbb{T} is involutory, i.e. $\mathbb{T}^2 = \mathbb{1}$.

2.4. EPR and thermodynamic potentials

The interplay between topological and thermodynamic properties of the system also shapes the expression of the EPR. Indeed, using equation (19), the edge affinities (9) and the EP (10) can be rewritten as

$$\mathbf{A} = \mathbb{X}_f^T \mathcal{F} + \mathbb{D}^T \Phi, \quad (24)$$

and

$$\dot{\sigma} = \mathbf{J}\mathbf{A} = \mathbf{J}(\mathbb{X}_f^T \mathcal{F} + \mathbb{D}^T \Phi), \quad (25)$$

respectively, where the stochastic Massieu potential

$$\Phi := \phi - \ln \mathbf{p} \quad (26)$$

is obtained by complementing the Massieu potential of the states

$$\phi := \mathcal{S} - \mathbb{L}(\mathbb{I}_p^{-1})\mathbf{f}_p \quad (27)$$

with the self-information $-\ln \mathbf{p}$. The first contribution in equations (24) and (25) describes the dissipative contribution of the nonconservative forces, whereas the second that of the potential ones.

Using equation (25), the full EPR (11) can be thus rewritten as

$$\dot{\Sigma} = d_t \Phi + \mathcal{F}^T \mathbf{I}_f + \dot{\nu}, \quad (28)$$

where

$$\Phi := \Phi^T \mathbf{p} \quad (29)$$

is a *nonequilibrium Massieu potential* of the system,

$$\mathbf{I}_f := \mathbb{X}_f \mathbf{J} \quad (30)$$

denotes the current of system quantities conjugated to the fundamental forces, and the residual term

$$\dot{\nu} := - (d_t \phi)^T \mathbf{p} \quad (31)$$

denotes the average of the variation in time of the Massieu potential of the states. Equation (28) discriminates the three core mechanisms that contribute to dissipation. The first term describes the dissipation associated to transient relaxation effects, and it vanishes at steady state. The second term describes the dissipation due to nonconservative flows of system quantities through the system, and it is the only nonvanishing term for a NESS. Finally, the third term in equation (28) is a *driving contribution* which describes the dissipation due to external time-dependent drivings.

In detailed-balanced systems—i.e. vanishing forces, $\mathcal{F} = 0$ —with no time-dependent driving, the EPR is fully characterized by the changes of nonequilibrium Massieu potential, Φ , equation (29). Under these conditions, this potential becomes a Lyapunov function which keeps increasing as the system relaxes to equilibrium. To review this fact, let us first note that the state

$$\mathbf{p}^{\text{eq}} = (p_n^{\text{eq}}) = (\exp\{\phi_n - \Phi_{\text{eq}}\}), \quad (32)$$

with $\Phi_{\text{eq}} = \ln \sum_n \exp\{\phi_n\}$ is the equilibrium state of the system, since it satisfies the detailed balance property (13) (consider equation (24) with $\mathcal{F} = 0$). Using this last expression, we obtain

$$\Phi = (\phi - \ln \mathbf{p})^T \mathbf{p} = \Phi_{\text{eq}} - D_{\text{KL}}(\mathbf{p} \parallel \mathbf{p}^{\text{eq}}), \quad (33)$$

where we introduced the Kullback–Leibler divergence $D_{\text{KL}}(\mathbf{p} \parallel \mathbf{p}^{\text{eq}}) = \mathbf{p}^T \ln \left(\frac{\mathbf{p}}{\mathbf{p}^{\text{eq}}} \right) \geq 0$, which is non-negative and vanishes solely at equilibrium. Using the fact that Φ_{eq} is constant, $d_t \Phi_{\text{eq}} = -\dot{\Sigma} \leq 0$, we recover the role of Φ as a Lyapunov function.

We conclude this section by writing the continuity equation constraining the evolution in time of the average of each conserved quantity,

$$d_t \langle L_\lambda \rangle = (d_t L_\lambda)^T \mathbf{p} + L_\lambda^T d_t \mathbf{p} = (d_t L_\lambda)^T \mathbf{p} + \ell_\lambda^T \mathbf{I}, \quad (34)$$

which is obtained using the master equation (2) together with equations (5) and (16). This equation clarifies how the changes of $\langle L_\lambda \rangle$ follows from either intrinsic variations of L_λ due to time-dependent driving (the first term on the rhs) or exchanges with the reservoirs (the second term on the rhs). Equation (34) generalizes equation (4) to the case of non-trivial, system-dependent conserved quantities.

3. Linear regime with protocols acting on the reservoirs

In this section we consider protocols that only act on the entropic fields characterizing the state of the reservoirs, \mathbf{f} , and do not affect the matrix \mathbb{X} of physical quantities exchanged with the reservoirs. We first obtain the linearized solution of the master equation (2) in terms of the fundamental forces and Massieu potentials, section 3.1. In section 3.2 we consider detailed-balanced dynamics to show how Green–Kubo relations and thermodynamic stability conditions are recovered from our approach. In section 3.3 we consider generic periodic drivings and show that the EPR in steady conditions can be written as the modulus of the protocol amplitudes with respect to a suitable scalar product. The matrix representing this scalar product is used to obtain a generalized Onsager matrix in section 3.4. This is the main result of this section. Finally, in sections 3.5 and 3.6 we analyze the response of the currents and the balance equations under this type of protocols.

3.1. Dynamical response of generic systems

To characterize the dynamical response of systems close to equilibrium, we first establish how the currents become linear in the edge affinities. Let the fields \mathbf{f}^{eq} identify some reference detailed balanced conditions described by transition rates \mathbf{w}^{eq} and whose equilibrium probability distribution is \mathbf{p}^{eq} . Upon an instantaneous and small displacement of these fields, $\mathbf{f}(t) = \mathbf{f}^{\text{eq}} + \delta\mathbf{f}(t)$, the transition rates are displaced as $\mathbf{w}(t) \approx \mathbf{w}^{\text{eq}} + \delta\mathbf{w}(t)$, and in turn the probability responds as $\mathbf{p}(t) \approx \mathbf{p}^{\text{eq}} + \delta\mathbf{p}(t)$. In $\delta\mathbf{p}$ and $\delta\mathbf{w}$, only the contributions linear in $\delta\mathbf{f}$ are retained. No probability current flows at equilibrium. Denoting by $\mathbf{j}^{\text{eq}} := \mathbf{w}^{\text{eq}}\mathbf{p}^{\text{eq}}$ the equilibrium fluxes, the first order contribution of the current (7) can be written as

$$\mathbf{J} \approx \delta\mathbf{J} = \mathbf{j}^{\text{eq}}\delta\mathbf{A}, \quad (35)$$

where the linearized affinities are obtained from equation (9), exploiting the identity for the linear corrections $\delta \ln \mathbf{p} = \delta\mathbf{p}/\mathbf{p}^{\text{eq}}$:

$$\mathbf{A} \approx \delta\mathbf{A} = -\mathbb{X}^T\delta\mathbf{f} - \mathbb{D}^T\delta \ln \mathbf{p}. \quad (36)$$

Equivalently, using equation (8) the affinities can be expressed in terms of the response of the rates

$$\delta\mathbf{A} = \left(\frac{\delta w_e}{w_e^{\text{eq}}} - \frac{\delta w_{-e}}{w_{-e}^{\text{eq}}} - \sum_n \mathbb{D}_{e,n} \frac{\delta p_n}{p_n^{\text{eq}}} \right), \quad (37)$$

which in turn respond as

$$\delta w_e = \sum_y \frac{\partial w_e}{\partial f_y} \delta f_y = \sum_y \left(\frac{\partial \ln s_e}{\partial f_y} - \frac{1}{2} \mathbb{X}_{ey} \right) w_e^{\text{eq}} \delta f_y. \quad (38)$$

For this last expression, we used the LDB (6) to write the rates in the form

$$w_e = s_e \exp \left\{ \frac{1}{2} (-\mathbb{X}^T \mathbf{f} + \mathbb{D}^T \mathbf{S})_e \right\}, \quad (39)$$

with $s_e := \sqrt{w_e w_{-e}}$ being a symmetric prefactor invariant under the exchange of e with $-e$. Without loss of generality, we only consider protocols such that $\delta\mathbf{S} = \sum_y \frac{\partial \mathbf{S}}{\partial f_y} \delta f_y = 0$, as the additional term could be treated as a perturbation in exchanged quantities \mathbb{X} , considered in section 4.

Equations (35)–(37) establish the linear response of currents and affinities. Note however that the response (36) is written in terms of two physically distinct contributions: the first one is due to the variation of the external intensive fields, $\delta\mathbf{f}$, while the second is a dynamic response and only depends on the current state of the system, which is represented by the instantaneous relative deviation from the equilibrium distribution, $\delta \ln \mathbf{p} = \delta\mathbf{p}/\mathbf{p}^{\text{eq}}$.

Before we proceed with our derivation of the linearized master equation, we introduce the Hermitian scalar product $\langle \cdot, \cdot \rangle$ over the space of edge vectors defined (in matrix notation and by components resp.) by

$$\langle \mathbf{a}, \mathbf{b} \rangle := \mathbf{a}^\dagger (\mathbf{j}^{\text{eq}} \mathbf{b}) = \sum_e j_e^{\text{eq}} a_e^* b_e, \quad (40)$$

which corresponds to an equilibrium average (with equilibrium fluxes as weights over the edges instead of probabilities). With a slight abuse of notation, we will use the same symbol for the matrix obtained from the weighted contraction of two matrices over edges defined by

$$\langle \mathbb{M}, \mathbb{N} \rangle_{ij} := \sum_e j_e^{\text{eq}} \mathbb{M}_{i,e}^* \mathbb{N}_{e,j}. \quad (41)$$

It will be clear from the context if the result of the operation is a scalar or a matrix. Finally note that the above definitions imply $\langle \mathbb{M}\mathbf{a}, \mathbb{N}\mathbf{b} \rangle = (\mathbb{M}\mathbf{a})^\dagger (\mathbb{J}^{\text{eq}} \mathbb{N}\mathbf{b}) = \mathbf{a}^\dagger \langle \mathbb{M}, \mathbb{N} \rangle \mathbf{b}$. This definition bears connections to the one introduced in reference [63].

The linearized version of master equation (2), can thus be written as

$$d_t \delta \mathbf{p} = \mathbb{D} \delta \mathbf{J} = \langle \mathbb{D}^\top, \delta \mathbf{A} \rangle \quad (42)$$

$$= -\langle \mathbb{D}^\top, \mathbb{X}^\top \rangle \delta \mathcal{F} - \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \delta \ln \mathbf{p}, \quad (43)$$

where we used equations (35) and (36). In addition, using equation (24), we can establish the alternative formulation in terms of the nonconservative forces and the Massieu potential,

$$d_t \delta \mathbf{p} = \langle \mathbb{D}^\top, \mathbb{X}_f^\top \rangle \delta \mathcal{F} + \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle (\delta \phi - \delta \ln \mathbf{p}), \quad (44)$$

which follows from rewriting the linear contribution to the affinities (36) as

$$\delta \mathbf{A} = \mathbb{X}_f^\top \delta \mathcal{F} + \mathbb{D}^\top (\delta \phi - \delta \ln \mathbf{p}), \quad (45)$$

with

$$\delta \mathcal{F} = -\delta f_f + \mathbb{L}_f \mathbb{L}_p^{-1} \delta f_p \quad (46)$$

$$\delta \phi = -\mathbb{L}(\mathbb{L}_p^{-1}) \delta f_p. \quad (47)$$

We remark that the perturbations acting on the symmetric part of the rates s_e do not contribute to $\delta \mathbf{p}$ near equilibrium. In other words, a perturbation on the symmetric part of the rates does not influence the linear response of the system [64, 65].

The solution of the linearized master equation (43) for a general perturbation over the intensive fields \mathbf{f} provides an ensemble description for near-equilibrium mesoscopic systems [64, 66] even when detailed balance is violated. This description is readily obtained in Fourier transform, which we recall here for convenience

$$\hat{g}(\omega) := \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau e^{-i\omega\tau} \check{g}(\tau), \quad (48)$$

(the corresponding inverse transform being $\check{g}(t) = \int_{-\infty}^{\infty} d\omega e^{i\omega t} \hat{g}(\omega)$). Indeed, by solving for the Fourier transform of the instantaneous deviations $\delta \ln \hat{\mathbf{p}}(\omega)$ in terms of the perturbation $\delta \hat{\mathbf{f}}(\omega)$, we obtain

$$\delta \ln \hat{\mathbf{p}}(\omega) = \frac{\delta \hat{\mathbf{p}}(\omega)}{\mathbf{p}^{\text{eq}}} = -\mathbb{A}(\omega) \langle \mathbb{D}^\top, \mathbb{X}^\top \rangle \delta \hat{\mathbf{f}}(\omega), \quad (49)$$

where

$$\mathbb{A}(\omega) := (i\omega \mathbb{P} + \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle)^{-1}, \quad (50)$$

and $\mathbb{P}_{nm} := \delta_{nm} p_m^{\text{eq}}$. The auxiliary matrix $\mathbb{A}(\omega)$ encodes how the probability vector responds to different frequencies. We can thus characterize the solution in time domain

$$\mathbf{p}(t) \approx \mathbf{p}^{\text{eq}} \left(1 - \int_{-\infty}^{\infty} d\omega e^{i\omega t} \mathbb{A}(\omega) \langle \mathbb{D}^\top, \mathbb{X}^\top \rangle \delta \hat{\mathbf{f}}(\omega) \right) \quad (51)$$

$$= \mathbf{p}^{\text{eq}} \left(1 + \int_{-\infty}^{\infty} d\omega e^{i\omega t} \mathbb{A}(\omega) \left(\langle \mathbb{D}^\top, \mathbb{X}_f^\top \rangle \delta \mathcal{F}(\omega) + \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \delta \hat{\phi}(\omega) \right) \right), \quad (52)$$

where we also used equation (24). This near-equilibrium solution of the master equation characterizes the dynamical response of the system to protocols with arbitrary time-dependence.

3.2. Response of unconditionally detailed-balance systems

For unconditionally detailed-balanced systems, we can obtain simple expressions for both the response of the system quantities and the total dissipation during the relaxation to equilibrium. These results will be later generalized to systems in which detailed balance is broken by nonconservative forces.

3.2.1. Linear response theory for system quantities

Let us consider an unconditionally detailed-balanced system that is initially prepared in an equilibrium steady state, $\mathbf{p}^{\text{eq}'}$, defined according to equation (32). Without loss of generality, we can write $\phi_n = \phi_n^{\text{eq}'} = S_n - L_{n\lambda} f'_\lambda$, as a one-to-one mapping can be constructed between \mathcal{Y} and Λ in absence of nonconservative forces—in other words, we regard \mathbb{L}_p as similar to an identity matrix. A small perturbation

of the intensive fields, $\mathbf{f} = \mathbf{f}' + \delta\mathbf{f}$, causes a change $\delta\mathbf{p}(t)$ in the probability vector. Since $\mathcal{F} = \mathbf{0}$, by solving equation (44) we get

$$\delta\mathbf{p}(t) = \int_0^t ds \exp\left\{- (t-s) \frac{\langle \mathbb{D}^\top, \mathbb{D}^\top \rangle}{\mathbb{P}}\right\} \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \delta\phi(s). \quad (53)$$

The matrix $\langle \mathbb{D}^\top, \mathbb{D}^\top \rangle$ enjoys the following important property

$$\begin{aligned} \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \mathbb{P}^{-1} &= \left(\sum_e \frac{j_e^{\text{eq}}}{p_m^{\text{eq}}} \mathbb{D}_{ne} \mathbb{D}_{me} \right) = \left(\sum_e w_{-e}^{\text{eq}} \mathbb{D}_{ne} \delta_{mo(e)} - \sum_e w_e^{\text{eq}} \mathbb{D}_{ne} \delta_{mo(-e)} \right) \\ &= \left(\sum_e \frac{j_e^{\text{eq}}}{p_n^{\text{eq}}} \mathbb{D}_{me} \mathbb{D}_{ne} \right) = (\langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \mathbb{P}^{-1})^\top, \end{aligned} \quad (54)$$

which follows from the definition of the incidence matrix \mathbb{D} and from the detailed-balance condition $j_e^{\text{eq}} = j_{-e}^{\text{eq}}$. The above property also justifies the notation used in the matrix exponential in (53) to stress the symmetry of the matrix at the exponent. Equation (53) shows how the instantaneous response of the probability vector is the outcome of the propagation of the changes of the Massieu potential of the states $\delta\phi$. The propagator, in turn, is essentially determined by topological and thermodynamic properties of the system through $\langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \mathbb{P}^{-1}$.

We now turn our attention to the linear response coefficients of the conserved quantities, L_λ , since these provide a fundamental description of the state of the system. After the perturbation, the deviations of the average L_λ from their equilibrium value are given by

$$\delta\langle L_\lambda(t) \rangle = \mathbf{L}_\lambda^\top \delta\mathbf{p} = \int_0^t ds \left\langle \mathbf{L}(t-s) \frac{\langle \mathbb{D}^\top, \mathbb{D}^\top \rangle}{\mathbb{P}} \delta\phi(s) \right\rangle_{\text{eq}}, \quad (55)$$

where we have introduced the evolved observable

$$\mathbf{L}_\lambda(t) := \mathbf{L}_\lambda^\top \exp\left\{-t \frac{\langle \mathbb{D}^\top, \mathbb{D}^\top \rangle}{\mathbb{P}}\right\}, \quad (56)$$

and the equilibrium average must be intended as a component-wise multiplication by \mathbf{p}^{eq} .

Equation (55) is the Agarwal fluctuation–response relation for the conserved quantities in an unconditionally detailed balanced systems [18, 30, 67–70]. Since we are considering perturbations of systems in thermal equilibrium, one can show that the response matrix is proportional to the derivative of the self-correlation matrix. In fact, since these perturbations only act on the intensive fields \mathbf{f} , we can compute explicitly $\delta\phi = -\sum_\lambda \mathbf{L}_\lambda \delta f_\lambda$, and from equation (53) we obtain the Green–Kubo relation

$$\frac{\delta\langle L_\lambda(t) \rangle}{\delta f_{\lambda'}(s)} = \frac{\partial}{\partial t} \langle \mathbf{L}_\lambda(t-s) \mathbf{L}_{\lambda'} \rangle_{\text{eq}}. \quad (57)$$

This formula immediately implies Onsager reciprocity

$$\frac{\delta\langle L_\lambda(t) \rangle}{\delta f_{\lambda'}(s)} = \frac{\delta\langle L_{\lambda'}(t) \rangle}{\delta f_\lambda(s)}, \quad (58)$$

because the exponential matrix appearing in (56) is symmetric.

We have here discussed the linear response regime of conserved quantities for the simplest class of dynamics—viz unconditionally detailed balance—and recovered the celebrated Onsager’s reciprocity relation. However, this proof cannot be used for system in which the nonconservative forces (such as externally imposed gradients) are present: proofs of Onsager reciprocity in nonequilibrium steady states indeed require different methods [19, 40, 41, 54, 71]. In section 3.4 we will give the construction of response functions expressed in terms of fundamental thermodynamic forces (20). Our approach will (i) provide a natural connection with results from macroscopic thermodynamics, and (ii) generalize the proof of Onsager symmetry to time-dependent nonconservative forces.

3.2.2. Relaxation between different equilibrium states and thermodynamic stability

We now specialize our discussion to purely relaxation processes and investigate the corresponding entropy dissipation. In these processes, the system transitions from one equilibrium state to another as the outcome of an instantaneous switch of the entropic fields from \mathbf{f}' to $\mathbf{f} = \mathbf{f}' + \delta\mathbf{f}$ at $t = 0$. Since no nonconservative force is present—i.e. $\mathcal{F} = 0$ —the two contributions to the EP (equation (28)) are the difference of Massieu potential between the initial and final state and the work done by the driving mechanism to produce the

switch. The latter contribution is readily evaluated: $\dot{\nu} = -(\mathbf{d}_t \phi)^T \mathbf{p} = -\delta_D(t) \sum_\lambda \delta f_\lambda \mathbf{L}_\lambda^T \mathbf{p}^{\text{eq}'}$, where $\delta_D(t)$ is a Dirac delta distribution.

Upon integration of the EPR (28) with the help of (33) we obtain

$$\Sigma := \int_0^\infty dt \dot{\Sigma}(t) = \Phi_{\text{eq}} - \Phi_{\text{eq}'} - \sum_\lambda \delta f_\lambda L_{\lambda n} p_n^{\text{eq}'} = D_{\text{KL}}(\mathbf{p}^{\text{eq}'} \parallel \mathbf{p}^{\text{eq}}). \quad (59)$$

The last equality follows from the definition (26), the fact that equilibrium Massieu potentials are constants, and the constraints on the conservation of probability, $\sum_n \delta p_n = 0$:

$$\begin{aligned} \Phi_{\text{eq}} &= (\phi - \ln \mathbf{p}^{\text{eq}})^T (\mathbf{p}^{\text{eq}'} + \delta \mathbf{p}) = \left(\phi' - \sum_\lambda \delta f_\lambda \mathbf{L}_\lambda^T - \ln \mathbf{p}^{\text{eq}} \right)^T \mathbf{p}^{\text{eq}'} \\ &= \Phi_{\text{eq}'} + \left(-\sum_\lambda \delta f_\lambda \mathbf{L}_\lambda^T - \ln \frac{\mathbf{p}^{\text{eq}}}{\mathbf{p}^{\text{eq}'}} \right)^T \mathbf{p}^{\text{eq}'} = \Phi_{\text{eq}'} - \sum_\lambda \delta f_\lambda \mathbf{L}_\lambda^T \mathbf{p}^{\text{eq}'} + D_{\text{KL}}(\mathbf{p}^{\text{eq}'} \parallel \mathbf{p}^{\text{eq}}). \end{aligned} \quad (60)$$

By solving the integral in equation (53) with $\delta \phi(s) = \theta(s) \sum_\lambda \delta f_\lambda \mathbf{L}_\lambda^T$ one obtains the total probability deviation

$$\Delta p_n := \lim_{t \rightarrow \infty} \delta p_n(t) = \sum_\lambda \delta f_\lambda \left(\langle L_\lambda \rangle_{\text{eq}} - L_{\lambda n} \right) p_n^{\text{eq}}. \quad (61)$$

Using this expression, and the conservation of probability, we can rewrite the total dissipation (59) at the leading order as

$$\Sigma \approx \frac{1}{2} \left\langle \left(\frac{\Delta \mathbf{p}}{\mathbf{p}^{\text{eq}}} \right)^2 \right\rangle_{\text{eq}} = \frac{1}{2} \sum_{\lambda, \lambda'} \delta f_\lambda \delta f_{\lambda'} \left(\langle L_\lambda L_{\lambda'} \rangle - \langle L_\lambda \rangle \langle L_{\lambda'} \rangle \right). \quad (62)$$

By recognizing the last term on the right-hand side as the covariance matrix of the conserved extensive quantities L_λ , we immediately see that the matrix

$$\frac{\partial^2 \Sigma}{\partial f_\lambda \partial f_{\lambda'}} = -\frac{\partial^2 \langle \Phi \rangle_{\text{eq}}}{\partial f_\lambda \partial f_{\lambda'}} = \frac{1}{2} \left(\langle L_\lambda L_{\lambda'} \rangle - \langle L_\lambda \rangle \langle L_{\lambda'} \rangle \right) \quad (63)$$

is positive semi-definite. The first equality holds because of (59). In the thermodynamic limit, this relations entail the so-called *thermodynamic stability conditions*. These constraints indeed follow from the fact that the principal minors of $\partial_\lambda \partial_{\lambda'} \langle \Phi \rangle_{\text{eq}}$ —which corresponds to quantities such as heat capacity and compressibility—inherit the negative semi-definiteness.

In the following section, we develop the above treatment in the more general setting in which nonconservative forces and arbitrary driving protocols are present.

3.3. EPR of periodic steady states

We here aim at expressing the EPR as bilinear form in terms of the protocol's Fourier amplitudes, since this provides a useful geometric characterization of the dissipation due to periodic protocols.

We start our derivation by linearizing the edge currents appearing in the EPR (11) in terms of the edge affinities (9),

$$\dot{\Sigma}(t) \approx \langle \delta \mathbf{A}(t), \delta \mathbf{A}(t) \rangle. \quad (64)$$

This expression is valid for weak perturbations with arbitrary time-dependence, and will serve as a basis for the result presented in section 4.4.2. Focusing here on the frequency domain and applying the Fourier transform (48) we obtain

$$\dot{\Sigma}(t) \approx \int d\omega d\omega' e^{i(\omega+\omega')t} \left\langle \delta \hat{\mathbf{A}}(\omega)^*, \delta \hat{\mathbf{A}}(\omega') \right\rangle. \quad (65)$$

We seek to express the EPR (65) in terms of the variations of the entropic fields $\delta \mathbf{f}$. We first combine equations (36) and (51) to express the response of the edge affinities as $\delta \hat{\mathbf{A}}(\omega) = \mathbb{R}(\omega) \delta \hat{\mathbf{f}}(\omega)$, with the response matrix given by

$$\mathbb{R}(\omega) := -\mathbb{X}^T + \mathbb{D}^T \mathbb{A}(\omega) \langle \mathbb{D}^T, \mathbb{X}^T \rangle. \quad (66)$$

Consider now the specific case of a periodic driving protocol of period T containing multiple commensurate frequencies $\omega_k = \frac{2\pi k}{T}$. The system will relax to a periodic steady state, as equation (51) shows for a protocol of the form $\delta \mathbf{f}(t) = \sum_k e^{i\omega_k t} \hat{\mathbf{f}}(\omega_k)$. This periodic state is reached after the longest relaxation

time of the dynamics. The edge affinities become

$$\delta\hat{\mathbf{A}}(\omega) = \sum_{k=-\infty}^{\infty} \delta\hat{\mathbf{A}}(\omega_k) \delta_{\mathbb{D}}(\omega - \omega_k) = \sum_{k=-\infty}^{\infty} \mathbb{R}(\omega_k) \delta\hat{\mathbf{f}}(\omega_k) \delta_{\mathbb{D}}(\omega - \omega_k), \quad (67)$$

and we can use this expression to characterize the lowest order contribution to the EPR (65) averaged over one period

$$\bar{\Sigma} := \frac{1}{T} \int_0^T dt \sum_{j,k} e^{i(\omega_j + \omega_k)t} \langle \delta\hat{\mathbf{A}}(\omega_j)^*, \delta\hat{\mathbf{A}}(\omega_k) \rangle \quad (68)$$

$$= \sum_{k=-\infty}^{\infty} \bar{\Sigma}(\omega_k), \quad (69)$$

with

$$\bar{\Sigma}(\omega_k) := \langle \delta\hat{\mathbf{A}}(\omega_k), \delta\hat{\mathbf{A}}(\omega_k) \rangle \quad (70)$$

$$= \delta\hat{\mathbf{f}}(\omega_k)^\dagger \langle \mathbb{R}(\omega_k), \mathbb{R}(\omega_k) \rangle \delta\hat{\mathbf{f}}(\omega_k). \quad (71)$$

Equation (69) is a first result obtained from our approach: the EP per period in the periodic steady state is written in terms of a scalar product over the space of the Fourier components of the perturbation. Different frequencies contribute to the total EPR independently from each other, as a consequence of the linearization procedure. This is instrumental for the next result of the paper, viz the construction of a generalized Onsager matrix. Notice that the expression (69) is such that the average EPR of each Fourier mode is non-negative, as it is the modulus of a vector, namely $\bar{\Sigma}(\omega_k) \geq 0$. It follows that $\bar{\Sigma} \geq 0$.

Another implication of equation (69) is related to time-reversal invariance. Since the matrix product (41) is Hermitian, we have that the EP corresponding to a forward protocol, $\mathbf{f}(t)$, coincides with that of the time reversal protocol, $\delta\mathbf{f}_{\text{TR}}(t) = \delta\mathbf{f}(-t)$. Indeed, from the equality $\delta\mathbf{f}_{\text{TR}}(\omega_k) = \int dt e^{i\omega_k t} \delta\mathbf{f}(-t) = \delta\mathbf{f}(\omega_k)^*$, we find that

$$\bar{\Sigma}_{\text{TR}}(\omega_k) = \langle \delta\hat{\mathbf{A}}^*(\omega_k), \delta\hat{\mathbf{A}}^*(\omega_k) \rangle = \bar{\Sigma}(\omega_k). \quad (72)$$

We remark that, because of the presence of equilibrium fluxes in the Hermitian product (40), the average EP of a mode depends on the complete knowledge of the equilibrium rates of the reference equilibrium state.

3.4. Response of the EPR: a generalized Onsager matrix

We now build on equation (69) and the use of the fundamental thermodynamic forces introduced in section 2.2 to construct a generalized Onsager matrix.

We preliminary recall the decomposition of the intensive fields introduced in equation (22),

$$\mathbb{T} \delta\hat{\mathbf{f}}(\omega_k) = \left(\delta\hat{\mathcal{F}}(\omega_k), \delta\hat{\mathbf{f}}_{\text{p}}(\omega_k) \right), \quad (73)$$

which is here expressed in the frequency domain. The first subvector accounts for the nonconservative forces $\delta\hat{\mathcal{F}}(\omega_k) = -\delta\hat{\mathbf{f}}_{\text{f}}(\omega_k) + \mathbb{I}_{\text{f}}^{-1} \delta\hat{\mathbf{f}}_{\text{p}}(\omega_k)$, while the second for the perturbation to the potential fields, which are left invariant by \mathbb{T} . The latter term is responsible for changes of the reference equilibrium. Inserting the identity matrix written as $\mathbb{1} = \mathbb{T}^2$ to the left and to the right of the quadratic form in equation (69), we can rewrite the EPR of a mode as

$$\bar{\Sigma}(\omega_k) := \begin{pmatrix} \delta\hat{\mathcal{F}}(\omega_k) \\ \delta\hat{\mathbf{f}}_{\text{p}}(\omega_k) \end{pmatrix}^\dagger \begin{pmatrix} \mathbb{O}_{\text{ff}}(\omega_k) & \mathbb{O}_{\text{fp}}(\omega_k) \\ \mathbb{O}_{\text{pf}}(\omega_k) & \mathbb{O}_{\text{pp}}(\omega_k) \end{pmatrix} \begin{pmatrix} \delta\hat{\mathcal{F}}(\omega_k) \\ \delta\hat{\mathbf{f}}_{\text{p}}(\omega_k) \end{pmatrix}. \quad (74)$$

The matrix

$$\mathbb{O}(\omega_k) = \mathbb{T}^\top \langle \mathbb{R}(\omega_k), \mathbb{R}(\omega_k) \rangle \mathbb{T} \quad (75)$$

appearing in equation (74) is the *generalized Onsager matrix* of the system. Indeed, it characterizes how the EPR responds to the variations of the fundamental forces $\hat{\mathcal{F}}(\omega_k)$ and the fields $\hat{\mathbf{f}}_{\text{p}}(\omega_k)$ at frequency ω_k . The explicit blocks of the generalized Onsager matrix (75) are expressed in terms of the matrices \mathbb{R}_{f} and \mathbb{R}_{p} defined by

$$\mathbb{R}_{\text{f}}(\omega_k) := -\mathbb{X}_{\text{f}}^\top + \mathbb{D}^\top \mathbb{A}(\omega_k) \langle \mathbb{D}^\top, \mathbb{X}_{\text{f}}^\top \rangle, \quad (76)$$

$$\mathbb{R}_p(\omega_k) := -\mathbb{X}_p^T + \mathbb{D}^T \mathbb{A}(\omega_k) \langle \mathbb{D}^T, \mathbb{X}_p^T \rangle, \quad (77)$$

where the indices p and f selects the rows of \mathbb{X} consistently with the identification of nonconservative forces and potential fields in equation (23). The generalized Onsager matrix in block form reads

$$\begin{aligned} \mathbb{O}(\omega_k) &= \begin{pmatrix} -\mathbb{1} & \mathbb{0} \\ (\mathbb{I}_p^{-1})^T \mathbb{I}_f^T & \mathbb{1} \end{pmatrix} \begin{pmatrix} \langle \mathbb{R}_f, \mathbb{R}_f \rangle & \langle \mathbb{R}_f, \mathbb{R}_p \rangle \\ \langle \mathbb{R}_p, \mathbb{R}_f \rangle & \langle \mathbb{R}_p, \mathbb{R}_p \rangle \end{pmatrix} \begin{pmatrix} -\mathbb{1} & \mathbb{I}_p^{-1} \\ \mathbb{0} & \mathbb{1} \end{pmatrix} \\ &= \begin{pmatrix} \langle \mathbb{R}_f, \mathbb{R}_f \rangle & -(\langle \mathbb{R}_f, \mathbb{R}_f \rangle \mathbb{I}_f \mathbb{I}_p^{-1} + \langle \mathbb{R}_f, \mathbb{R}_p \rangle) \\ -(\langle \mathbb{R}_f, \mathbb{R}_f \rangle \mathbb{I}_f \mathbb{I}_p^{-1} + \langle \mathbb{R}_f, \mathbb{R}_p \rangle)^T & (\mathbb{I}_p^{-1})^T \mathbb{I}_f^T \langle \mathbb{R}_f, \mathbb{R}_f \rangle \mathbb{I}_f \mathbb{I}_p^{-1} + (\mathbb{I}_p^{-1})^T \mathbb{I}_f^T \langle \mathbb{R}_f, \mathbb{R}_p \rangle + \langle \mathbb{R}_p, \mathbb{R}_f \rangle \mathbb{I}_f \mathbb{I}_p^{-1} + \langle \mathbb{R}_p, \mathbb{R}_p \rangle \end{pmatrix}. \end{aligned} \quad (78)$$

Equation (74) is a central result of this work. It is the bilinear form that gives the EPR for each frequency mode of the protocol. Crucially, equation (74) discriminates the perturbations of different nature. The blocks corresponding to $\hat{\mathcal{F}}(\omega_k)$ describe the response to perturbation affecting the nonconservative forces. In contrast, the blocks corresponding $\hat{f}_p(\omega_k)$ describe how the EPR responds to variations of the reference equilibrium. The generalized Onsager matrix (75) is symmetric for each frequency as it results from applying the matrix \mathbb{T} and its transpose to the symmetric matrices $\langle \mathbb{R}(\omega_k), \mathbb{R}(\omega_k) \rangle$.

The first diagonal block at zero frequency $\mathbb{O}_{ff}(0)$, which we call *static*, plays a special role as it gives the total dissipation for steady-state processes. In a later subsection, we will indeed prove that the other three blocks in equation (74) do not contribute to the dissipation in steady state, viz $\mathbb{O}_{fp}(0) = \mathbb{O}_{pf}^T(0)$ and $\mathbb{O}_{pp}(0)$ all vanish.

Physically, finite values of $\delta \hat{f}_p(0)$ must be understood as adiabatic, infinitesimal changes of the reference equilibrium, thus not affecting the EPR in a periodic steady state.

We remark that, in contrast to previous generalizations of the Onsager matrix, our construction guaranties that (i) such a matrix is symmetric, (ii) it solely depends on the system and not on the protocol, and (iii) the time-reversal operation ($\omega \rightarrow -\omega$ in frequency space) leaves the matrix invariant, cf references [53, 55].

3.5. Response of the currents and conserved quantities

We focus now on the physical currents in the periodic steady state, $\mathbf{I} = \sum_k \hat{\mathbf{I}}(\omega_k) e^{i\omega_k t}$, whose variations read

$$\delta \mathbf{I} = \mathbb{X} \delta \mathbf{J} = \langle \mathbb{X}^T, \delta \mathbf{A} \rangle. \quad (79)$$

Using the response function introduced in equation (66), we readily obtain the Fourier components

$$\delta \hat{\mathbf{I}}(\omega_k) = \langle \mathbb{X}^T, \mathbb{R}(\omega_k) \rangle \delta \hat{\mathcal{F}}(\omega_k). \quad (80)$$

The validity of Onsager reciprocal relations is clear: from equation (66), one immediately sees that the Onsager response matrix of the physical currents, $\langle \mathbb{X}^T, \mathbb{R}(\omega_k) \rangle$ in equation (80), is symmetric.

To recover the Onsager reciprocal relation for perturbations described in terms of fundamental forces and potential fields, an appropriate linear transformation of the currents is required. Using the decomposition in terms of the fundamental forces and potential fields, equations (22) and (73), together with equation (80), we obtain

$$\begin{pmatrix} \delta \hat{\mathbf{I}}_f(\omega_k) \\ -(\mathbb{I}_p^{-1})^T \mathbb{I}_f^T \delta \hat{\mathbf{I}}_f - \delta \hat{\mathbf{I}}_p(\omega_k) \end{pmatrix} = -\mathbb{T}^T \delta \hat{\mathbf{I}}(\omega_k) = \mathbb{T}^T \langle -\mathbb{X}^T, \mathbb{R}(\omega_k) \rangle \mathbb{T} \begin{pmatrix} \delta \hat{\mathcal{F}}(\omega_k) \\ \delta \hat{f}_p(\omega_k) \end{pmatrix}. \quad (81)$$

Again, it is readily seen that $\mathbb{T}^T \langle -\mathbb{X}^T, \mathbb{R}(\omega_k) \rangle \mathbb{T}$ is symmetric.

We can now characterize the response of conserved quantities. By rewriting the balance equation (34) in terms of the perturbations in the frequency domain, and using equation (81), we obtain

$$i\omega_k \delta \langle \widehat{L}_\lambda(\omega_k) \rangle = \ell_\lambda^T \langle \mathbb{X}^T, \mathbb{R}(\omega_k) \rangle \mathbb{T} \begin{pmatrix} \delta \hat{\mathcal{F}}(\omega_k) \\ \delta \hat{f}_p(\omega_k) \end{pmatrix}. \quad (82)$$

This relation generalizes the fluctuation–response relation (55) to nonconservative dynamics ($\delta \mathcal{F} \neq 0$). Equation (82) will be further generalized in the next section, where we will also account for the possibility that the observables itself depends on time, cf equation (108).

3.6. Nonequilibrium steady states and relation between response matrices

We now show that the linear regime thermodynamics of nonequilibrium steady states is fully characterized by a single system-dependent matrix (the ordinary Onsager matrix), in terms of which we obtain the response of both the EPR and the physical currents. In this regime, obtained when $\omega_k = 0$, for all k , the right-hand side of equation (82) must vanish for any perturbation $\delta\hat{f}$. This implies that

$$\ell_\lambda^\top \langle \mathbb{X}^\top, \mathbb{R}(0) \rangle = \mathbf{0}. \quad (83)$$

A fact anticipated in section 3.4, we now prove that $\mathbb{O}_{\text{pf}}(0)$, $\mathbb{O}_{\text{fp}}(0)$, and $\mathbb{O}_{\text{pp}}(0)$ all vanish, as well as the corresponding blocks of the response matrix of the currents, equation (81). To this end, using equations (66) and (50), we obtain the identity

$$\langle \mathbb{R}(0), \mathbb{R}(0) \rangle = \langle -\mathbb{X}^\top, \mathbb{R}(0) \rangle, \quad (84)$$

and then we rewrite equation (83) in matrix form by separating the blocks corresponding to forces and potential fields,

$$\mathbf{0} = \mathbb{I}_f^\top \langle \mathbb{R}_f(0), \mathbb{R}(0) \rangle + \mathbb{I}_p^\top \langle \mathbb{R}_p(0), \mathbb{R}(0) \rangle. \quad (85)$$

By multiplying both sides by the inverse of \mathbb{I}_p^\top , we obtain

$$(\mathbb{I}_p^{-1})^\top \mathbb{I}_f^\top \langle \mathbb{R}_f(0), \mathbb{R}(0) \rangle = -\langle \mathbb{R}_p(0), \mathbb{R}(0) \rangle. \quad (86)$$

Finally, using some algebra and this last equality in equation (78) (with $\omega = 0$) allows us to see that all blocks but $\mathbb{O}_{\text{ff}}(0)$ vanish.

We now turn to the response matrix of the currents appearing in equation (81). First, we apply the identity (84) to the generalized Onsager matrix (75), yielding

$$\mathbb{T}^\top \langle -\mathbb{X}^\top, \mathbb{R}(0) \rangle \mathbb{T} = \mathbb{O}(0), \quad (87)$$

i.e. that the generalized Onsager matrix coincides—at steady state—with the response matrix of the currents, equation (81). Therefore, this latter matrix has only one nonvanishing block coinciding with $\mathbb{O}_{\text{ff}}(0)$. The equivalence of these matrices is valid solely at steady state,

$$\delta\hat{\mathcal{I}}_f(0) = \langle -\mathbb{X}_f^\top, \mathbb{R}_f(0) \rangle \delta\hat{\mathcal{F}}(0) = \mathbb{O}_{\text{ff}}(0) \delta\hat{\mathcal{F}}(0), \quad (88)$$

in agreement with classical treatments of the linear regime [7]. A physical consequence of the above reasoning is that no steady current can be sustained by just shifting the equilibrium state by $\delta\hat{f}_p(0)$. This also clarifies that the response matrix of the currents, equation (81), does not coincide with the generalized Onsager matrix (75) at arbitrary frequency.

We conclude this section with a simple yet important physical implication of the linear response regime of periodically driven systems: no current can flow against the average thermodynamic force. Indeed, the direction of the currents is determined by the average thermodynamic force applied since equation (88) can be rewritten as

$$\bar{\mathcal{I}}_f \approx \delta\hat{\mathcal{I}}_f(0) = \mathbb{O}_{\text{ff}}(0) \delta\hat{\mathcal{F}}(0) = \mathbb{O}_{\text{ff}}(0) \overline{\delta\mathcal{F}}. \quad (89)$$

In other words, for perturbations to which this theory applies, it is not possible to establish a current against a nonconservative force $\delta\mathcal{F}$ by acting on the potential fields f_p . The components $\bar{\mathcal{I}}_p$ can be obtained as linear combinations of the fundamental currents $\bar{\mathcal{I}}_f$, as can be seen from equation (81).

3.7. Linear response theory for tightly-coupled systems

An important case in applications is the one of *tightly-coupled* systems [72], e.g. when transport of energy can only be achieved by moving particles from one reservoir to another, as in the example 5.2.

Mathematically, this is expressed by the existence of a conservation law $\ell_{\text{t.c.}}$ that satisfies

$$\ell_{\text{t.c.}}^\top \mathbb{X} = \mathbf{0}, \quad (90)$$

that is a stronger condition compared to equation (15). It implies a constraint for the instantaneous currents in the reservoirs, as it is shown by multiplying the definition on the right by the probability flux \mathbf{j}

$$\ell_{\text{t.c.}}^\top \mathbf{I} = \mathbf{0}. \quad (91)$$

In the simple case of a system with two entropic fields and exchanged quantities, the above condition is equivalent to require the proportionality of the two currents. In general, equation (91) means that the emergence of system-specific conservation laws forces different transport mechanisms to happen

simultaneously. The immediate consequence of (90) is that both the response matrix of the currents (80) and the generalized Onsager matrix (75) have $\ell_{t.c.}$ as left null vector, and will thus be degenerate.

4. General protocol involving reservoirs and system quantities

We now consider the most general class of protocols: those that act not only on the intensive fields of the reservoirs, \mathbf{f} , but also on the extensive quantities of the system, \mathbb{X} . This is motivated, for instance, by experiments in which an electric or magnetic field is used to change the energy levels of the system. We first derive the dynamical as well as topological effects that these protocols have on the system. We then investigate thermodynamic forces and currents, highlighting the differences with the simplified picture presented in section 3. Finally, we establish a general expression of the near equilibrium EPR that is valid at finite times, and examine its consequences for the minimum EP principle and for the adiabatic regime of driving.

4.1. Response of topology and dynamics

The additional perturbation of the system quantities is denoted by $\mathbb{X}(t) = \mathbb{X}^{eq} + \delta\mathbb{X}(t)$, and affects the state-wise value of the conserved quantities $\mathbb{L} \mapsto \mathbb{L} + \delta\mathbb{L}$. In fact, adapting the definition of conservation laws and the balance equation equations (16) and (15), we get the equation to compute the change in the conservation law matrix

$$\mathbb{C}^T(\mathbb{X} + \delta\mathbb{X})^T(\mathbb{I} + \delta\mathbb{I}) = 0, \quad (92)$$

that in turn gives the balance obeyed by the perturbed conserved quantity

$$\delta\mathbb{X}^T \ell_\lambda + \mathbb{X}^T \delta\ell_\lambda = \mathbb{D}^T \delta\mathbf{L}_\lambda. \quad (93)$$

We mention that, as pointed out in section 2.2 and explained in more details in reference [29], only the *non-trivial* (system-specific) conservation law vectors can depend on the protocol, giving $\delta\mathbb{I} \neq 0$, since the *trivial* ones (energy conservation, etc) will always be independent on the instantaneous value of the systems' parameters.

Aware of the subtleties of this general type of perturbation, we now proceed to describe how the linearized dynamics is affected. The expression of the affinities in equation (36) and the corresponding master equation in equation (42), must now be replaced by

$$\mathbf{A} \approx \delta\mathbf{A} = -\delta\mathbb{X}^T \mathbf{f}^{eq} - \mathbb{X}^T \delta\mathbf{f} - \mathbb{D}^T \delta \ln \mathbf{p}, \quad (94)$$

and

$$d_t \delta\mathbf{p} = -\langle \mathbb{D}^T, \delta\mathbb{X}^T \rangle \mathbf{f}^{eq} - \langle \mathbb{D}^T, \mathbb{X}^T \rangle \delta\mathbf{f} - \langle \mathbb{D}^T, \mathbb{D}^T \rangle \delta \ln \mathbf{p}, \quad (95)$$

respectively. In contrast, equations (45) and (44)—which feature the decomposition in forces and potential fields—remain formally correct since $\mathcal{F}^{eq} = \mathbf{0}$. However, the perturbations of the Massieu potential now read

$$\delta\mathcal{F} = -\delta\mathbf{f}_f + \mathbb{I}_f \mathbb{I}_p^{-1} \delta\mathbf{f}_p + \delta(\mathbb{I}_f \mathbb{I}_p^{-1}) \mathbf{f}_p^{eq} \quad (96)$$

$$\delta\phi = -\mathbb{L} (\mathbb{I}_p^{-1}) \delta\mathbf{f}_p - \delta\mathbb{L} (\mathbb{I}_p^{-1}) \mathbf{f}_p^{eq} - \mathbb{L} \delta(\mathbb{I}_p^{-1}) \mathbf{f}_p^{eq}, \quad (97)$$

cf equation (47). Note that the nonconservative forces are affected by perturbations of system quantities solely through the conservation law vector (the last term in equation (96)), see example 5.2.

4.2. Current response

The response of the physical currents changes when the perturbation acts both on \mathbb{X} and \mathbf{f} compared to the situation in section 3.5. It features time-dependent terms containing $\delta\mathbb{X}$ and $\delta\mathbf{f}$. Since at the reference equilibrium $\mathbf{J}^{eq} = \mathbf{0}$ by definition, the first order contribution to the physical currents is

$$\delta\mathbf{I} = \delta(\mathbb{X}\mathbf{J}) = \mathbb{X}\delta\mathbf{J} = -\langle \mathbb{X}^T, \delta(\mathbb{X}^T \mathbf{f}) \rangle + \mathbb{D}^T \delta\mathbf{p} \quad (98)$$

$$= -\langle \mathbb{X}^T, \mathbb{X}^T \rangle \delta\mathbf{f} - \langle \mathbb{X}^T, \delta\mathbb{X}^T \rangle \mathbf{f}^{eq} - \langle \mathbb{X}^T, \mathbb{D}^T \rangle \delta\mathbf{p}. \quad (99)$$

Comparing equation (99) to equation (79) combined with (36), we note that an additional term containing the change $\delta\mathbb{X}$ of exchanged quantities appears. In addition, $\delta\mathbb{X}$ also implicitly determines the solution $\delta\mathbf{p}$ to the linearized master equation, equation (95). As already seen, the situation is conceptually

simpler in the language of fundamental forces and Massieu potentials of the state. Using the decomposition (19), we rewrite equation (99) as

$$\delta \mathbf{I} = \langle \mathbb{X}^T, \mathbb{X}_f^T \rangle \delta \mathcal{F} + \langle \mathbb{X}^T, \mathbb{D}^T \rangle \delta(\phi - \ln \mathbf{p}), \quad (100)$$

where we used $\mathcal{F}^{\text{eq}} = \mathbf{0}$. This expression still contains $\delta \mathbf{p}$, which can here be regarded as an implicit function of $\delta \mathcal{F}$ and $\delta \phi$ through the master equation (44). By explicating the solution for each Fourier mode,

$$\delta \ln \hat{\mathbf{p}}(\omega_k) = \mathbb{A}(\omega_k) \left(\langle \mathbb{D}^T, \mathbb{X}_f^T \rangle \delta \hat{\mathcal{F}}(\omega_k) + \langle \mathbb{D}^T, \mathbb{D}^T \rangle \delta \hat{\phi}(\omega_k) \right), \quad (101)$$

we can rewrite the current as

$$\begin{pmatrix} \hat{\mathbf{I}}_f(\omega_k) \\ \hat{\mathbf{I}}_p(\omega_k) \end{pmatrix} = \begin{pmatrix} \mathbb{Q}_{ff}(\omega_k) & \mathbb{Q}_{fp}(\omega_k) \\ \mathbb{Q}_{pf}(\omega_k) & \mathbb{Q}_{pp}(\omega_k) \end{pmatrix} \begin{pmatrix} \delta \hat{\mathcal{F}}(\omega_k) \\ \delta \hat{\phi}(\omega_k) \end{pmatrix}, \quad (102)$$

where

$$\begin{pmatrix} \mathbb{Q}_{ff}(\omega) \\ \mathbb{Q}_{pf}(\omega) \end{pmatrix} := \langle \mathbb{X}^T, \mathbb{R}_f(\omega) \rangle \quad (103)$$

and

$$\begin{pmatrix} \mathbb{Q}_{fp}(\omega) \\ \mathbb{Q}_{pp}(\omega) \end{pmatrix} := \langle \mathbb{X}^T, \mathbb{D}^T \rangle \{ \mathbb{1} - \mathbb{A}(\omega) \langle \mathbb{D}^T, \mathbb{D}^T \rangle \} \quad (104)$$

$$= i\omega \langle \mathbb{X}^T, \mathbb{D}^T \rangle \mathbb{A}(\omega) \mathbb{P}. \quad (105)$$

We obtained the equality (105) by writing $\mathbb{1} = \mathbb{A}(\omega) \mathbb{A}^{-1}(\omega)$ and using the definition (50).

Since the number of potential fields (the dimension of \mathbf{I}_p) is different from the number of nodes over which the Massieu potential ϕ is defined, the response matrix \mathbb{Q} is not square in general, and Onsager symmetry for the current response coefficients is lost. However, the symmetry is restored at $\omega = 0$, when solely $\mathbb{Q}_{ff}(\omega) = \langle \mathbb{R}_f(0), \mathbb{R}_f(0) \rangle = \mathbb{Q}_{ff}^T(\omega)$ does not vanish. Indeed, $\mathbb{Q}_{fp}(0) = \mathbf{0}$ and $\mathbb{Q}_{pp}(0) = \mathbf{0}$ from the definition (105), while $\mathbb{Q}_{pf}(0) = \mathbf{0}$ is a consequence of equation (83).

We conclude this subsection with a remark concerning the possibility of *pumping*, i.e. inducing a current by changing the Massieu potentials ϕ in absence of net time-averaged forces ($\delta \hat{\mathcal{F}}(0) = \mathbf{0}$). Since $\mathbb{Q}_{fp}(0) = \mathbf{0}$ and $\mathbb{Q}_{pp}(0) = \mathbf{0}$, the zero-frequency response to perturbation of the potentials ϕ reduces to zero. This confirms the intuition gained in the discussion of equation (89): no pumping is possible in the realm of linear ST. This extends the original *no-pumping theorem* [73, 74] to cases in which the nonconservative forces are instantaneously different from zero, complementing previous results on nonequilibrium systems with a specific form of the transition rates [75]. This is consistent with previous observations on driving-induced net average currents in systems governed by time-dependent master equations [59, 76], where the observed effect is nonlinear in the driving amplitude.

4.3. Response of conserved quantities

The analysis of the response of the conserved quantities in section 3.5 is here repeated for the case of general perturbations. This time, one needs to introduce the values of the conserved quantities associated to the reference equilibrium state, that we denote by $\mathbf{L}_\lambda^{\text{eq}}$. The overall change of the average conserved quantities can be then properly defined,

$$\delta \langle L_\lambda(t) \rangle := \mathbf{L}_\lambda^T(t) \mathbf{p}(t) - (\mathbf{L}_\lambda^{\text{eq}})^T \mathbf{p}^{\text{eq}}, \quad (106)$$

and, upon linearizing and Fourier transforming its time derivative, with the help of equation (2), we find

$$i\omega \delta \langle \widehat{L}_\lambda(\omega) \rangle = i\omega \langle \delta \hat{L}_\lambda(\omega) \rangle_{\text{eq}} + (\mathbf{L}_\lambda^{\text{eq}})^T \mathbb{D} \delta \hat{\mathbf{J}}(\omega), \quad (107)$$

where $\langle \delta \hat{L}_\lambda(\omega) \rangle_{\text{eq}} := \delta \hat{\mathbf{L}}_\lambda^T(\omega) \mathbf{p}^{\text{eq}}$. In the periodic steady state, using equations (16), (5), and (102) we find

$$i\omega_k \delta \langle \widehat{L}_\lambda(\omega_k) \rangle = i\omega_k \langle \delta \hat{L}_\lambda(\omega_k) \rangle_{\text{eq}} + \mathbf{e}^T \mathbb{Q}(\omega_k) \begin{pmatrix} \delta \hat{\mathcal{F}}(\omega_k) \\ \delta \hat{\phi}(\omega_k) \end{pmatrix}. \quad (108)$$

This important relation is the general version of the Agarwal–Green–Kubo response formula when the conservation laws are chosen as observables and the perturbation can act both on the reservoirs and on the system quantities.

The generalized version of the Agarwal–Green–Kubo relations (55) are recovered in detailed balanced systems, for which $\delta\mathcal{F} = \mathbf{0}$. Indeed, after expliciting \mathbb{Q} , equation (105), with the help of equation (16), equation (108) becomes

$$\delta\langle\widehat{L_\lambda(\omega_k)}\rangle = \langle\delta\hat{L}_\lambda(\omega)\rangle_{\text{eq}} + \mathbf{L}_\lambda^\top\langle\mathbb{D}^\top, \mathbb{D}^\top\rangle_{\mathbb{A}(\omega_k)}\mathbf{p}^{\text{eq}}\delta\hat{\phi}(\omega_k). \quad (109)$$

By anti-transforming and using equation (56), we recover the expression of the response coefficients in terms of integrals of equilibrium correlators,

$$\delta\langle L_\lambda(t)\rangle = \langle\delta L_\lambda(t)\rangle_{\text{eq}} + \int_{-\infty}^{+\infty} d\tau \left\langle \mathbf{L}_\lambda(t-\tau)^\top \frac{\langle\mathbb{D}^\top, \mathbb{D}^\top\rangle}{\mathbb{P}} \delta\phi(\tau) \right\rangle_{\text{eq}}. \quad (110)$$

The first contribution accounts for the explicit time-dependency of δL_λ on the driving protocol, and was assumed to be zero in the derivation of equation (55).

4.4. Response of the EPR

We complete our analysis by expressing the perturbations of the EPR. Remarkably, two distinct decomposition of the EPR are possible, and they shed light on different aspects of the dynamics near equilibrium. These decompositions constitute the main result of this paper. The first decomposition is presented in section 4.4.1 and holds for periodic protocols in the periodic steady state. It provides a generalization of equations (74) and (78). The second decomposition, introduced in section 4.4.2, holds at finite time for an arbitrary protocol. It tells apart the contribution to the dissipation due to nonconservative forces from that caused by the time-dependent protocol and transient relaxations. This latter decomposition is related to the minimum EP theorem of Prigogine [5], which we recover as a corollary.

4.4.1. Time-averaged EPR decomposition

For systems in the periodic steady state caused by generic protocols, the EPR (70) retains its validity. By expanding the edge affinity vector in terms of the variations of the nonconservative forces $\delta\hat{\mathcal{F}}$ and of the Massieu potentials $\delta\hat{\phi}$, equation (45), we obtain the average EPR per period of a single mode

$$\bar{\Sigma}(\omega_k) = \begin{pmatrix} \delta\hat{\mathcal{F}}(\omega_k) \\ \delta\hat{\phi}(\omega_k) \end{pmatrix}^\top \tilde{\mathbb{O}}(\omega_k) \begin{pmatrix} \delta\hat{\mathcal{F}}(\omega_k) \\ \delta\hat{\phi}(\omega_k) \end{pmatrix} \quad (111)$$

with the Onsager matrix defined as

$$\tilde{\mathbb{O}}(\omega_k) := \begin{pmatrix} \mathbb{O}_{\text{ff}}(\omega_k) & \omega_k^2 \langle \mathbb{X}_f^\top, \mathbb{D} \rangle \mathbb{P} \mathbb{A}(-\omega_k) \mathbb{A}(\omega_k) \mathbb{P} \\ \omega_k^2 \mathbb{P} \mathbb{A}(-\omega_k) \mathbb{A}(\omega_k) \mathbb{P} \langle \mathbb{D}^\top, \mathbb{X}_f^\top \rangle & \omega_k^2 \mathbb{P} \mathbb{A}(-\omega_k) \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle \mathbb{A}(\omega_k) \mathbb{P} \end{pmatrix}. \quad (112)$$

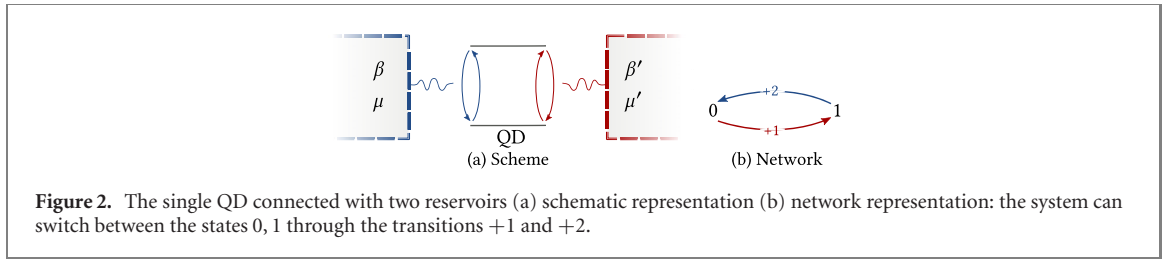
To characterize the latter $(|\mathcal{Y}| - |\Lambda| + |\mathcal{V}|) \times (|\mathcal{Y}| - |\Lambda| + |\mathcal{V}|)$ -matrix, we also used the response term $\delta \ln \hat{\mathbf{p}}(\omega_k)$ in equation (101), as well as the property of the matrix $\mathbb{A}(\omega)$ already used in (105), viz $\mathbb{1} - \mathbb{A}(\omega) \langle \mathbb{D}^\top, \mathbb{D}^\top \rangle = i\omega \mathbb{A} \mathbb{P}$. Equation (111) gives the most general formulation of the time-averaged EPR of each frequency mode in the periodic steady state. It is expressed as a quadratic form of the nonconservative forces and the Massieu potentials. A different representation of the EPR could be obtained choosing the variations $\delta\mathbb{X}$ as control parameters. However, equation (111) is the representation with the minimal number of ‘generalized thermodynamic forces’: the nonconservative forces and variations of the Massieu potentials. In fact, if we subtract the dimension of the vectors featuring (111), namely $|\mathcal{Y}| - |\Lambda| + |\mathcal{V}|$, from the number of independent entries of the matrix \mathbb{X} , $|\mathcal{Y}| + |\mathcal{K}|(|\mathcal{V}| - 1)$, we reduce the dimensionality by $\Delta = |\Lambda| - |\mathcal{K}| + |\mathcal{V}|(|\mathcal{K}| - 1) \geq 0$, with the equality attained by unconditionally detailed balanced systems with a single conserved quantity. We emphasize that such a complexity reduction is obtained thanks to systematic identification and use of the conservation laws of the system.

We finally notice that the generalized Onsager matrix (112) is symmetric at every frequency, and that all blocks except the top-left one, $\mathbb{O}_{\text{ff}}(\omega_k)$ vanish identically at $\omega = 0$. It reduces to equation (78) when the protocol does not affect the system quantities.

4.4.2. Finite-time EPR decomposition

In contrast to the previous characterization of the EPR, the decomposition of the EPR that we describe here is valid at finite times and not only in the periodic steady state. It is expressed in terms of the instantaneous steady-state distribution $\mathbf{p}^{\text{ss}}(t)$, which is obtained from the linearized master equation, equations (43) and (44), as

$$\langle \mathbb{D}^\top, \delta \mathbf{A}^{\text{ss}} \rangle = \langle \mathbb{D}^\top, \mathbb{X}_f^\top \delta \mathcal{F} + \mathbb{D}^\top (\delta\phi - \delta \ln \mathbf{p}^{\text{ss}}) \rangle = \mathbf{0}. \quad (113)$$



Using this relation and the following identity (see the expression of the affinity (45)),

$$\delta\mathbf{A} + \mathbb{D}^T \delta \ln \mathbf{p} = \mathbb{X}_f^T \delta \mathcal{F} + \mathbb{D}^T \delta \phi = \delta \mathbf{A}^{ss} + \mathbb{D}^T \delta \ln \mathbf{p}^{ss}, \quad (114)$$

we can rewrite the EPR (64) as

$$\dot{\Sigma} \approx \langle \delta \mathbf{A}^{ss}, \delta \mathbf{A}^{ss} \rangle + (\delta \ln \mathbf{p} - \delta \ln \mathbf{p}^{ss})^T \langle \mathbb{D}^T, \mathbb{D}^T \rangle (\delta \ln \mathbf{p} - \delta \ln \mathbf{p}^{ss}). \quad (115)$$

The steady-state affinities $\delta \mathbf{A}^{ss}$ appearing in the first term on the right-hand side can be expressed in terms of the sole nonconservative forces $\delta \mathcal{F}$. Indeed, using the solution of equation (113) obtained through the pseudoinverse matrix $\langle \mathbb{D}^T, \mathbb{D}^T \rangle^{-1} = \lim_{\omega \rightarrow 0} \mathbb{A}(\omega)$,

$$\delta \ln \mathbf{p}^{ss} = \langle \mathbb{D}^T, \mathbb{D}^T \rangle^{-1} \langle \mathbb{D}^T, \mathbb{X}^T \rangle \delta \mathcal{F} + \delta \phi, \quad (116)$$

we can rewrite the steady-state affinities (see equation (45)) as

$$\delta \mathbf{A}^{ss} = (\mathbb{X}^T - \mathbb{D}^T \langle \mathbb{D}^T, \mathbb{D}^T \rangle^{-1} \langle \mathbb{D}^T, \mathbb{X}^T \rangle) \delta \mathcal{F}. \quad (117)$$

We complete our derivation by substituting this expression and the definition of generalized Onsager matrix, equation (75), into the EPR (115),

$$\dot{\Sigma} \approx \delta \mathcal{F}^T \mathbb{O}_{ff}(0) \delta \mathcal{F} + (\delta \ln \mathbf{p} - \delta \ln \mathbf{p}^{ss})^T \langle \mathbb{D}^T, \mathbb{D}^T \rangle (\delta \ln \mathbf{p} - \delta \ln \mathbf{p}^{ss}). \quad (118)$$

This decomposition of the dissipation rate into two separately positive quadratic forms is a main result of this paper. The first term describes the dissipation due to the currents generated by the nonconservative forces, and it features the block of the Onsager matrix $\mathbb{O}_{ff}(0)$. It vanishes solely for unconditionally detailed balanced systems or for detailed-balanced driving protocols, when no $\delta \mathcal{F}$ are present or $\delta \mathcal{F}$ vanish at all times, respectively. In contrast, the second term accounts for the dissipation caused by transient effects, i.e. instantaneous deviations from the steady state.

Remark. Equation (118) is reminiscent but not identical to the adiabatic–nonadiabatic decomposition of the EPR [60, 77, 78] (the discrete analogous of the Hatano–Sasa decomposition [79]). Indeed, in this decomposition, the steady-state distribution is introduced to separate steady-state contributions of the EPR (11) from transient ones, see also reference [80]. The decomposition procedure solely acts at the level of the affinities, as reflected in equation (113), and leaves the currents appearing in the EPR unchanged. As a consequence, both terms of the adiabatic–nonadiabatic decomposition depend on $\delta \mathcal{F}$ and $\delta \ln \mathbf{p} - \delta \ln \mathbf{p}^{ss}$. However, being in the linear regime allows us to decompose the currents as well, since they are proportional to the affinities. In this way, one can disentangle completely the contributions involving $\delta \mathcal{F}$ and $\delta \ln \mathbf{p} - \delta \ln \mathbf{p}^{ss}$, and recover equation (118). \square

Remark. The decomposition of the EP in equation (118) can be specialized for a detailed-balanced protocol $\delta \mathcal{F} = \mathbf{0}$. In this case $\delta \ln \mathbf{p}^{ss} = \delta \phi + \delta \Phi$, with $\delta \Phi = \Phi^{eq'} - \Phi^{eq}$ being a constant vector, and hence belonging to the kernel of $\langle \mathbb{D}^T, \mathbb{D}^T \rangle$. If we now substitute this expression for $\delta \ln \mathbf{p}^{ss}$ as well as $\delta \ln \mathbf{p}$ from equation (44) in equation (118), we obtain

$$\dot{\Sigma} \approx \mathbf{d}_t \delta \mathbf{p}^T \langle \mathbb{D}^T, \mathbb{D}^T \rangle^{-1} \mathbf{d}_t \delta \mathbf{p}. \quad (119)$$

This can be interpreted as the EPR of a system relaxing toward an equilibrium state that is changing in time at finite speed. In this sense, it extends the result on the EPR of relaxation obtained in section 3.2.2, that holds only for instantaneous switching protocols, to cases in which the integral (53) cannot be computed explicitly. \square

Remark. An interesting limit is the one of adiabatic driving, that is obtained when $\delta \mathcal{F} = \mathbf{0}$ and the Massieu potential has the form $\phi = \phi(kt)$, with the driving speed parameter k much smaller than the intrinsic timescales of the system. The probability distribution is assumed to follow the driving as $\delta \mathbf{p} \approx \delta \mathbf{p}^{(0)}(kt) + O(k)$. By inserting this adiabatic ansatz in the master equation (44), we find

$\delta \ln \mathbf{p}(kt) = \delta \phi(kt) + \delta \Phi(kt) + O(k)$. Substituting back in (119), we find an adiabatic expansion of the EPR for detailed-balance protocols

$$\dot{\Sigma} \approx k^2 d_t [\mathbf{p}^{\text{eq}}(\delta \phi + \delta \Phi)]^T \langle \mathbb{D}^T, \mathbb{D}^T \rangle^{-1} d_t [\mathbf{p}^{\text{eq}}(\delta \phi + \delta \Phi)]. \quad (120)$$

This allows us to interpret the inverse weighted Laplacian $\langle \mathbb{D}^T, \mathbb{D}^T \rangle^{-1}$ as a metric distance between equilibrium states, i.e. as a *thermodynamic length* [45]. Compared to previous formulations [45, 47], our formulation accounts for variations in both systems quantities and entropic fields. \square

We conclude this section by discussing an important implication of equation (118). Since both terms in equation (118) are separately positive-definite, and since the sole dependence on the instantaneous state comes from the second term, the EPR is minimized by the steady-state distribution, i.e. when $\delta \mathbf{p}(t) = \delta \mathbf{p}^{\text{ss}}$ at all times. This is the *minimum entropy production principle* derived by Prigogine, valid in the phenomenological framework of linear irreversible thermodynamics [5, 81–83]. A related result is the fact that the EPR is a Lyapunov function of the relaxation dynamics. This fact was derived long ago [19, 22, 23] for stochastic dynamics, and in our formalism is readily recovered. Indeed, starting from equation (64) the derivative of the EPR reads

$$d_t \dot{\Sigma} \approx d_t \langle \delta \mathbf{A}, \delta \mathbf{A} \rangle = 2 \langle \delta \mathbf{A}, d_t \delta \mathbf{A} \rangle. \quad (121)$$

Taking the time derivative of (36) in absence of time-dependent driving and using the linearized master equation (42) gives us

$$d_t \delta \mathbf{A} = -\mathbb{D}^T \mathbb{P}^{-1} \langle \mathbb{D}^T, \delta \mathbf{A} \rangle. \quad (122)$$

Substituting this expression back into equation (121), we find

$$d_t \dot{\Sigma} \approx -2 \langle \delta \mathbf{A}, \mathbb{D}^T \rangle \mathbb{P}^{-1} \langle \mathbb{D}^T, \delta \mathbf{A} \rangle \approx -2 \mathcal{I}[\mathbf{p}(t)] \leq 0, \quad (123)$$

where the last equality uses the definition of Fisher information [84] $\mathcal{I}[\mathbf{p}(t)] := \langle (d_t \ln \mathbf{p})^2 \rangle$ computed up to linear orders using equation (42).

5. Examples

In this section we illustrate our theory on some examples that help elucidating the main results.

5.1. Quantum dot in contact with a time-dependent reservoir

The simplest system we consider is a single quantum dot working in the Coulomb blockade regime, where only a single electronic state with energy ϵ is available, figure 4(a). The quantum dot is connected to a lead with inverse temperature β and chemical potential μ . We call $p(t)$ the probability of occupation at time t , namely $p(t) = P(n = 1, t)$, with n being the occupation number of the quantum dot. This probability evolves according to the master equation

$$d_t p(t) = -(w_{-1} + w_{+1})p(t) + w_{+1}, \quad (124)$$

where $w_{\pm 1}$ are the transition rates of the charge and discharge events. The LDB (6) takes the form

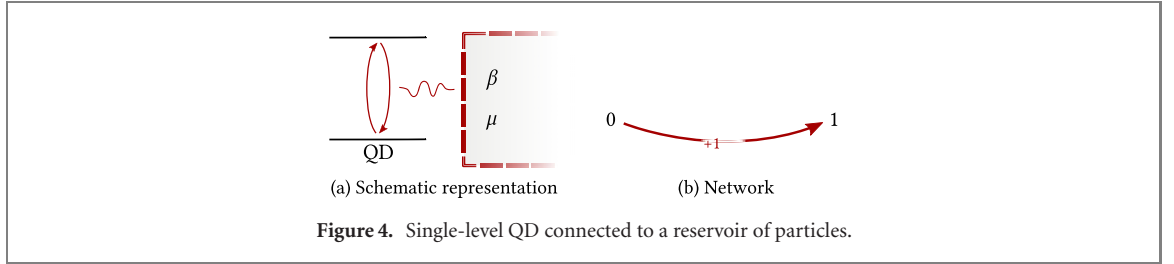
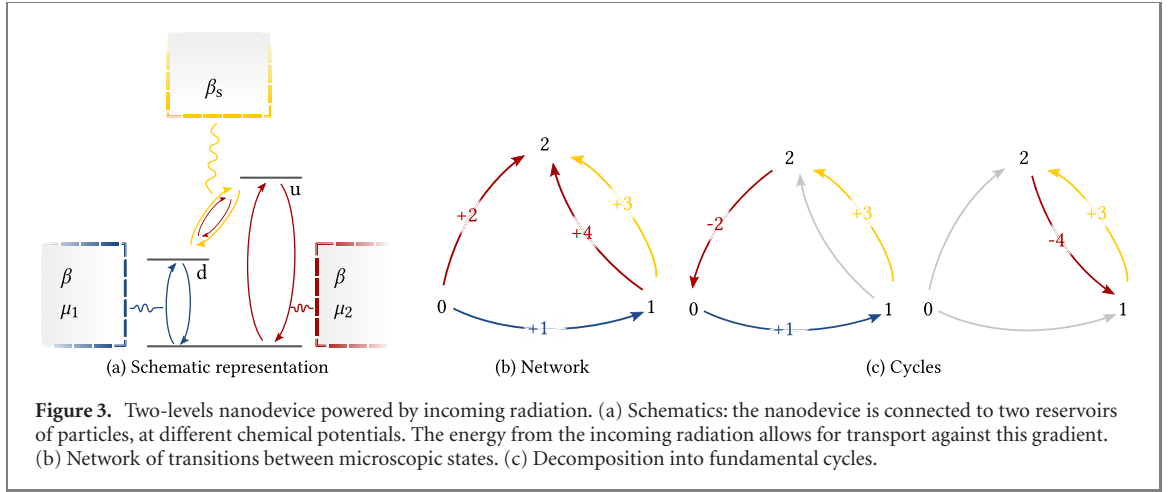
$$\frac{w_{+1}}{w_{-1}} = e^{-\beta(t)(\epsilon - \mu)} \quad (125)$$

with $(\epsilon - \mu)$. The equilibrium value of the occupancy is $p^{\text{eq}} = e^{-\beta(\epsilon - \mu)}(1 + e^{-\beta(\epsilon - \mu)})^{-1}$. The occupation number is $n \in \{0, 1\}$, and the only forward transition is labelled +1. The network representation of the system is shown in figure 4(b). Its incidence matrix reads

$$\mathbb{D} = \begin{matrix} & +1 \\ \begin{matrix} 0 \\ 1 \end{matrix} & \begin{pmatrix} -1 \\ 1 \end{pmatrix} \end{matrix}. \quad (126)$$

This system is unconditionally detailed-balanced because of the absence fundamental nonconservative forces. The extensive quantities corresponding to each of the states of the system are contained in the matrix

$$\mathbb{Y} = \begin{matrix} & E & N \\ \begin{matrix} 0 \\ 1 \end{matrix} & \begin{pmatrix} 0 & 0 \\ \epsilon & 1 \end{pmatrix} \end{matrix}. \quad (127)$$



The matrix of the exchanged quantities can be written as

$$\mathbb{X}^T = +1 \begin{pmatrix} E_1 & N_1 \\ \epsilon & 1 \end{pmatrix} \quad (128)$$

using the fact that during the transition path we are simultaneously exchanging both energy and particles. We see immediately that there is a tight-coupling conservation law $\ell_{t.c.}^T = (1, -\epsilon)$ such that $\ell_{t.c.}^T \mathbb{X} = \mathbf{0}$, since matter and energy are exchanged by the same physical mechanism. The entropic fields that characterize the reservoirs are $\mathbf{f} = (\beta, -\mu\beta)$. The equilibrium flux reduces to a single scalar value

$$j^{eq} = w_{-1}^{eq} p^{eq} = w_{+1}^{eq} (1 - p^{eq}). \quad (129)$$

The response matrices are constructed starting from the auxiliary matrices

$$i\omega \mathbb{P} + \langle \mathbb{D}^T, \mathbb{D}^T \rangle = \begin{pmatrix} i\omega p^{eq} + j^{eq} & -j^{eq} \\ -j^{eq} & i\omega(1 - p^{eq}) + j^{eq} \end{pmatrix} \quad (130)$$

$$\langle \mathbb{D}^T, \mathbb{X}^T \rangle = \begin{pmatrix} -\epsilon j^{eq} & -j^{eq} \\ \epsilon j^{eq} & j^{eq} \end{pmatrix}. \quad (131)$$

The dynamic response in both frequency and time domain is computed plugging the above expressions into equations (49) and (53) and rearranging them as

$$\frac{\partial \hat{p}}{\partial \hat{f}_1}(\omega) = -\frac{\epsilon j^{eq}}{(w_{+1}^{eq} + w_{-1}^{eq} + i\omega)} = \epsilon \frac{\partial \hat{p}}{\partial \hat{f}_2}, \quad (132)$$

$$\frac{\delta p}{\delta f_1}(\tau) = -\epsilon j^{eq} e^{-(w_{+1}^{eq} + w_{-1}^{eq})\tau} \theta(\tau) = \epsilon \frac{\delta p}{\delta f_2}(\tau). \quad (133)$$

We now specialize the analysis to the case of periodic driving on the temperature of the reservoir. The response function (80) of the currents to a generic change of the entropic fields $\delta \hat{\mathbf{f}}$ is

$$\nabla \hat{I}(\Omega) = \frac{i\Omega j^{eq}}{w_{+1}^{eq} + w_{-1}^{eq} + i\Omega} \begin{pmatrix} \epsilon^2 & \epsilon \\ \epsilon & 1 \end{pmatrix}. \quad (134)$$

The generalized Onsager matrix (78) here consists of the \mathbb{O}_{pp} block only,

$$\mathbb{O}(\Omega) = \mathbb{O}_{pp}(\Omega) = \frac{j^{eq} \Omega^2}{(w_{+1}^{eq} + w_{-1}^{eq})^2 + \Omega^2} \begin{pmatrix} \epsilon^2 & \epsilon \\ \epsilon & 1 \end{pmatrix}. \quad (135)$$

Since the system is tightly-coupled, both matrices have zero determinant, as it can be checked directly multiplying on by the conservation law $\ell_{t.c.}$. Notice also that in equations (134) and (135) both the forward and backward equilibrium rates are needed, and this implies the full knowledge of the transition rates, not just their ratio as fixed by LDB, even if we are in presence of a single reservoir. When computing steady-state responses, however, this additional information on the transition rates is no longer needed, as it can be checked directly by imposing $\Omega = 0$.

To make things more concrete, we compute the currents activated by a periodic perturbation on the temperature and the dissipation associated to it. Note also that in terms of β and μ the probability displacement is

$$\delta p = \left(\frac{\delta p}{\delta f_1} - \mu \frac{\delta p}{\delta f_2} \right) \delta \beta - \frac{\delta p}{\delta f_2} \delta \mu \quad (136)$$

$$= \frac{\delta p}{\delta f_2} ((\epsilon - \mu) \delta \beta - \delta \mu), \quad (137)$$

because of equation (132). State observables are characterized by response functions that are directly obtained from the dynamic response, i.e. $\delta \langle E \rangle = \epsilon \delta p = \epsilon \delta \langle N \rangle$. The frequency-dependent response function of the energy to the temperature is then

$$\frac{\partial \langle \hat{E}(\omega) \rangle}{\partial \hat{\beta}} = \epsilon (\epsilon - \mu) \frac{\partial \hat{p}}{\partial f_2}(\omega). \quad (138)$$

Note that its real and imaginary parts satisfy the Kramers–Kronig relations. The Fourier representation of the current component I in the periodic state for the perturbation $\delta \beta(t) = e^{i\Omega t} \delta \beta(0)$ is obtained from equation (80)

$$\delta \hat{I}(\Omega) = \langle \mathbb{X}^\top, \mathbb{R}(\Omega) \rangle \delta \hat{f}(\Omega) \quad (139)$$

and its response to temperature perturbation is obtained from the response to the entropic fields using the relation

$$\delta I = (\epsilon - \mu) \frac{\partial I}{\partial f_2} \delta \beta - \beta \frac{\partial I}{\partial f_2} \delta \mu. \quad (140)$$

We can then introduce the thermal admittance defined as $Y(\Omega) := (\epsilon - \mu) \frac{\delta I}{\delta f_2}$ with real and imaginary part given by

$$\Re Y(\Omega) = (\epsilon - \mu) \frac{\Omega^2 j^{\text{eq}}}{(w_{-1}^{\text{eq}} + w_{+1}^{\text{eq}})^2 + \Omega^2}, \quad (141)$$

$$\Im Y(\Omega) = (\epsilon - \mu) \frac{\Omega j^{\text{eq}} (w_{-1}^{\text{eq}} + w_{+1}^{\text{eq}})}{(w_{-1}^{\text{eq}} + w_{+1}^{\text{eq}})^2 + \Omega^2}. \quad (142)$$

The EP in this case is

$$\bar{\Sigma} \approx (\epsilon - \mu)^2 \mathbb{O}_{2,2}(\Omega) \delta \beta^2. \quad (143)$$

In figure 5 we show how equations (141) and (143) behave when varying the driving frequency.

5.2. Electrical transport through a quantum dot

We now extend the setting of example 5.1 by an additional reservoir of particles at inverse temperature β' and chemical potential μ' . This is the minimal setup to display non-trivial realizations of the quantities introduced in section 2.2. The incidence matrix after the addition of a second reservoir becomes

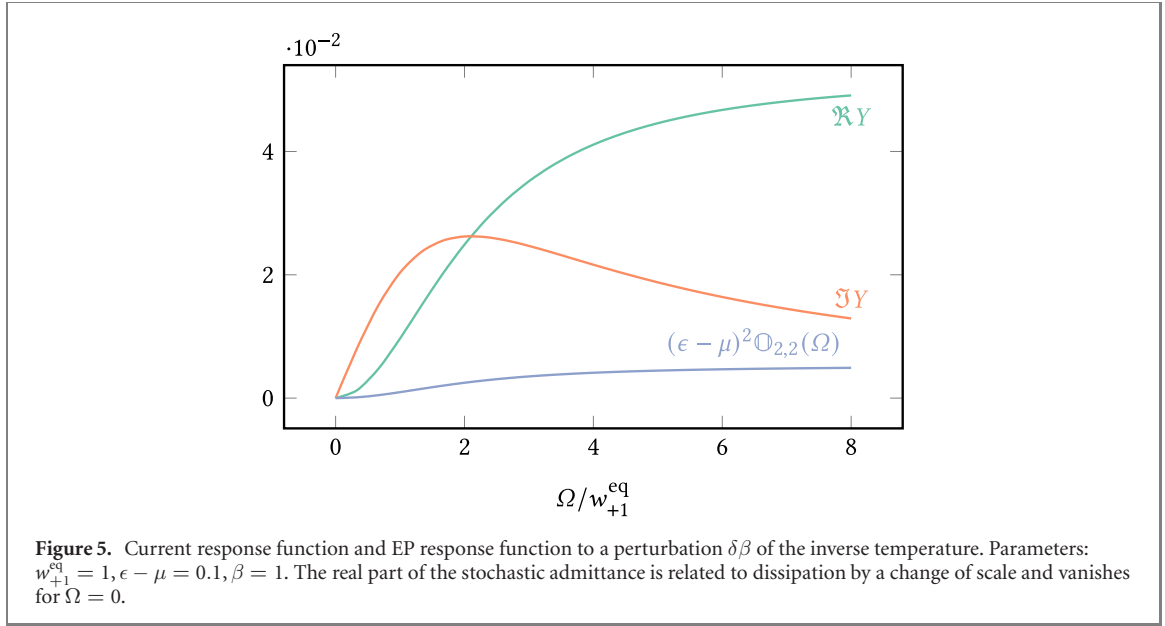
$$\mathbb{D} = \begin{matrix} & +1 & +2 \\ \begin{matrix} 0 \\ 1 \end{matrix} & \begin{pmatrix} -1 & 1 \\ 1 & -1 \end{pmatrix} \end{matrix}, \quad (144)$$

where a new transition path labeled +2 appears. The vector of entropic fields is then given by

$$\mathbf{f} = (\beta \quad -\beta\mu \quad \beta' \quad -\beta'\mu')^\top. \quad (145)$$

The matrix of extensive quantities coincide with (127), as the states of the system are unaffected by the connection of the second reservoir. The extensive quantities exchanged by the reservoirs with the system in transitions +1 and +2 are

$$\mathbb{X}^\top = \begin{matrix} & E_l & N_l & E_r & N_r \\ \begin{matrix} +1 \\ +2 \end{matrix} & \begin{pmatrix} \epsilon & 1 & 0 & 0 \\ 0 & 0 & -\epsilon & -1 \end{pmatrix} \end{matrix}. \quad (146)$$



LDB reads

$$\frac{w_{+1}}{w_{-1}} = e^{-\beta(\epsilon-\mu)}, \quad \frac{w_{+2}}{w_{-2}} = e^{\beta'(\epsilon-\mu')}, \quad (147)$$

and the reference equilibrium is defined setting $\beta = \beta'$ and $\mu = \mu'$. Under these conditions the rates satisfy detailed-balance for the equilibrium probability of occupancy $p^{\text{eq}} = e^{-\beta(\epsilon-\mu)}(1 + e^{-\beta(\epsilon-\mu)})^{-1}$. The equilibrium fluxes are

$$\mathbf{j}^{\text{eq}} = \begin{pmatrix} j_{+1}^{\text{eq}} \\ j_{+2}^{\text{eq}} \end{pmatrix} = \begin{pmatrix} w_{+1}(1 - p^{\text{eq}}) \\ w_{+2}p^{\text{eq}} \end{pmatrix}. \quad (148)$$

Define $j^{\text{eq}} := j_1^{\text{eq}} + j_2^{\text{eq}}$ and $w_{\pm} := w_{\pm 1} + w_{\pm 2}$. The matrices that appear in the response functions are then

$$i\omega\mathbb{P} + \langle \mathbb{D}^{\text{T}}, \mathbb{D}^{\text{T}} \rangle = \begin{pmatrix} i\omega(1 - p^{\text{eq}}) + j^{\text{eq}} & -j^{\text{eq}} \\ -j^{\text{eq}} & i\omega p^{\text{eq}} \end{pmatrix} \quad (149)$$

$$\langle \mathbb{D}^{\text{T}}, \mathbb{X}^{\text{T}} \rangle = j^{\text{eq}} \begin{pmatrix} -\epsilon & -1 & -\epsilon & -1 \\ \epsilon & 1 & \epsilon & 1 \end{pmatrix}. \quad (150)$$

The dynamic response of the occupation probability to a generic perturbation δf is

$$\delta \hat{p}(\omega) = -\frac{1}{2} \frac{w_{\pm}^{\text{eq}}}{(w_{+}^{\text{eq}} + w_{-}^{\text{eq}}) + i\omega} (\epsilon, 1, \epsilon, 1)^{\text{T}} \delta \hat{f}(\omega). \quad (151)$$

This system has a single cycle $\mathbf{C} = (+1, -1)$ in $\ker \mathbb{D}$, and thus $\mathbb{M} = \mathbb{X}\mathbf{C} = (\epsilon, 1, -\epsilon, -1)$. The conservation laws are thus given by the equation $\epsilon x_1 + x_2 - \epsilon x_3 - x_4 = 0$. Notice that the system is tight-coupled: in addition to the (trivial) conservation of energy and particles $\ell_E = (1, 0, 1, 0)$ and $\ell_N = (0, 1, 0, 1)$, we have the proportionality link between the exchange of energy and particles $\ell_{\text{t.c.}} = (-1/\epsilon, 1, 0, 0)$.

Indeed, the conserved quantities \mathbb{L} corresponding to ℓ_E and ℓ_N , coincide with the *trivial* ones contained in (127). In contrast, the last conservation law, $\ell_{\text{t.c.}}$, is system-specific and reflects the tight-coupling between energy and mass transport. The corresponding conserved quantity $L_{\text{t.c.}}$ can be chosen to be any constant vector.

We now select the right reservoir (characterized by the fields β' and μ') as the one that identifies the reference equilibrium of the system. The other reservoir—the left one—is instead seen as the one breaking detailed balance and driving the system out of equilibrium. Because of the tight-coupling conservation law, one (rather than two, see equation (21)) fundamental force is engendered. To characterize this force, we split the entropic field vector is $\mathbf{f}_f = \beta$, and $\mathbf{f}_p = (-\beta\mu, \beta', -\beta'\mu')^{\text{T}}$, which corresponds to splitting the matrix of conservation laws as

$$\mathbb{f}_f = (0, 1, -1/\epsilon), \quad \mathbb{f}_p = \begin{pmatrix} 1 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix}. \quad (152)$$

The transformation matrix \mathbb{T} then reads

$$\mathbb{T} = \begin{pmatrix} -1 & -1/\epsilon & 1 & 1/\epsilon \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (153)$$

Applying equation (20) we obtain

$$\delta\mathcal{F} = [(\epsilon - \mu')\beta' - (\epsilon - \mu)\beta]/\epsilon, \quad (154)$$

which can be read as an effective difference of chemical potentials rescaled by inverse temperatures. Notice how this nonconservative force depends explicitly on the system quantity ϵ , and hence on the protocol. However, notice also that this dependence disappears when $\beta = \beta'$. The Massieu potential of the states contains no entropic contribution and reads

$$\phi = (0, \beta'(\epsilon - \mu'))^\top. \quad (155)$$

We now consider a perturbation with a single frequency Ω . The physical currents and the EP response matrices read respectively

$$\mathbb{T}^\top \nabla \hat{I}(\Omega) \mathbb{T} = \frac{1}{(w_+^{\text{eq}} + w_-^{\text{eq}} + i\Omega)} \begin{pmatrix} j_{+1}^{\text{eq}} \epsilon^2 (w_{+2}^{\text{eq}} + w_{-2}^{\text{eq}} + i\Omega) & 0 & -ij_{+1}^{\text{eq}} \Omega \epsilon^2 & -ij_{+1}^{\text{eq}} \Omega \epsilon \\ 0 & 0 & 0 & 0 \\ -ij_{+1}^{\text{eq}} \Omega \epsilon^2 & 0 & i\Omega \epsilon^2 (j_{+1}^{\text{eq}} + j_{+2}^{\text{eq}}) & i\Omega \epsilon (j_{+1}^{\text{eq}} + j_{+2}^{\text{eq}}) \\ -ij_{+1}^{\text{eq}} \Omega \epsilon & 0 & i\Omega \epsilon (j_{+1}^{\text{eq}} + j_{+2}^{\text{eq}}) & i\Omega (j_{+1}^{\text{eq}} + j_{+2}^{\text{eq}}) \end{pmatrix}, \quad (156)$$

$$\mathbb{O}(\Omega) = \frac{1}{[(w_+^{\text{eq}} + w_-^{\text{eq}})^2 + \Omega^2]} \begin{pmatrix} j_{+1}^{\text{eq}} \epsilon^2 [(w_+^{\text{eq}} + w_-^{\text{eq}})(w_{+2}^{\text{eq}} + w_{-2}^{\text{eq}}) + \Omega^2] & 0 & -j_{+1}^{\text{eq}} \Omega^2 \epsilon^2 & -j_{+1}^{\text{eq}} \Omega^2 \epsilon \\ 0 & 0 & 0 & 0 \\ -j_{+1}^{\text{eq}} \Omega^2 \epsilon^2 & 0 & \Omega^2 \epsilon^2 j_+^{\text{eq}} & \Omega^2 \epsilon j_+^{\text{eq}} \\ -j_{+1}^{\text{eq}} \Omega^2 \epsilon & 0 & \Omega^2 \epsilon j_+^{\text{eq}} & \Omega^2 j_+^{\text{eq}} \end{pmatrix}. \quad (157)$$

As discussed in section 4.2, the existence of a tight coupling conservation law makes these two matrices degenerate at every frequency. Notice, furthermore, that these two matrices coincide at steady state, i.e. when $\Omega = 0$.

5.3. Photoelectric two-levels nanodevice

As a final example, we consider a photoelectric nanodevice that uses energy from incoming radiation to transport particles between two reservoirs maintained at different chemical potential [85], depicted in figure 3. In this model, the transport of energy and matter are decoupled. This follows from the presence of a transition between the two levels that is mediated by the absorption or emission of a photon, and not by an exchange of electrons with the reservoirs. This implies that the system is not tight-coupled for generic choices of the parameters, as we will show in the following.

The incidence matrix reads

$$\mathbb{D} = \begin{matrix} & +1 & +2 & +3 & +4 \\ \begin{matrix} 0 \\ 1 \\ 2 \end{matrix} & \begin{pmatrix} -1 & -1 & 0 & 0 \\ 1 & 0 & -1 & -1 \\ 0 & 1 & 1 & 1 \end{pmatrix} \end{matrix}, \quad (158)$$

where the different labels refer to the transitions depicted in figure 5. The vector of entropic fields is then given by

$$\mathbf{f} = (\beta \quad -\beta\mu_1 \quad -\beta\mu_2 \quad \beta_s)^\top. \quad (159)$$

The matrix of extensive quantities exchanged between system and reservoirs reads

$$\mathbb{X}^\top = \begin{matrix} & E_g & N_l & N_r & E_s \\ \begin{matrix} +1 \\ +2 \\ +3 \\ +4 \end{matrix} & \begin{pmatrix} \epsilon_d & 1 & 0 & 0 \\ \epsilon_u & 0 & 1 & 0 \\ 0 & 0 & 0 & \epsilon_u - \epsilon_d \\ \epsilon_u - \epsilon_d & 0 & 0 & 0 \end{pmatrix} \end{matrix}. \quad (160)$$

The cycles in the kernel of \mathbb{D} are contained in the matrix

$$\mathbb{C} = \begin{pmatrix} 1 & 0 \\ -1 & 0 \\ 0 & 1 \\ 1 & -1 \end{pmatrix}, \quad (161)$$

and the physical topology matrix reads

$$\mathbb{M} = \begin{pmatrix} 0 & \epsilon_d - \epsilon_u \\ 1 & 0 \\ -1 & 0 \\ 0 & \epsilon_u - \epsilon_d \end{pmatrix}. \quad (162)$$

The conservation law vectors are $\ell_1 = (1, 0, 0, 1)$ and $\ell_2 = (0, 1, 1, 0)$, and they are related through the balance (16) respectively to the conservation of energy and particles. We choose the fields β , μ_1 as the ones fixing the reference equilibrium. The two fundamental forces that result from such a choice are obtained applying equation (20), and are $\mathcal{F}_1 = \beta(\mu_2 - \mu_1)$ and $\mathcal{F}_2 = \beta - \beta_s$. The former corresponds to the force created by the chemical potential gradient of the particle reservoir. The latter is instead created by the thermal gradient generated by the coupling of the system plus particle reservoirs with the radiation source. Thus the reference equilibrium is achieved when the temperature of the radiative source is the same as the one of the ground, $\beta_s = \beta$, and when the two reservoirs have the same chemical potential, $\mu_2 = \mu_1$.

For this model, the generalized Onsager matrix $\mathbb{O}(\omega)$ and response matrix of the current \mathbb{Q} can be obtained analytically, using the same procedure of the previous examples. However, the resulting expressions of the two matrices are neither compact nor enlightening enough to be included. Nevertheless, we can make a conceptually important observation by studying the determinant of the generalized Onsager matrix evaluated in the limit $\omega \rightarrow 0$, that is

$$\det \mathbb{O}(0) = 16 \frac{(\epsilon_d - \epsilon_u)^4 e^{\beta(3\mu_1 + \mu_2 + 2\epsilon_d + 2\epsilon_u) + \beta_s(\epsilon_d - \epsilon_u)}}{(e^{\beta(\mu_1 + \epsilon_d)} + e^{\beta(\epsilon_d + \epsilon_u)} + e^{\beta(\mu_1 + \epsilon_u)})^4}. \quad (163)$$

Since it is proportional to a power of the difference between the energy levels ($\epsilon_d - \epsilon_u$), the matrix becomes singular when the two levels have the same energy. This condition also makes the system tightly coupled. In fact, if $\epsilon_d = \epsilon_u$, a new conservation law appears, namely $\ell_{t,c} = (0, 0, 0, 1)$, which belongs to the cokernel of \mathbb{X} .

6. Conclusions and outlook

We now summarize our results going from the most general to the special cases covered by our theory.

The near equilibrium decomposition of the EPR in equation (118) is a direct consequence of the systematic introduction of conservation laws in ST [29]. This decomposition describes the dissipation at finite times caused by a general, non-periodic protocol acting both on the extensive quantities of the states of the system and on the intensive parameters of the reservoirs. The decomposition strongly constrains the dynamics near equilibrium, as it immediately implies the minimum EP principle, in absence of time-dependent driving. In our formulation, the EPR is made up of two separately positive contributions. The first one is a quadratic form in the vector $\delta\mathcal{F}$ of fundamental thermodynamic forces and identifies the static part of the Onsager matrix. This term accounts for the dissipation due to maintaining a steady state, while the other contribution describes the dissipation due to purely dynamical effects as it vanishes at steady state or for adiabatic driving.

In the special case of periodic driving, the system settles on a periodic steady state. We derived the linear response matrix (100) for the Fourier components of the currents, concluding that it is not possible to generate currents against a nonconservative force by only performing time-dependent driving on the fields that define the equilibrium. Response coefficients for conserved quantities can also be derived and expressed via Green–Kubo-like formulas (107), and this general expression reduces to the classic one (110) when detailed balanced protocols are considered. The EPR (111) allows to identify the minimal subset of generalized thermodynamic forces that define a periodic protocol, namely the nonconservative forces (20) and the Massieu potential (27) of each state of the system. The decomposition (111), as well as the more general (118), also describes the regime of adiabatic driving, for which the only dissipation and currents are associated to the existence of nonzero average nonconservative forces.

Finally, restricting ourselves to protocols that only act on the reservoir intensive parameters, we obtained an expression for the response of the currents (81) and for the EPR in equation (75), and we call the latter a

generalized Onsager matrix (for each frequency mode present in the driving). To account for all possible mechanisms of dissipation, the set of fundamental forces must be extended by using the potential fields that define the time-dependent equilibrium. Employing the formalism of conservation laws [29], we are able to construct these matrices such that they are always symmetric. Even if the explicit time dependence of the protocol does not destroy the symmetry, it makes the response matrix of the currents different from the generalized Onsager matrix (75), unless steady perturbations are used. Our conclusion is that time-dependent driving near equilibrium does not break the time-reversal symmetry of the dynamics as far as response coefficients are concerned, as these are computed employing equilibrium quantities only. This provides an alternative point of view compared to that presented in references [53, 55], where a single, non-symmetric and protocol-dependent matrix was used to obtain both the time averaged currents and the EPR. Furthermore, we proved the connection between the existence of tight-coupling conservation laws and the vanishing of the determinant of the generalized Onsager matrix.

The present work only focuses on the thermodynamics of linear response around equilibrium. Linear response of far-from-equilibrium steady has been explored using ST in recent years [65, 86–90] but the implications of a proper identification of the thermodynamic forces in such situations is left for future work. This work also provides a useful starting point to design optimal protocols in the linear regime. In fact, equations (71) and (74) allow for straightforward optimization procedures to obtain protocols that minimize dissipation in steady or periodic working conditions. This complements previous studies on optimal protocols in the linear regime [91, 92] as well as the approaches based on thermodynamic length [45, 47, 93, 94], that are concerned with transient transformations between different steady states.

Acknowledgments

This work was supported by grants from the European Research Council, project NanoThermo (ERC-2015-CoG Agreement No. 681456, DF and ME), and from the Simons Foundation (691552, RR).

Data availability statement

No new data were created or analysed in this study.

ORCID iDs

Danilo Forastiere  <https://orcid.org/0000-0001-8803-0148>

References

- [1] Prigogine I 1947 *Etude thermodynamique des phénomènes irréversibles* (Paris: Dunod)
- [2] Callen H B 1948 *Phys. Rev.* **73** 1349
- [3] Prigogine I 1949 *Physica* **15** 272
- [4] de Groot S R and Mazur P 1962 *Non-Equilibrium Thermodynamics* (Amsterdam: North-Holland)
- [5] Glansdorff P and Prigogine I 1971 *Thermodynamic Theory of Structure, Stability and Fluctuations* (New York: Wiley-Interscience)
- [6] Callen H B 1985 *Thermodynamics and An Introduction to Thermostatistics* (New York: Wiley)
- [7] Onsager L 1931 *Phys. Rev.* **37** 405
- [8] Onsager L 1931 *Phys. Rev.* **38** 2265
- [9] Callen H B, Barasch M L and Jackson J L 1952 *Phys. Rev.* **88** 1382
- [10] Callen H B and Greene R F 1952 *Phys. Rev.* **86** 702
- [11] Greene R F and Callen H B 1952 *Phys. Rev.* **88** 1387
- [12] Wigner E P 1954 *J. Chem. Phys.* **22** 1912
- [13] Kirkwood J G 1946 *J. Chem. Phys.* **14** 180
- [14] Callen H B and Welton T A 1951 *Phys. Rev.* **83** 34
- [15] Green M S 1952 *J. Chem. Phys.* **20** 1281
- [16] Green M S 1954 *J. Chem. Phys.* **22** 398
- [17] Bergmann P G and Lebowitz J L 1955 *Phys. Rev.* **99** 578
- [18] Kubo R 1957 *J. Phys. Soc. Japan* **12** 570
- [19] Schnakenberg J 1976 *Rev. Mod. Phys.* **48** 571
- [20] Sekimoto K 2010 *Stochastic Energetics* vol 799 (Berlin: Springer)
- [21] Hill T L 1977 *Free Energy Transduction in Biology* (New York: Academic)
- [22] Jiu-Li L, Van Den Broeck C and Nicolis G 1984 *Z. Phys. B* **56** 165
- [23] Mou C Y, Luo J-I and Nicolis G 1986 *J. Chem. Phys.* **84** 7011
- [24] Jarzynski C 1997 *Phys. Rev. E* **56** 5018
- [25] Crooks G E 1999 *Phys. Rev. E* **60** 2721
- [26] Seifert U 2005 *Phys. Rev. Lett.* **95** 040602

- [27] Seifert U 2012 *Rep. Prog. Phys.* **75** 126001
- [28] Zhang X-J, Qian H and Qian M 2012 *Phys. Rep.* **510** 1
- [29] Rao R and Esposito M 2018 *New J. Phys.* **20** 023007
- [30] Pigolotti S and Peliti L 2021 *Stochastic Thermodynamics: An Introduction* (Princeton, NJ: Princeton University Press)
- [31] Esposito M 2012 *Phys. Rev. E* **85** 041125
- [32] Maes C 2021 *SciPost Phys. Lect. Notes* **32**
- [33] Falasco G and Esposito M 2021 *Phys. Rev. E* **103** 042114
- [34] Esposito M, Harbola U and Mukamel S 2009 *Rev. Mod. Phys.* **81** 1665
- [35] Breuer H-P and Petruccione F 2002 *The Theory of Open Quantum Systems* (Oxford: Oxford University Press)
- [36] Bochkov G and Kuzovlev Y E 1977 *Zh. Eksp. Teor. Fiz.* **72** 238
- [37] Evans D J, Cohen E G D and Morriss G P 1993 *Phys. Rev. Lett.* **71** 2401
- [38] Gallavotti G and Cohen E G D 1995 *Phys. Rev. Lett.* **74** 2694
- [39] Gallavotti G 1996 *Phys. Rev. Lett.* **77** 4334
- [40] Kurchan J 1998 *J. Phys. A: Math. Gen.* **31** 3719
- [41] Lebowitz J L and Spohn H 1999 *J. Stat. Phys.* **95** 333
- [42] Maes C 1999 *J. Stat. Phys.* **95** 367
- [43] Pekola J P 2015 *Nat. Phys.* **11** 118
- [44] Ciliberto S 2017 *Phys. Rev. X* **7** 021051
- [45] Crooks G E 2007 *Phys. Rev. Lett.* **99** 100602
- [46] Feng E H and Crooks G E 2009 *Phys. Rev. E* **79** 012104
- [47] Sivak D A and Crooks G E 2012 *Phys. Rev. Lett.* **108** 190602
- [48] Izumida Y and Okuda K 2009 *Phys. Rev. E* **80** 021121
- [49] Brandner K, Saito K and Seifert U 2015 *Phys. Rev. X* **5** 031019
- [50] Proesmans K and Van den Broeck C 2015 *Phys. Rev. Lett.* **115** 090601
- [51] Proesmans K, Cleuren B and Van den Broeck C 2016 *J. Stat. Mech.* **023202**
- [52] Cleuren B and Proesmans K 2020 *Physica A* **552** 122789
- [53] Proesmans K and Fiore C E 2019 *Phys. Rev. E* **100** 022141
- [54] Andrieux D and Gaspard P 2004 *J. Chem. Phys.* **121** 6167
- [55] Brandner K and Seifert U 2016 *Phys. Rev. E* **93** 062134
- [56] Polettni M, Bulnes-Cuetara G and Esposito M 2016 *Phys. Rev. E* **94** 052117
- [57] Altaner B, Wachtel A and Vollmer J 2015 *Phys. Rev. E* **92** 042133
- [58] Rao R and Esposito M 2018 *J. Chem. Phys.* **149** 245101
- [59] Forastiere D, Falasco G and Esposito M 2020 *J. Chem. Phys.* **152** 134101
- [60] Esposito M, Harbola U and Mukamel S 2007 *Phys. Rev. E* **76** 031132
- [61] Freitas N, Delvenne J-C and Esposito M 2021 *Phys. Rev. X* **11** 031064
- [62] Horn R A and Johnson C R 2012 *Matrix Analysis* (Cambridge: Cambridge University Press)
- [63] Polettni M and Esposito M 2013 *Phys. Rev. E* **88** 012112
- [64] Maes C and Netočný K 2010 *J. Math. Phys.* **51** 015219
- [65] Baiesi M and Maes C 2013 *New J. Phys.* **15** 013004
- [66] McLennan J A Jr 1959 *Phys. Rev.* **115** 1405
- [67] Mori H 1958 *Phys. Rev.* **112** 1829
- [68] Green M S 1960 *Phys. Rev.* **119** 829
- [69] Agarwal G S 1972 *Z. Phys. A* **252** 25
- [70] Marconi U M B, Puglisi A, Rondoni L and Vulpiani A 2008 *Phys. Rep.* **461** 111
- [71] Hill T L 1982 *Nature* **299** 84
- [72] Van den Broeck C 2005 *Phys. Rev. Lett.* **95** 190602
- [73] Chernyak V Y and Sinityn N A 2008 *Phys. Rev. Lett.* **101** 160601
- [74] Rahav S, Horowitz J and Jarzynski C 2008 *Phys. Rev. Lett.* **101** 140602
- [75] Maes C, Netočný K and Thomas S R 2010 *J. Chem. Phys.* **132** 234116
- [76] Sinityn N A and Nemenman I 2007 *Phys. Rev. Lett.* **99** 220408
- [77] Esposito M and Van den Broeck C 2010 *Phys. Rev. Lett.* **104** 090601
- [78] Esposito M and Van den Broeck C 2010 *Phys. Rev. E* **82** 011143
- [79] Hatano T and Sasa S-i 2001 *Phys. Rev. Lett.* **86** 3463
- [80] Rao R and Esposito M 2018 *Entropy* **20** 635
- [81] Keizer J and Fox R F 1974 *Proc. Natl Acad. Sci. USA* **71** 192
- [82] Hunt K, Hunt P and Ross J 1987 *Physica A* **147** 48
- [83] Ross J and Vlad M O 2005 *J. Phys. Chem. A* **109** 10607
- [84] Cover T M and Thomas J A 1991 *Information Theory and Statistics* (New York: Wiley)
- [85] Ruten B, Esposito M and Cleuren B 2009 *Phys. Rev. B* **80** 235122
- [86] Prost J, Joanny J-F and Parrondo J 2009 *Phys. Rev. Lett.* **103** 090601
- [87] Speck T and Seifert U 2009 *Phys. Rev. E* **79** 040102
- [88] Altaner B, Polettni M and Esposito M 2016 *Phys. Rev. Lett.* **117** 180601
- [89] Vroylandt H, Lacoste D and Verley G 2018 *J. Stat. Mech.* **023205**
- [90] Owen J A, Gingrich T R and Horowitz J M 2020 *Phys. Rev. X* **10** 011066
- [91] Bauer M, Brandner K and Seifert U 2016 *Phys. Rev. E* **93** 042112
- [92] Proesmans K, Cleuren B and Van den Broeck C 2016 *Phys. Rev. Lett.* **116** 220601
- [93] Zulkowski P R, Sivak D A, Crooks G E and DeWeese M R 2012 *Phys. Rev. E* **86** 041148
- [94] Mandal D and Jarzynski C 2016 *J. Stat. Mech.* **063204**