Master equation based formulation of nonequilibrium statistical mechanics

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For a nonequilibrium system characterized by its state space, by a dynamics defined by a transfer matrix and by a reference equilibrium dynamics given by a detailed-balance transfer matrix, we define various nonequilibrium concepts: relative entropy, dissipation during the relaxation to the stationary state, path entropy, cost for maintaining the system in a nonequilibrium state, fluctuation-dissipation theory, and finally a tree integral formula for the stationary state. © *1996 American Institute of Physics.* [S0022-2488(96)02808-3]

I. INTRODUCTION

For systems that are not in equilibrium, much of the general power of thermodynamics and statistical mechanics is lost. For chemical reactions, for fluids, for dynamic critical phenomena, or metastable states, and for many, many natural, social, and economic systems, specific methods have been developed to deal with time-dependent collective phenomena (see among many possible references [1–9]). The absence of overriding laws, such as the entropy-related variational principles of equilibrium statistical mechanics, has long been lamented, although there have been many attempts, for example to define generalizations of thermodynamic functions (see Refs. 1, 2, 6, and 10–12 for recent definitions). In the present paper we use a dynamical framework broad enough to cover most of the phenomena of interest and find that there *are* general statements that can be made. Of course, there is a kind of complementarity principle. The vast range of nonequilibrium phenomena in open systems precludes certain kinds of specific predictions and forces on us a level of abstraction that may limit usefulness.

The framework is the master equation. A state space and transition probabilities between states are given. This will not describe situations where quantum interference is important, but is nevertheless rather comprehensive—even finite memory effects can be included by enlarging the state space. In its various forms, for example, the Fokker–Planck equation, the master equation has already been used in many contexts. Our goal will be to seek general versions of the broadest kind of equilibrium information, things analogous to entropy inequalities, fluctuation-dissipation theorems, and the characterization of the steady state, when there is one.

Label the states x, $y \in X$ and the transition probabilities R_{xy}, defined as the (conditional) probability that the state of the system at time $t+\Delta t$ is x, given that it was y at time t. For most of the present paper, we take X and Δt finite. The stochastic matrix R is *not* assumed to satisfy detailed balance (for any vector) and indeed *it is this feature that is of greatest interest.* To avoid irrelevant mathematical complications, R is assumed to be irreducible.

For some of our results it would be easy to take continuum limits. Indeed in previous works (Refs. 13–15) we used the master equation approach advocated here to define a metastable state and in Ref. 16 to establish "self-organized criticality" (see Refs. 17 and 18) in a model system. These results were based on showing the disappearance of an energy gap, clearly going beyond the finite state context. Similarly, in Ref. 19 various critical properties in directed percolation derive

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from the spectrum and eigenfunctions of the matrix R, in particular, its infinite size limit.

Within this framework there emerges the important general concept of *current*. By this we mean the flow of probability that can exist, even in the stationary state: $J_{xy}=R_{xy}\tilde{p}_y-R_{yx}\tilde{p}_x$, where $R\tilde{p}=\tilde{p}$. Currents, in particular current loops, which do not exist at equilibrium, are essential to anything one would consider *complex* as a dynamical system (see, e.g., Ref. 20 for different perspectives and Ref. 21 for other opinions). One must be careful here to distinguish currents in X from currents in an underlying physical coordinate space. For example, in this terminology, heat conduction with detailed balance is not complex, even though it is not in equilibrium. We shall discuss these matters in another publication.

The present paper is intended as an exposition of our general framework. In developing this framework we have had a number of examples in mind and in future publications we intend to exhibit these applications. However, because the present exposition is already rather lengthy, we will give only minimal indications of these examples. It is also clear that the wealth of potential applications will require tweaking of our framework. For example, directed percolation on finite systems generally has a trivial (absorbing) stationary state. By minor modification of the dynamics the interesting behavior of such systems can be studied with the present techniques (see, for example Ref. 22). However, in the present paper we do not focus on those issues.

Summary of results. A natural construct is the relative entropy $S(p|q)=-\Sigma_x p_x \log(p_x/q_x)$ of two distributions. For equilibrium theory this is already important [e.g., if q is the Gibbs state, $S(p|q)$ is essentially a thermodynamic potential] and it is also used extensively in information theory. It is easy to show (and well known) that

$$S(p|q)\leqslant S(Rp|Rp)\leqslant 0.$$

Much of this work focuses on the invariant state of R, which is called $\tilde{p}$. Thus $\tilde{p}=R\tilde{p}$. The analog of the entropy increase in equilibrium systems is the fact that $S(p|\tilde{p})$ can only increase as R is successively applied to p. In fact, we have a stronger statement: If δ is small and $p_\delta(x)\equiv\tilde{p}(x)\exp(\delta\varphi_1(x))$, then $S(p_\delta|\tilde{p})\sim-\delta^2\langle\varphi_1^2\rangle_{\tilde{p}}/2$. This allows bounds on the rate of approach to stationarity (i.e., $\tilde{p}$). Let ${}^tR$ be the transpose of R and $({}^tR)^*$ be its adjoint with respect to the inner product using $\tilde{p}$ as a weight. Let φ_1 be the eigenvector of ${}^tR({}^tR)^*$ with maximum eigenvalue $\mu_{\max}$, different from 1, then

$$S(Rp|\tilde{p})-S(p|\tilde{p})\geqslant\tfrac{1}{2}\langle\delta^2\varphi_1^2\rangle(1-\mu_{\max}).$$

This is a statement about dissipation and fluctuations, although in a moment we shall get to the usual form. In this context we are also able to get results on "excess work," a concept that has been used in the chemical literature.[10]

One question of great interest is, what does it "cost" to keep the system out of equilibrium? The matrix R can describe a system with temperature gradients, with sunlight, with wind, with currency exchange rate shifts. How can one associate a general cost? Given the broad nature of our goals, we preferred not to model the reservoirs that maintain R's imbalances. Rather, we assume that R is to be compared with a fiducial W, which is a transition matrix with an equilibrium state and detailed balance $[W_{xy}p_{\rm eq}(y)=W_{yx}p_{\rm eq}(x)]$. For example, if R describes Rayleigh–Benard flow, then W could represent a world uniformly at the temperature of the upper (or the colder) plate. The cost should then be what it takes to heat the lower plate. The choice of W is made by the observer and is partly conventional, depending on what the observer or designer intends to do with the R matrix. For example, in a Carnot cycle completed by a necessarily out-of-equilibrium engine, depending on whether the cycle is used to move a car or function as a refrigerator, W would be the thermal state at low or high temperature, respectively. However, within our general framework one is not committed to such a detailed point of view.

By considering a path entropy, we find it appropriate to define

$$\Delta_1 \mathcal{I}(R,\tilde{p},W) = -\sum_{x,y} R_{xy}\tilde{p}(y)\log\left(\frac{R_{xy}}{W_{xy}}\right),$$

to be the dissipation per time step required to maintain the state $\tilde{p}$, against a tendency to relax to equilibrium. The following remarkable inequality emerges:

$$S(W\tilde{p}|p_{\rm eq}) - S(\tilde{p}|p_{\rm eq}) \leqslant 4\,\Delta_1 \mathcal{I}(R,\tilde{p},W).$$

This inequality is proved for $\tilde{p}$ near equilibrium. It means that if we start with a certain stationary state and switch off the reservoirs defining R, so that $\tilde{p}$ starts to relax to the equilibrium $p_{\rm eq}$ by the W evolution, the dissipation per unit time is always less than four times the cost to maintain $\tilde{p}$, as defined by the path entropy.

To state a nonequilibrium fluctuation-dissipation theorem in a form similar to its usual equilibrium formulation, it is necessary to climb down from the grand generality adopted until this point. A distinction must be drawn between fast and slow variables—the motion of one dissipates while the others fluctuate. In the context of our master equation model we achieve this result. In fact, what we get is stronger than what is known in the equilibrium case. In particular, we have independent expressions for fluctuation and dissipation and the comparison of these expressions gives the fluctuation-dissipation statement, while traditional derivations do not give separate expressions for fluctuation and dissipation. Note that the state around which this generalized fluctuation dissipation theorem holds is *not* equilibrium but is the stationary state $\tilde{p}$. More precisely, let A be a slow variable of the system that is chosen to be a left eigenvector of the transfer matrix R whose eigenvalue is close to 1, and let p_α be a perturbation of the stationary state, such that $\langle A(0)\rangle_{p_\alpha}$ (average of A at time 0 in the state p_α) is given, then the dissipation is

$$\langle A(\Delta t) - A(0)\rangle \sim \langle A(0)\rangle_{p_\alpha}(\lambda - 1),$$

while the fluctuation is

$$\langle (A(\Delta t) - A(0))\rangle \sim (\lambda - 1)(-2\langle A^2\rangle_{p_\alpha}) + O(\alpha).$$

The elimination of $\lambda - 1$ then provides the analog of the fluctuation-dissipation relation in a nonequilibrium stationary state.

Finally, we state a generalization of the Onsager reciprocity relations for a general nonequilibrium system. The Onsager coefficients L_{kj} are not, in general, symmetric, but they are symmetric in the case of detailed balance dynamics.

In general, most of our statements (with the exception of the statements of Secs. II A and II B), hold for states near the stationary state, or for stationary states near an equilibrium state of reference (and an R matrix near a detailed balance matrix W). In our general framework, it is difficult to estimate how "near" one must be so that our statements remain valid. In particular, we do not discuss criticality (although everything we say is valid in this context too). Finally, we present a general expression for the stationary state of any stochastic matrix R. This is potentially important: for equilibrium theory, merely writing down the Gibbs state, $\exp(-\beta H)$, is a major step toward calculating various quantities. Our expression for the state is in terms of a sum over spanning trees built out of R and is reminiscent of a path integral formula; actually it is a "tree-integral formula." At the computational level this may turn out to be difficult to work with. (An Ising model with 5 spins has a 32×32 transition matrix. The number of spanning trees on 32 objects is about 10^{45}.) However, for formal manipulations it should be useful; for example, it could lead to an abstract definition of nonequilibrium phase transition.

II. DISSIPATION DURING THE RELAXATION TO THE STATIONARY STATE

In the following, X denotes a discrete space with points $x,y,\ldots$. We start by recalling elementary facts about probability distributions and their entropy on X.

A. Relative entropy of two probability distributions

Let p and q be two probability distributions on X. The relative entropy is defined to be

$$S(p|q)=-\sum_{x\in X} p(x)\log\frac{p(x)}{q(x)}.$$

By convention, $0\log 0=0$. It follows that

$$S(p|q)\leqslant 0. \tag{2.1}$$

The proof is immediate. We have

$$S(p|q)=-\sum p(x)\log\frac{p(x)}{q(x)}=-\sum q(x)L\left(\frac{p(x)}{q(x)}\right),$$

where $L(\xi)\equiv\xi\log\xi$. But $L'(\xi)=1+\log\xi$ and $L''\xi)=(1/\xi)\geqslant 0$, so L is convex. As a consequence, $L(\Sigma_x q(x)\alpha(x))\leqslant\Sigma q(x)L(\alpha(x))$, provided $\Sigma_x q(x)=1$. So

$$S(p|q)\leqslant -L\left(\sum_x q(x)\frac{p(x)}{q(x)}\right)=-L(1)=0.$$

Remark: If $\tilde{p}$ is an equilibrium distribution of the form $\tilde{p}(x)=\exp(-\beta E_x)/\mathcal{Z}$, the quantity S is (up to a sign) the corresponding thermodynamic potential. Specifically, $S(q|p)=\beta[\tilde{F}-F_q]$, where $\tilde{F}=-T\log\mathcal{Z}$ (the usual free energy) and $F_q=\langle E\rangle_q-T(-\Sigma q\log q)$.

B. Increase of the relative entropy

The result below, Eq. (2.2), is derived as in Ref. 23 but adapted to our notation.

We consider two distributions p_0,q_0 and a Markov chain on X, with transition matrix R_{xy} ($R_{xy}\equiv R_{x\leftarrow y}$ is the probability that starting from y, one has a transition $y\to x$ in unit time step). We call p_1, q_1 the probability distributions at time 1,

$$p_1(x)=\sum R_{xy}p_0(y),\quad q_1(x)=\sum R_{xy}q_0(y).$$

Then

$$S(p_0|q_0)\leqslant S(p_1|q_1). \tag{2.2}$$

Proof: Consider the states of the Markov chain at times 0 and 1, namely $\{x_0,x_1\}$. If the initial probability distribution is p_0, the joint law of $\{x_0,x_1\}$ is $P(x_0,x_1)=R_{x_1x_0}p_0(x_0)$, and if the initial probability distribution is q_0, the joint law of $\{x_0,x_1\}$ is $Q(x_0,x_1)=R_{x_1x_0}q_0(x_0)$. Then

$$S(P|Q)=-\sum_{x_0,x_1}P(x_0,x_1)\log\frac{P(x_0,x_1)}{Q(x_0,x_1)}=-\sum_{x_0,x_1}R_{x_1x_0}p_0(x_0)\log\frac{p_0(x_0)}{q_0(x_0)}=S(p_0|q_0).$$

Now, we compute $S(P|Q)$ in a different way. We can write P and Q by conditioning the past x_0 knowing the future x_1 in the following way:

$$P(x_0,x_1)=p_1(x_1)r_{x_1x_0}, \quad Q(x_0,x_1)=q_1(x_1)s_{x_1x_0},$$

where $p_1(x_1)$ [resp., $q_1(x_1)$] are the distribution probabilities of x_1, the initial distribution of x_0 being $p_0(x_0)$ [resp., $q_0(x_0)$], and where $r_{x_1x_0}$ is the distribution probability of x_0, knowing that the position at time 1 of the chain is x_1 [given the fact that the distribution of x_1 is $p_1(x_1)$], and, in the same manner, $s_{x_1x_0}$ is the distribution probability of x_0 knowing that at time 1 the position of the chain is x_1 [given the fact that the distribution of x_1 is $q_1(x_1)$].

Then the same computation proves that

$$S(P|Q)=S(p_1|q_1)+\sum_{x_1} p_1(x_1)S(r_{x_1,*}|s_{x_1,*}),$$

where

$$S(r_{x_1,*}|s_{x_1,*})=-\sum_{x_0} r_{x_1x_0}\log\frac{r_{x_1x_0}}{s_{x_1x_0}}\leqslant 0.$$

So we have

$$S(p_0|q_0)=S(P|Q)\leqslant S(p_1|q_1).$$

Remark: For $\tilde{p}=\exp(-\beta E_x)/\mathcal{Z}$, this shows that $F_q=\Sigma E_x q(x)-TS(q|1)$ can only decrease. (This F_q is the same as defined in our remark at the end of the previous section.)

C. Computation near the stationary state

Although Eq. (2.2) is known in the information theory context, the matter we now discuss appears more relevant to physical and chemical systems as such. To the extent that similar or weaker results are known, they arise in the statistical mechanics literature. As we proceed, we shall give references wherever appropriate. In any case the results we now derive are not contained in Ref. 23. We shall see that they are completely general and do not refer to any special feature of the physical or chemical systems we consider.

As above, our system is described by a state space X and its evolution can be represented by a stochastic matrix R_{xy} (which is the probability of a transition $y\to x$ in a unit time step Δt). We assume that R has a unique stationary state $\tilde{p}(x)$ satisfying

$$\tilde{p}(x)=\sum_y R_{xy}\tilde{p}(y). \tag{2.3}$$

Remark: Note that by virtue of (2.2), for any q,

$$S(Rq|\tilde{p})\geqslant S(q|\tilde{p}). \tag{2.4}$$

We consider a neighboring state $p_\delta(x)$, where δ is a small parameter, namely

$$p_\delta(x)\equiv\tilde{p}(x)\exp(\varphi(x,\delta)),$$

where

$$\varphi(x,\delta)\equiv\delta\varphi_1(x)+\delta^2\varphi_2(x)+\cdots.$$

Then, for small δ, it follows that

$$S(p_\delta|\tilde{p}) \sim -\frac{\delta^2}{2}\langle\varphi_1^2\rangle_{\tilde{p}}, \tag{2.5}$$

where $\langle\varphi_1^2\rangle_{\tilde{p}}$ is the mean square average of φ_1 in the state $\tilde{p}$,

$$\langle\varphi_1^2\rangle_{\tilde{p}} = \sum_x \tilde{p}(x)\varphi_1^2(x).$$

Proof: We start from the definition

$$S(p_\delta|\tilde{p}) = \sum p_\delta(x)\log\frac{p_\delta(x)}{\tilde{p}(x)}.$$

Then

$$\frac{\partial S}{\partial\delta} = -\sum \frac{\partial p_\delta(x)}{\partial\delta}\left(1+\log\frac{p_\delta(x)}{\tilde{p}(x)}\right),$$

$$\frac{\partial^2 S}{\partial\delta^2} = -\sum \frac{\partial^2 p_\delta(x)}{\partial\delta^2}\left(1+\log\frac{p_\delta(x)}{\tilde{p}(x)}\right) - \sum \frac{1}{p_\delta(x)}\left(\frac{\partial p_\delta(x)}{\partial\delta}\right)^2.$$

Now we have $\Sigma p_\delta(x)=1$. Therefore $\Sigma\partial p_\delta(x)/\partial\delta=0$ and $\Sigma\partial^2 p_\delta(x)/\partial\delta^2=0$. It follows that

$$S(p_\delta|\tilde{p}) \sim -\frac{\delta^2}{2}\sum_x \frac{1}{p_\delta(x)}\left(\frac{\partial p_\delta(x)}{\partial\delta}\right)^2\Bigg|_{\delta=0}.$$

But

$$\left.\frac{\partial p_\delta(x)}{\partial\delta}\right|_{\delta=0} = p_\delta(x)\varphi_1(x).$$

D. Variation of entropy near the stationary state

We again consider a state $p_\delta(x)$ near the stationary state, and for simplicity we drop the δ index. We note that

$$p(x) = \tilde{p}(x)\exp(\delta\varphi_1(x)+\delta^2\varphi_2(x)+\cdots).$$

We consider at time step Δt (one time step) the evolution of p, namely

$$p(x,\Delta t) = \sum_y R_{xy}p(y).$$

We know by Eq. (2.2) of Sec. II B that

$$S(p|\tilde{p}) \leqslant S(p(\cdot,\Delta t)|\tilde{p}),$$

but here we shall find the difference between these two entropies. We can write

$$p(x,\Delta t) = \tilde{p}(x)\exp(\delta\psi_1(x)+\delta^2\psi_2(x)+\cdots).$$

Now

$$\tilde{p}(x)\exp(\delta\psi_1(x)+\delta^2\psi_2(x)+\cdots)=\sum_y R_{xy}\tilde{p}(y)\exp(\delta\varphi_1(y)+\delta^2\varphi_2(y)+\cdots).$$

Comparing the terms of order δ, we find

$$\psi_1(x)=\frac{1}{\tilde{p}(x)}\sum_y R_{xy}\tilde{p}(y)\varphi_1(y), \tag{2.6}$$

or in vector notation,

$$\psi_1=\left(\operatorname{diag}\frac{1}{\tilde{p}}\right)R(\operatorname{diag}\tilde{p})\varphi_1. \tag{2.6'}$$

Then, in our context, we have

$$S(p(\cdot,\Delta t)|\tilde{p})-S(p|\tilde{p})\sim-\frac{1}{2}\left[\left\langle\left(\frac{1}{\operatorname{diag}\tilde{p}}R\ \operatorname{diag}\tilde{p}\ \delta\varphi_1\right)^2\right\rangle_{\tilde{p}}-\langle(\delta\varphi_1)^2\rangle_{\tilde{p}}\right]. \tag{2.7}$$

We shall study a lower bound for this quantity. To do this, we maximize the quantity

$$\max\left\langle\left(\frac{1}{\operatorname{diag}\tilde{p}}R\ \operatorname{diag}\tilde{p}\varphi_1\right)^2\right\rangle_{\tilde{p}},$$

subject to the conditions $\langle\varphi_1^2\rangle_{\tilde{p}}=1$ and $\langle\varphi_1\rangle_{\tilde{p}}=0$. [The last condition is a consequence of $\Sigma_x p_\delta(x)=1$, so that $\Sigma\varphi_1(x)\tilde{p}(x)=0$.]

To find this maximum, we introduce a Lagrange multiplier μ for the constraint $\langle\varphi_1^2\rangle_{\tilde{p}}=1$ and we assume that φ_1 has been found. Then, for any variation $\varphi_1+\epsilon_1$, we must have

$$\sum_x\tilde{p}(x)\left(\frac{1}{\operatorname{diag}\tilde{p}}R\ \operatorname{diag}\tilde{p}\varphi_1\right)_x\left(\frac{1}{\operatorname{diag}\tilde{p}}R\ \operatorname{diag}\tilde{p}\epsilon_1\right)_x-\mu\sum\tilde{p}(x)\epsilon_1(x)=0.$$

Rearranging, the factor of $\epsilon_1(x)$ must vanish identically, so that

$${}^tR\ \operatorname{diag}\left(\frac{1}{\tilde{p}}\right)R(\operatorname{diag}\tilde{p})\varphi_1=\mu\varphi_1. \tag{2.8}$$

Denote $M={}^tR$ (so $M_{xy}=R_{yx}$). We notice that the adjoint M^* of M for the scalar product,

$$(v|w)_{\tilde{p}}=\sum\tilde{p}(x)v(x)^*w(x), \tag{2.9}$$

is just

$$M^*=\operatorname{diag}\left(\frac{1}{\tilde{p}}\right)R\ \operatorname{diag}\tilde{p}, \tag{2.10}$$

and so Eq. (2.8) can be rewritten as

$$MM^*\varphi_1=\mu\varphi_1. \tag{2.11}$$

MM^* is self-adjoint with respect to the scalar product $(\ |\ )_{\tilde{p}}$. Moreover, all its eigenvalues are obviously positive. Call $N=MM^*$. We show that all eigenvalues of N are less than 1. Let φ be eigenvector of eigenvalue μ,

$$\max_{y\in X}|N\varphi(y)|\leqslant\sum R_{xy}\frac{1}{\tilde{p}(x)}R_{xy}\tilde{p}(z)|\varphi(z)|\leqslant\max_{z}|\varphi(z)|\left(\sum_{x}R_{xy}\frac{1}{\tilde{p}(x)}\sum_{z}R_{xy}\tilde{p}(z)\right)$$

$$\leqslant\max|\varphi(z)|\left(\sum_{x}R_{xy}\frac{1}{\tilde{p}(x)}\tilde{p}(x)\right)=\max|\varphi(z)|,$$

but because of Eq. (2.11), $N\varphi=\mu\varphi$. We see that if $\varphi\not\equiv 0$, $\mu\leqslant 1$.

Obviously the matrix $N=MM^*$ has the eigenvalue 1 with trivial eigenvector $\{1\}$ because

$$\sum_{x,z}R_{xy}\frac{1}{\tilde{p}(x)}R_{xz}\tilde{p}(z)=1,\quad\text{for all } y.$$

Now in the variational problem above, we considered an eigenvector φ_1 that is orthogonal to the trivial eigenvector $\{1\}$ (because we imposed $\langle\varphi_1\rangle_{\tilde{p}}=0$). So, *we have proved that*

$$S(p(\cdot,\Delta t)|\tilde{p})-S(p|\tilde{p})\geqslant\tfrac{1}{2}\langle\delta^2\varphi_1^2\rangle_{\tilde{p}}(1-\mu_{\max}),\tag{2.12}$$

where $\mu_{\max}$ *is the maximal eigenvalue of* MM^* *corresponding to an eigenvector* φ_1 *orthogonal to the trivial eigenvector* $\{1\}$.

It remains to prove that $\mu_{\max}<1$. Suppose that $v(x)$ is an eigenvector of eigenvalue 1 but different from the trivial eigenvector $\{1\}$; thus $v(x)$ is orthogonal to $\{1\}$ for the scalar product $(\ |\ )_{\tilde{p}}$,

$$\sum\tilde{p}(x)v(x)=0.$$

We have

$$v(x)=\sum_{y,z}R_{yx}\frac{1}{\tilde{p}(y)}R_{yz}\tilde{p}v(z).$$

But $\tilde{p}(x)\neq 0$ for all x, so that at least one $v(x)$ must be negative. As a consequence, we obtain from the preceding inequality a strict inequality,

$$|v(x)|<\sum_{y,z}R_{yx}\frac{1}{\tilde{p}(y)}R_{yz}\tilde{p}(z)|v(z)|,$$

or finally

$$\max_{x\in X}|v(x)|<\max_{z\in X}|v(z)|,$$

which is a contradiction

Remark 1: All this assumes that $\tilde{p}(x)\neq 0$ for all x because we need to define $1/\tilde{p}(x)$ for all x. This is the case if R is irreducible. If R is reducible, one can sometimes introduce small matrix elements to make it irreducible while preserving its essential features, as in Ref. 22.

Remark 2: This "universal" inequality,

$$S(p(\cdot,\Delta(\cdot)|\tilde{p})-S(p(\cdot)|\tilde{p})\geq\tfrac{1}{2}\langle\delta^2\varphi_1^2\rangle_{\tilde{p}}(1-\mu_{\max})$$

is a kind of *fluctuation dissipation inequality* for nearby states, p, because the left side is the dissipation (in one time step) and the right side involves a fluctuation $\langle\delta^2\varphi_1^2\rangle$ of the relative free energy of p with respect to $\tilde{p}$ (and a factor that is $1-\mu_{\max}$).

Remark 3: If R satisfies detailed balance, then

$$M^*={}^tR$$

and

$$MM^*=({}^tR)^2.$$

In general, we see that the relevant operator is MM^* and not $M^2=({}^tR)^2$.

E. The notion of excess work

The notion of excess work has been introduced by Ross, Hunt, and Hunt[10] and we can give a meaning to it in our abstract setting.

We start from a stationary state $\tilde{p}(x)$ (and as usual the stochastic matrix, R). We can do two things.

(i) We force a variation of the state $\tilde{p}$ (by an external process) so that we have a displaced state $p(x)$,

$$p(x)=\tilde{p}(x)\exp(\delta\varphi_1(x)+\delta^2\varphi_2(x)+\cdots).$$

The relative cost in entropy for doing this is

$$S(p|\tilde{p})\sim-\frac{\delta^2}{2}\langle\varphi_1^2(x)\rangle_{\tilde{p}},$$

as we know from Eq. (2.5).

(ii) We start from a certain state q,

$$q(x)=\tilde{p}(x)\exp(\delta\psi_1(x)+\delta^2\psi_2(x)+\cdots),$$

and let it evolve in one time step Δt in such a way that the variation of free energy is exactly $\delta\varphi_1(x)$ (up to δ^2), so that we *want*

$$q(x,\Delta t)=\tilde{p}(x)\exp(\delta\psi_1(x)+\delta\varphi_1(x)+\cdots).$$

This implies

$$\tilde{p}(x)[\varphi_1(x)+\psi_1(x)]=\sum_y R_{xy}\tilde{p}(y)\psi_1(y). \tag{2.13}$$

The variation of entropy is then easily seen to be

$$S(q(\cdot,\Delta t)|\tilde{p})-S(q(\cdot)|\tilde{p})=-\frac{\delta^2}{2}(\langle\varphi_1^2\rangle_{\tilde{p}}+2(\psi_1|\varphi_1)_{\tilde{p}}),$$

with ψ_1 fixed by Eq. (2.13), or

$$\left[I-\operatorname{diag}\left(\frac{1}{\tilde{p}}\right)R\ \operatorname{diag}\tilde{p}\right]\psi_1\equiv-\varphi_1.$$

Alternatively,

$$\operatorname{diag}\tilde{p}(\varphi_1+\varphi_1)=R(\operatorname{diag}\tilde{p})\psi_1=-R(\operatorname{diag}\tilde{p})\left[I-\operatorname{diag}\left(\frac{1}{\tilde{p}}\right)R\ \operatorname{diag}\tilde{p}\right]^{-1}\varphi_1.$$

The excess work W_{exc} is given by (see Ref. 10)

$$\begin{aligned}
W_{\mathrm{exc}}&=S(q(\cdot,\Delta t)|\tilde{p})-S(q(\cdot)|\tilde{p})+S(p|\tilde{p})\\
&=-\delta^2\sum_x\tilde{p}(x)\varphi_1(x)(\varphi_1(x)+\psi_1(x))\\
&=-\delta^2\sum\varphi_1(x)(R(\operatorname{diag}\tilde{p})\psi_1)(x)\\
&=\delta^2\sum_x\left(\left[I-\left(\operatorname{diag}\frac{1}{\tilde{p}}\right)R(\operatorname{diag}\tilde{p})\right]\psi_1\right)_x((R\ \operatorname{diag}\tilde{p})\psi_1)_x\\
&=\delta^2\sum_x\tilde{p}(x)\psi_1(x)\left[({}^tR\psi_1)_x-\left({}^tR\ \operatorname{diag}\frac{1}{\tilde{p}}R\ \operatorname{diag}\tilde{p}\psi_1\right)_x\right]\\
&=\delta^2[(\psi_1|M\psi_1)-(\psi_1|MM^*\psi_1)],
\end{aligned}$$

where $M={}^tR$, $M^*=\operatorname{diag}(1/\tilde{p})R(\operatorname{diag}\tilde{p})$, and the scalar product has the weight $\tilde{p}$ as usual. One can rewrite this as

$$W_{\mathrm{exc}}=\delta^2\left[\left(\psi_1\left|\frac{M+M^*}{2}\psi_1\right.\right)-(\psi_1|MM^*\psi_1)\right]. \tag{2.14}$$

Notice that if R *satisfies detailed balance*, we have seen that

$$M^*=M={}^tR,$$

and so, because ${}^tR-({}^tR)^2\geqslant 0$, because ${}^tR$ has eigenvalue less than or equal to 1, we have

$$W_{\mathrm{exc}}=\delta^2[(\psi_1|({}^tR-({}^tR)^2)\psi_1)]\geqslant 0,$$

which is exactly Ross' result in our abstract context.

Remark: Ross *et al.* derive this result for the stochastic matrix corresponding to a master equation of a linear chemical system, in which case it is known that the master equation satisfies detailed balance. This is not the case for nonlinear chemical reactions. But we still have an expression for the excess work, in general.

Remark: For a stochastic matrix S that is self-adjoint with respect to a scalar product, it is clear that

$$(\psi|S\psi)\geqslant(\psi|S^2\psi).$$

Moreover, to prove that the excess work is positive, we would have to prove that $M+M^*-2MM^*$ is a positive matrix self-adjoint with respect to the scalar product $(\ |\ )_{\bar{p}}$.

III. PATH ENTROPIES AND DISSIPATION TO MAINTAIN THE STATIONARY STATE

To maintain a system in a nonequilibrium state against an equilibrated environment, it is necessary to dissipate energy. We introduce measures for the rate of dissipation of free energy.

A. Absolute path entropy

(a) Measure on a space of paths. Let X be our usual state space. A path up to time $T(T=n\ \Delta t)$ is a sequence $\gamma=\{x_0,x_1,\ldots,x_T\}$ of points in X. A path is then a sequence of transformations. For example, a Carnot cycle or a biochemical cycle will be realized by closed paths.

If p is an initial distribution, we define a probability measure $\mu^{(R,p)}(\gamma)$ on the space of paths by the formula

$$\mu^{(R,p)}(\gamma)=R_{x_Tx_{T-1}}R_{x_{T-1}x_{T-2}}\cdots R_{x_1x_0}p(x_0). \tag{3.1}$$

(b) We define an absolute entropy as

$$\sigma(T|R,p)=-\sum_{\gamma=\text{path up to } T}\mu^{(R,p)}(\gamma)\log\,\mu^{(R,p)}(\gamma)$$

$$=-\sum_{\gamma}\,\mu^{(R,p)}(\gamma)\left[\sum_{n=0}^{T-1}\,\log R_{x_{n+1}x_n}+\log\,p(x_0)\right].$$

As usual, we define recursively

$$(R^kp)(x)=\sum_y R_{xy}(R^{k-1}p)(y).$$

Then

$$\sum_{\gamma}\,\mu^{(R,p)}(\gamma)\log R_{x_{n+1}x_n}=\sum R_{x_Tx_{T-1}}\cdots R_{x_{n+1}x_n}\log R_{x_{n+1}x_n}(R^np)(x_n)$$

$$=\sum_{x_{n+1},x_n}R_{x_{n+1}x_n}\log R_{x_{n+1}x_n}(R^np)(x_n).$$

But

$$\sum_{x_{n+1}}R_{x_{n+1}x_n}(R^np)(x_n)=1.$$

Because $L(\xi)=\xi\log\xi$ is convex, we have

$$\sum_{x_{n+1},x_n} R_{x_{n+1}x_n}(R^n p)(x_n)\log R_{x_{n+1}x_n}$$

$$\geqslant \sum_{x_{n+1}} \left(\sum_{x_n} R_{x_{n+1}x_n}(R^n p)(x_n)\right)\log\left(\sum_{x_n} R_{x_{n+1}x_n}(R^n p)(x_n)\right)$$

$$= \sum_{x_{n+1}} (R^{n+1}p)(x_{n+1})\log(R^{n+1}p)(x_{n+1})$$

$$\equiv -S(R^{n+1}p) \quad \text{(absolute entropy of } R^{n+1}p\text{)}.$$

Finally,

$$\sigma(T|R,p)\leqslant \sum_{n=0}^{T} S(R^n p).$$

In particular, if $p=\tilde{p}$,

$$\sigma(T|R,\tilde{p})\leqslant (T+1)S(\tilde{p}).$$

B. Relative entropy on paths

The preceding concept involved the transition matrix R alone and, as such, did not measure the cost of the process R itself. Now, to quantify the extra dissipation needed to maintain a nonequilibrium state in a larger environment, we represent the action of this environment on our system by a stochastic matrix W satisfying the detailed balance. Namely, under the influence of W, the system X relaxes to an equilibrium state p_{eq} and we assume that for all x,y,

$$W_{yx}p_{\text{eq}}(x)=W_{xy}p_{\text{eq}}(y).$$

On the path space of X, we can consider the measure $\mu^{(R,p)}$, as well as the measures $\mu^{(W,q)}$, $\mu^{(W,p_{\text{eq}})}$.

The extra dissipation needed to maintain the R-dynamics in the larger equilibrium environment where action on the system is given by W is represented by the relative path entropy:

$$\mathscr{S}(T|(R,p),(W,q))=-\sum_{\text{path to } T} \mu^{(R,p)}(\gamma)\log\frac{\mu^{(R,p)}(\gamma)}{\mu^{(W,q)}(\gamma)}. \tag{3.2}$$

This can be rewritten using our basic quantity, $S(p|q)$, the relative entropy of Eq. (2.1),

$$\mathscr{S}(T|(R,p),(W,q))=S(p|q)-\sum_{\gamma=\text{path to } T} \mu^{(R,p)}(\gamma)\log\frac{R_{x_T x_{T-1}}\cdots R_{x_1 x_0}}{W_{x_T x_{T-1}}\cdots W_{x_1 x_0}}.$$

Analysis of $\mathscr{S}$. In the preceding equation, the second term is

$$\sum_{\gamma}\sum_{n=0}^{T-1} \mu^{(R,p)}(\gamma)\log\frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}}.$$

Now consider

$$\sum_{\gamma} \mu^{(R,p)}(\gamma)\log\frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}} = \sum_{x_{n+1},x_n} (R^n p)(x_n) R_{x_{n+1}x_n} \log\frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}}$$

$$= \sum_{x_n} (R^n p)(x_n) \sum_{x_{n+1}} W_{x_{n+1}x_n} \frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}} \log\frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}}. \tag{3.3}$$

Using the convexity of $\xi\log\xi$, this is

$$\geqslant \sum_{x_n} (R^n p)(x_n) \left(\sum_{x_{n+1}} W_{x_{n+1}x_n} \frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}} \right) \left(\log\left(\sum_{x_{n+1}} W_{x_{n+1}x_n} \frac{R_{x_{n+1}x_n}}{W_{x_{n+1}x_n}} \right) \right),$$

and so, this is positive or zero because $\Sigma_{x_{n+1}} R_{x_{n+1}x_n} = 1$. Let us define

$$\Delta_1 \mathscr{S}(R,q,W) = -\sum_{x,y} R_{xy} q(y) \log\frac{R_{xy}}{W_{x,y}}. \tag{3.4}$$

Then, from Eq. (3.3),

$$\mathscr{S}(T|(R,p),(W,q)) = S(p|q) + \sum_{t=0}^{T-1} \Delta_1 \mathscr{S}(R,R^t p,W), \tag{3.5}$$

and we have proved above that

$$\Delta_1 \mathscr{S}(R,q,W) \leqslant 0. \tag{3.6}$$

In particular,

$$\mathscr{S}(T|(R,p),(W,q)) \leqslant 0. \tag{3.7}$$

Note that *this is also decreasing with T*, i.e., *each increment* $\Delta_1 \mathscr{S}(R,R^t p,W) \leqslant 0$.

A Particular case: Take $p = \tilde{p}$, the stationary state, so that $R^t \tilde{p} = \tilde{p}$ for all t. Then $S(T|(R,\tilde{p}),(W,q)) = S(\tilde{p}|q) + T\,\Delta_1 S(R,\tilde{p},W)$. In this case, the fundamental quantity,

$$\Delta_1 \mathscr{S}(R,\tilde{p},W) = -\sum_{xy} R_{xy} \tilde{p}(y) \log\frac{R_{xy}}{W_{xy}} \tag{3.8}$$

is the *rate of dissipation per unit time step to maintain the stationary state* $\tilde{p}$ *(in the R-dynamics) against the W dynamics.*

We introduce

$$R_{xy} = W_{xy} e^{f_{xy}}. \tag{3.9}$$

Then

$$\Delta_1 \mathscr{S}(R,\tilde{p},W) = -\sum_{xy} R_{xy} \tilde{p}(y) f_{xy}. \tag{3.10}$$

Remark: If $f_{xy} = \varphi_x - \varphi_y$ then it is immediate that $\Delta_1 S(R,\tilde{p},W) = 0$. But in this case we would have

$$W_{xy} e^{\varphi_y} \tilde{p}(y) = \tilde{p}(x),$$

so that $e^{\varphi_y}\tilde{p}(y)$ would be the equilibrium state $p_{\rm eq}$ of W. But also $\Sigma_x R_{xy}=1$, so that $\Sigma_x W_{xy}e^{-\varphi_x} = e^{-\varphi_y}$. This implies that $\{e^{-\varphi_x}\}$ would be the left eigenvector of eigenvalue 1 of W and (in the absence of degeneracy) this would be a constant. Then $\tilde{p}$ would just be the equilibrium $p_{\rm eq}$ and $R=W$.

C. The minimal rate of dissipation of a given state

In this section, we start with a given $\tilde{p}$ (in a detailed balance dynamics W) and we want to build an R-dynamics for which $\tilde{p}$ is stationary but that minimizes the dissipation of energy with respect to an underlying detailed balance dynamics W. The rate of dissipation is $\Delta_1\mathscr{S}(R,\tilde{p},W)$. We define, as in Eq. (3.9),

$$R_{xy}=W_{xy}e^{f_{xy}},$$

and we want to minimize

$$|\Delta_1\mathscr{S}(R,\tilde{p},W)|=\sum_{xy} W_{xy}e^{f_{xy}}\tilde{p}(y)f_{xy},$$

subject to the conditions

(i) $\Sigma_x W_{xy}e^{f_{xy}} = 1$, for all y;

(ii) R preserves the stationary state $\tilde{p}$, or $\Sigma_y W_{xy}e^{f_{xy}}\tilde{p}(y) = \tilde{p}(x)$ for all x. We examine the effect of a variation δf_{xy} on $|\Delta_1\mathscr{S}(R,\tilde{p},W)|$,

$$\delta|\Delta_1\mathscr{S}(R,\tilde{p},W)|=\sum_{yx} W_{yx}\tilde{p}(x)\exp(f_{yx})(1+f_{yx})\,\delta f_{yx}. \tag{3.11}$$

The variations of the two constraints are

$$\delta\left(\sum_y W_{yx}\exp(f_{yx})-1\right)=\sum_y W_{yx}\exp(f_{yx})\,\delta f_{yx}, \tag{3.12}$$

$$\delta\left(\sum_y W_{yx}\exp(f_{yx})\tilde{p}(y)\right)=\sum_x W_{yx}\exp(f_{yx})\tilde{p}(x)\,\delta f_{yx}. \tag{3.13}$$

As usual, we introduce two Lagrange multipliers, λ_x for the constraints (3.12) and μ_y for the constraints (3.13), and write

$$0=\delta|\Delta_1\mathscr{S}(R,\tilde{p},W)|+\sum_x \lambda_x\delta\left(\sum_y W_{yx}\exp(f_{yx})-1\right)$$
$$+\sum_y \mu_y\delta\left(\sum_x W_{yx}\exp(f_{yx})\tilde{p}(x)-\tilde{p}(y)\right).$$

After rearrangement, this gives

$$0=\sum_{yx}\delta f_{yx}\exp(f_{yx})W_{yx}[\tilde{p}(x)(1+f_{yx})+\lambda_x+\mu_y\tilde{p}(x)].$$

If $W_{yx}=0$, we have no condition. But, if $W_{yx}\neq 0$ we obtain

$$f_{yx}=-\frac{\lambda_x}{\tilde{p}(x)}-\mu_y-1,$$

and then λ_x and μ_y are determined using the two constraints:

$$1=\sum_y W_{yx}\exp\left(-\frac{\lambda_x}{\tilde{p}(x)}\right)\exp(-\mu_y-1),$$

$$\tilde{p}(y)=\sum_x W_{yx}\exp\left(-\frac{\lambda_x}{\tilde{p}(x)}\right)\exp(-\mu_y-1)\tilde{p}(x).$$

Then, we see that $R_{yx}=0$ when $W_{yx}=0$ (this is our ansatz anyway), and

$$R_{yx}=W_{yx}\exp(\rho_y-\sigma_x), \tag{3.14}$$

when $W_{yx}\neq 0$, so that we can always use Eq. (3.14). Using (3.14), we can compute the rate of dissipation

$$|\Delta_1\mathcal{S}(R,\tilde{p},W)|=\sum_{y,x} W_{yx}\exp(f_{yx})f_{yx}\tilde{p}(x).$$

But

$$\sum_x W_{yx}e^{-\sigma_x}\tilde{p}(x)=e^{-\rho_y}\tilde{p}(y),\quad \sum_y W_{yx}e^{\rho_y}=e^{\sigma_x},$$

so that finally the minimal rate of dissipation is

$$|\Delta_1\mathcal{S}(R,\tilde{p},W)|=\sum_x \tilde{p}(x)(\rho_x-\sigma_x). \tag{3.15}$$

D. Analysis near equilibrium

In this section, we consider an equilibrium state p_{eq} with its detailed balance dynamics given by the matrix W. We further consider a nonequilibrium state $\tilde{p}$, which is close to p_{eq}, whose dynamics are given by a (non-detailed-balance) stochastic matrix R, close to W. We fix the notation as follows:

$$\tilde{p}(x)=p_{\mathrm{eq}}(x)\exp(\varphi(x,\delta))\equiv p_{\mathrm{eq}}(x)\exp(\delta\varphi_1(x)+\delta^2\varphi_2(x)+\cdots), \tag{3.16}$$

$$R_{xy}=W_{xy}\exp(f(x,y,\delta))\equiv W_{xy}\exp(\delta f_{xy}^{(1)}+\delta^2 f_{xy}^{(2)}+\cdots). \tag{3.17}$$

1. Identities satisfied by $f^{(1)}$ and φ_1

We differentiate the relation $\Sigma_y R_{yx}=1$ with respect to δ at $\delta=0$. This gives

$$\sum_y W_{yx}f_{yx}^{(1)}=0. \tag{3.18}$$

Similarly, the relation $\tilde{p}(x)=\Sigma_y R_{xy}\tilde{p}(y)$ is differentiated with respect to δ at $\delta=0$. This implies

$$p_{\mathrm{eq}}(x)\varphi_1(x)=\sum_y W_{xy}f_{yx}^{(1)}p_{\mathrm{eq}}(y)+\sum_y W_{xy}\varphi_1(y)p_{\mathrm{eq}}(y), \tag{3.19}$$

or using detailed balance,

$$W_{xy}p_{\rm eq}(y)=W_{yx}p_{\rm eq}(x),$$

$$\varphi_1(x)=\sum_y W_{yx}f^{(1)}_{yx}+\sum_y W_{yz}\varphi_1(y). \tag{3.20}$$

2. Variation of the entropy near the equilibrium state

Suppose that we start from the stationary state $\tilde{p}(x)$ and let it evolve spontaneously using the detailed balance dynamics W. We can compute in one time step Δt, the variation of relative entropy as in Sec. II D, except now $\tilde{p}\to p_{\rm eq}$ and $R\to W$. We obtain

$$S(\tilde{p}(\cdot,\Delta t)|p_{\rm eq})-S(\tilde{p}|p_{\rm eq})=\frac{\delta^2}{2}\left[\langle\varphi_1^2\rangle_{p_{\rm eq}}-\left\langle\left(\frac{1}{\text{diag } p_{\rm eq}}W\ \text{diag}\ p_{\rm eq}\varphi_1\right)^2\right\rangle_{p_{\rm eq}}\right].$$

But for detailed balance we have $((1/\text{diag}\, p_{\rm eq})W\ \text{diag}\, p_{\rm eq})_{xy}=(1/p_{\rm eq}(x))W_{xy}p_{\rm eq}(y)=W_{yx}$. Thus

$$S(\tilde{p}(\cdot,\Delta t)|p_{\rm eq})-S(\tilde{p}(\cdot)\cdot|p_{\rm eq})=\frac{\delta^2}{2}\left[\langle\varphi_1^2\rangle_{p_{\rm eq}}-\langle({}^tW\varphi_1)^2\rangle_{p_{\rm eq}}\right]. \tag{3.21}$$

3. Rate of dissipation

We now compute the rate of dissipation $|\Delta_1\mathcal{I}(\mathrm{R},\tilde{\mathrm{p}},\mathrm{W})|$. In Sec. III C, we obtained

$$|\Delta_1\mathcal{I}(R,\tilde{p},W)|=\sum_{xy}\ \exp(f(x,y,\delta))\tilde{p}(y)f(x,y,\delta),$$

which is evidently 0 for $\delta=0$ because $f(x,y,0)=0$. We expand this quantity in powers of δ up to second order. We write

$$\tilde{p}(x)=p_{\rm eq}(x)+\delta p_1(x)+O(\delta^2).$$

(We will not need the second order term in $\tilde{p}$.)

$$|\Delta_1\mathcal{I}(R,\tilde{p},W)|=\sum_{xy}W_{xy}\left\{1+\delta f^{(1)}_{xy}+\delta^2 f^{(2)}_{xy}+\frac{\delta^2}{2}(f^{(1)}_{xy})^2\right\}$$
$$\times\{p_{\rm eq}(y)+\delta p_1(y)\}\{\delta f^{(1)}_{xy}+\delta^2 f^2_{xy}\}+O(\delta^3).$$

The first-order term in δ is

$$\sum_y p_{\rm eq}(y)\sum_x W_{xy}f^{(1)}_{xy}.$$

But this is 0 because of relation (3.15) above,

$$\sum_x W_{xy}f^{(1)}_{xy}=0.$$

So we compute the second-order terms in δ; these are

$$\sum_{x,y} W_{xy} p_{\mathrm{eq}}(y)(f_{xy}^{(2)}+(f_{xy}^{(1)})^2)+\sum_{x,y} W_{xy} f_{xy}^{(1)} p_1(y).$$

But expanding $\Sigma_x R_{xy}=1$ up to second order gives

$$\sum_{x} W_{xy} f_{xy}^{(1)}=0 \quad \text{[relation (3.18) above]},$$

$$\sum_{x} W_{xy}\left(f_{xy}^{(2)}+\frac{1}{2}(f_{xy}^{(1)})^2\right)=0.$$

There remains only a single term, so that

$$|\Delta_1 \mathscr{S}(R,\tilde{p},W)|=\frac{\delta^2}{2}\sum_{x,y} W_{xy} p_{\mathrm{eq}}(y)(f_{xy}^{(1)})^2. \tag{3.22}$$

4. Minimal rate of dissipation

Starting from this last equation let us compute the minimal rate of dissipation, given $\tilde{p}$ and W. We thus have to minimize $|\Delta_1 \mathscr{S}(R,\tilde{p},W)|$ with respect to $f_{xy}^{(1)}$ under the conditions (3.18) and (3.20).

Introducing Lagrange multipliers λ_x, μ_y for (3.18) and (3.20), we see that

$$\sum_{x,y} W_{xy} p_{\mathrm{eq}}(y) f_{xy}^{(1)}\,(\delta f_{xy}^{(1)})+\sum_{x}\lambda_x\sum_{y} W_{yx}\,\delta f_{yx}^{(1)}+\sum_{x}\mu_x\sum_{y} W_{yx}\,\delta f_{xy}^{(1)}=0.$$

This implies that

$$W_{xy} p_{\mathrm{eq}}(y) f_{xy}^{(1)}+W_{xy}\lambda_y+\mu_x W_{yx}=0.$$

Using detailed balance, $W_{yx}=W_{xy}p_{\mathrm{eq}}(y)/p_{\mathrm{eq}}(x)$, we see that either

$$(1)\quad W_{xy}=0, \quad \text{or}$$

$$(2)\quad f_{xy}^{(1)}+\frac{\lambda_y}{p_{\mathrm{eq}}(y)}+\frac{\mu_x}{p_{\mathrm{eq}}(x)}=0,$$

which is exactly what we obtained above in a more general setting, namely

$$f_{xy}^{(1)}=\rho(x)-\sigma(y).$$

Let us assume that $f_{xy}^{(1)}$ has this form and determine σ and ρ using the constraints (3.18) and (3.20) above. This gives the following.

(1) For the constraint (3.18), $\Sigma_x W_{xy} f_{xy}^{(1)}=0$,

$$\sigma(x)=\sum_{y}\rho(y)W_{yx} \quad \text{or} \quad \sigma=\rho^t W. \tag{3.23}$$

(2) For the constraint (3.20), $\sigma_1(x)=\Sigma_y W_{yx} f_{xy}^{(1)}+\Sigma_y W_{yx}\varphi_1(y)$,

$$\varphi_1(x)=\sum_y W_{yx}(\rho(x)-\sigma(y))+\sum_y W_{yx}\varphi_1(y)$$

$$=\rho(x)-\sum_y \sigma(y)W_{yx}+\sum_y W_{yx}\varphi_1(y)$$

$$=(\rho(I-{}^tW)(I+{}^tW))_x+\sum_y W_{yx}\varphi_1(y).$$

This can be solved as

$$\varphi_1(I-{}^tW)=\rho(I+{}^tW)(I-{}^tW)$$

or

$$\varphi_1=\rho(I+{}^tW). \tag{3.24}$$

Now we can compute the minimal rate of dissipation (to maintain $\tilde{p}$ against the thermal detailed balance dynamics given by W),

$$|\Delta_1\mathscr{S}(R,\tilde{p},W)|=\frac{\delta^2}{2}\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)(f_{xy}^{(1)})^2=\frac{\delta^2}{2}\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)(\rho(x)-\sigma(y))^2.$$

Let us expand, using (3.23), $\sigma=\rho^tW$,

$$\sum_{x,y} W_{xy}(y)(\rho(x)-\sigma(y))^2$$

$$=\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)\rho(x)^2-2\sum_{x,yp} p_{\mathrm{eq}}(y)\rho(x)\sigma(y)+\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)\sigma(y)^2.$$

We consider the three terms above,

$$\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)\rho(x)^2=\sum_x p_{\mathrm{eq}}(x)\rho(x)^2-2\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)\rho(x)\sigma(y)$$

$$=-2\sum_y (\rho^tW)_y p_{\mathrm{eq}}(y)(\rho^tW)_y,$$

$$\sum_{x,y} W_{xy}p_{\mathrm{eq}}(y)\sigma(y)^2=\sum_y p_{\mathrm{eq}}(y)\sigma(y)^2=\sum_y p_{\mathrm{eq}}(y)((\rho^tW)_y),$$

so that

$$|\Delta_1\mathscr{S}(R,\tilde{p},W)|=\frac{\delta^2}{2}(\|\rho\|_{p_{\mathrm{eq}}}^2-\|\rho^tW\|_{p_{\mathrm{eq}}}^2). \tag{3.25}$$

5. Comparison of the rate of dissipation and the variation of S

We come back to relation (3.21), namely

$$S(\tilde{p}(\cdot,\Delta t)|p_{\mathrm{eq}})-S(\tilde{p}|p_{\mathrm{eq}})=\frac{\delta^2}{2}[\|\varphi_1\|_{p_{\mathrm{eq}}}^2-\|\varphi_1{}^tW\|_{p_{\mathrm{eq}}}^2]. \tag{3.26}$$

We see that this is similar to the relation (3.25) for the minimal rate of dissipation in the R-dynamics, except for the fact that $\varphi_1=\rho(I+{}^tW)$. [See the relation (3.24).]

We know that $\Sigma p_{\rm eq}(x)\varphi_1(x)=0$ so that φ_1 can be expanded in terms of the left eigenvectors of W with eigenvalues different from 1. Because of the (3.24), the same expansion is possible for ρ, and we therefore write

$$\rho=\sum c_n\theta_n^{(L)},$$

where $\theta_n^{(L)}$ are the left eigenvectors of W of eigenvalue less than 1.

Moreover, $\{\theta_n^{(L)}\}$ form an orthonormal basis for the equilibrium scalar product $p_{\rm eq}$ (because ${}^tW$ is self-adjoint for this scalar product due to detailed balance). It follows that

$$S(\tilde{p}(\cdot,\Delta t)|p_{\rm eq})-S(\tilde{p}(\cdot)|p_{\rm eq})=\frac{\delta^2}{2}\sum_n |c_n|^2|(1+\lambda_n)|^2(1-|\lambda_n^2|)$$

and

$$|\Delta_1\mathscr{S}(R,\tilde{p},W)|=\frac{\delta^2}{2}\sum_x |c_n|^2(1-|\lambda_n^2|).$$

Because $0\leqslant|\lambda_n|\leqslant 1$, we see that we always have

$$\tfrac{1}{4}[S(\tilde{p}(\cdot,\Delta t)|p_{\rm eq})-S(\tilde{p}(\cdot)|p_{\rm eq})]\leqslant|\Delta_1\mathscr{S}(R,\tilde{p},W)|. \tag{3.27}$$

The interpretation of this inequality is clear. If we start with the stationary state $\tilde{p}$ and switch off the R dynamics (so that $\tilde{p}$ starts to evolve by the detailed balance dynamics toward $p_{\rm eq}$), the dissipation is less than four times the cost to maintain $\tilde{p}$ using the dynamics in the larger environment (acting with W) on the system.

IV. FLUCTUATION AND DISSIPATION FOR SLOW VARIABLES

A. The macroscopic entropy

1. Fast and slow variables

Usually, a system with state space X is characterized by a small number of "slow" variables and by other "fast" variables. Such variables can be distinguished in terms of the eigenvalues of the master equation, i.e., in our case, in terms of the eigenvalues of the stochastic matrix R. Essentially, the slow variables are functions f on X, such that their set of values $\{f_x\}$ $(x\in X)$ can be decomposed on left eigenvectors of the matrix R associated with eigenvalues of R very close to 1 (but not equal to 1).

In this section, we shall assume that the system is characterized by only one slow variable $A_x(x\in X)$ taking values $a,a',\ldots$. We shall denote by u the other coordinates, so that a point x in X is identified with a couple (a,u), with $a=A(x)$.

2. Reduced description

At this point, it is customary to describe the system by the variable A alone. This is the reduced or macroscopic description, which is a coarse grained description of the full description by (a,u). We also must change the time scale, because in the time scale Δt (of the R dynamics), A does not evolve in an appreciable manner. So the relevant time scale becomes much longer, and the general idea is that, relative to this longer time scale, A varies but u readjusts itself instantaneously to its relative stationary distribution. This is the idea of all macroscopic descriptions (see Refs. 1, 6, and 7, among many references).

In our context, we can make this precise in the following way. We start as usual from a stationary state $\tilde{p}(x)$ and we define a reduced (or macroscopic) state $\tilde{P}$ by

$$\tilde{P}(x)=\sum_{\{x\in X|A(x)=a\}}\tilde{p}(x)=\sum_{u}\tilde{p}(x,u). \tag{4.1}$$

Following customary practice, we define a function $\Sigma(a)$ by the formula

$$\tilde{P}(a)=\exp(-\Sigma(a)) \tag{4.2}$$

(see, for example, Ref. 1, in the context of an equilibrium situation, and Refs. 2, 6, 10, and 24 for generalizations to nonequilibrium situations). Here $\Sigma(a)$ is a Lyapunov function for the reduced evolution of the A variable, as we shall see below.

In the case of equilibrium, $\Sigma(a)$ is the Einstein entropy. We shall assume now that the average of A in the stationary state is 0.

$$\langle A\rangle_{\tilde{p}}\equiv\sum A(x)\tilde{p}(x)=0. \tag{4.3}$$

3. Relation to the relative entropy

Let us assume that we have prepared the system in the state $\tilde{p}(x)$ (the stationary state for the R-dynamics), but that we observe in a particular sample of the system a certain fluctuation of A, so that A takes a value $a\neq 0$. Then, the probability distribution of the fast variables u, given the fact one observes the fluctuation a of A, is the conditional stationary probability distribution

$$\tilde{\pi}_a(u)=\frac{\tilde{p}(a,u)}{\tilde{P}(a)}, \tag{4.4}$$

and the quasistationary state is thus a state q_a,

$$q_a(x)\equiv\tilde{\pi}_a(u)\,\delta(A(x)-a). \tag{4.5}$$

It follows immediately that the relative entropy of q_a with respect to $\tilde{p}$, namely $S(q_a|\tilde{p})$ [see Eq. (1.1)], is in fact, $-\Sigma(a)$,

$$S(q_a|\tilde{p})=-\Sigma(a), \tag{4.6}$$

where $\Sigma(a)$ is defined as in Eq. (4.2). We calculate this as follows.

$$S(q_a|\tilde{p})=-\sum_{a}\tilde{\pi}_a(u)\,\delta(A(x)-a)\log\frac{\tilde{\pi}_a(u)}{\tilde{p}(a,u)}=-\log\frac{1}{\tilde{P}(a)}=-\Sigma(a).$$

This explains why $\Sigma(a)$ could be taken as a Lyapunov function for the evolution of a. If we wait an appropriate time, a would vary by a small quantity δa, while the fast u variables would recover their conditional stationary distribution. Our earlier assumption on time scales is precisely the assumption that such an appropriate time exists. Then the state q_a would become $q_{a+\delta a}$ by the R evolution, the variables u keeping their conditional stationary distribution. Under this circumstance,

$$S(q_{a+\delta a}|\tilde{p})-S(q_a|\tilde{p})\geq 0,$$

since $q_{a+\delta a}\approx Rq_a$ [cf. Eq. (2.4)]. This implies

$$\Sigma(a+\delta a)\leqslant\Sigma(a).$$

B. Fluctuation dissipation, in general

The usual near-equilibrium fluctuation dissipation theory is a formal consequence of the fact that a certain state $p_{\rm eq}$ is a stationary state of the W evolution: Writing the stationarity of the equilibrium state explicitly leads to an identity that can then be reinterpreted as a physical relation between fluctuation and dissipation (see Refs. 1, 6, 12, and 25 among many references). In the language of the previous section, if we are in a stationary state, and if we observe an actual fluctuation of A equal to a [this fluctuation has a probability $\tilde{P}(a)$], then the dissipation induced by the reduction to 0 of this fluctuation is related in a natural way to this fluctuation (in a linear way), the proportionality coefficient being some given "transport" characteristic of the system (see Ref. 23).

We will show that it is possible to derive the fluctuation-dissipation theory in our context using the formalism of Sec. IV A. Moreover, our demonstration is not limited to near-equilibrium situations (and gives, in fact, a correction to it, as well as a finite-size effect correction).

There are various ways to derive fluctuation-dissipation theorems, each of which provides, in general, extra information, in particular about the transport or relaxation coefficients. Here we shall relate the "transport" coefficient to the eigenvalues of R. In our abstract context, we shall choose an analog of linear response theory (as presented, for example, in Ref. 25). We produce the fluctuation of the macroscopic variable A using an external force that modifies the stationary state $\tilde{p}$.

C. Linear response theory: General computation

The general situation is as in Sec. IV A: we distinguish a variable $A(x)$ $(x\in X)$ and other variables u, so that $x=(a,u)$, where $a=A(x)$. Moreover, we have the stationary state $\tilde{p}(x)$ with respect to the R dynamics as usual. We finally assume that A has average 0 in the stationary state [Eq. (4.3)].

1. The displaced state

We define, in analogy with the analysis of Sec. II B, a displaced state,

$$p_\alpha(x)=\frac{1}{Z_\alpha}\tilde{p}(x)\exp(\alpha A(x)+\cdots), \tag{4.7}$$

where α is a small parameter and the ellipses represents higher-order terms in α. Then α can be viewed as a "conjugate field," αA being an extra energy (this field is imposed by an external source or observer on which the system does not react).

We have, because (by assumption) $\langle A(\,\cdot\,)\rangle_{\tilde{p}}=0$,

$$Z_\alpha=\sum_x \tilde{p}(x)\exp(\alpha A(x)+\cdots)=1+\frac{\alpha^2}{2}\langle A(x)^2\rangle_{\tilde{p}}+\cdots,$$

so that one can suppress Z_α as being higher order in α, and write simply

$$p_\alpha(x)=\tilde{p}(x)\exp(\alpha A(x)+\cdots). \tag{4.8}$$

Now, we have

$$\langle A\rangle_{p_\alpha}=\langle A\rangle_{\tilde{p}}+\alpha\langle A^2\rangle_{\tilde{p}}+\cdots,$$

so that for small α,

$$\langle A\rangle_{p_\alpha}=\alpha\langle A^2\rangle_{\tilde{p}}, \tag{4.9}$$

and $\langle A^2\rangle_{\tilde{p}}$ appears as usual to be the susceptibility for A.

2. Variation of the mean value

We start at time $t=0$ from the state $p_\alpha(x)$ given by (4.1). In one time step, the state becomes

$$p_\alpha(x,\Delta t)=\sum_y R_{xy}p_\alpha(y)=\sum_y R_{xy}\tilde{p}(y)\exp(\alpha A(y)),$$

and at time Δt,

$$\langle A(\Delta t)\rangle_{p_\alpha}=\sum_x A(x)p_\alpha(x,\Delta t).$$

Expanding

$$\langle A(\Delta t)\rangle_{p_\alpha}=\sum_{x,y} A(x)R_{xy}\tilde{p}(y)+\alpha\sum_{x,y} A(x)R_{xy}\tilde{p}(y)A(y)+\cdots .$$

The first term is 0 because this is $\Sigma_x A(x)\tilde{p}(x)$. Finally, we have, modulo terms of order α^2,

$$\langle A(\Delta t)-A(0)\rangle_{p_\alpha}=\alpha\sum_{x,y} A(x)(R_{xy}-\delta_{xy})A(y)\tilde{p}(y). \tag{4.10}$$

We can also eliminate α using Eq. (4.9) and get

$$\langle A(\Delta t)-A(0)\rangle_{p_\alpha}=\frac{\langle A\rangle_{p_\alpha}}{\langle A^2\rangle_{\tilde{p}}}\sum_{x,y} A(x)(R_{xy}-\delta_{xy})A(y)\tilde{p}(y). \tag{4.11}$$

3. Second moment

We now want to compute $\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}$

(a) Computation of $\langle A(\Delta t)^2\rangle_{p_\alpha}$. This is, modulo terms in α^2,

$$\langle A(\Delta t)^2\rangle_{p_\alpha}=\sum_{x,y} A(x)^2R_{xy}\tilde{p}(y)\exp(\alpha A(y))=\langle A^2\rangle_{\tilde{p}}+\alpha\sum_{x,y} A(x)^2R_{xy}\tilde{p}(y)A(y). \tag{4.12}$$

(b) Computation of $\langle A^2\rangle_{p_\alpha}$. Again up to order α^2,

$$\langle A(0)^2\rangle_{p_\alpha}=\langle A^2\rangle_{\tilde{p}}+\alpha\langle A^3\rangle_{\tilde{p}}. \tag{4.13}$$

(c) Computation of $\langle A(\Delta t)A(0)\rangle_{p_\alpha}$

$$\begin{aligned}\langle A(\Delta t)A(0)\rangle_{p_\alpha}&=\sum_{x,y} A(x)R_{xy}A(y)\tilde{p}(y)\exp(\alpha A(y))\\&=\sum_{x,y} A(x)R_{xy}A(y)\tilde{p}(y)+\alpha\sum_{x,y} A(x)R_{xy}A(y)^2\tilde{p}(y).\end{aligned} \tag{4.14}$$

(d) Computation of $\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}$ From Eqs. (4.12)–(4.14) we obtain, after rearrangement,

$$\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}=-2\sum_{x,y} A(x)(R_{xy}-\delta_{xy})A(y)\tilde{p}(y)+\alpha\sum_{x,y}[A(x)^2A(y)(R_{xy}\tilde{p}(y) -R_{yx}\tilde{p}(x))-\tilde{p}(y)A(x)A(y)^2(R_{xy}-\delta_{xy})]. \tag{4.15}$$

We see that this term is again of order Δt because $R_{xy}-\delta_{xy}$ is of order Δt and because we can rewrite

$$R_{xy}\tilde{p}(y)-R_{yx}\tilde{p}(x)=(R_{xy}-\delta_{xy})\tilde{p}(y)-(R_{yx}-\delta_{yx})\tilde{p}(x).$$

D. The case of a left eigenvector of *R*

We shall now take for A a slow variable of the system, i.e., a variable that decays in one of the slowest possible modes. One way to do this is to choose for A a left eigenvector of the transfer matrix R with eigenvalue λ near 1 (but not exactly 1). (In fact, for our purposes the essential point is that the eigenvalue associated with A satisfy $1>\lambda\gg|\lambda'|$ for all other eigenvalues λ'. The closeness of λ to 1 is not used significantly.)

1. First moment

We assume for all $y\in X$, that

$$\lambda A_y=\sum_x A_x R_{xy}.$$

From Eq. (4.11) we obtain to first order in α,

$$\langle A(\Delta t)-A(0)\rangle_{p_\alpha}=\langle \dot{A}(0)\rangle_{p_\alpha}(\lambda-1). \tag{4.16}$$

2. Second moment

We obtain from Eq. (4.15),

$$\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}=(\lambda-1)[-2\langle A^2\rangle_{\tilde{p}}-\alpha\langle A^3\rangle_{\tilde{p}}]+\alpha\sum_{x,y} A(x)^2A(y)(R_{xy}\tilde{p}(y)-R_{yx}\tilde{p}(x)). \tag{4.17}$$

3. The case of detailed balance

When R satisfies detailed balance, the second term on the right-hand side of Eq. (4.17) above vanishes identically and

$$\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}=(1-\lambda)[-2\langle A^2\rangle_{\tilde{p}}+\alpha\langle A^3\rangle_{\tilde{p}}]. \tag{4.18}$$

Moreover, in this case, we can compute the rate of dissipation in one unit time step Δt starting from the state p_α and using the R dynamics. This is

$$S(p_\alpha(\cdot,\Delta t)|\tilde{p})-S(p_\alpha|\tilde{p})=\frac{\alpha^2}{2}\langle A^2\rangle_{\tilde{p}}(1-\lambda)(1+\lambda). \tag{4.19}$$

We saw the same result in Sec. II. To see the correspondence, take $\varphi_1=A$ and notice that $M=M^*={}^tR$, so that the μ eigenvalue of MM^* corresponding to $\varphi_1=A$ is $\mu=\lambda^2$.

4. Fluctuation dissipation

We return to the general (non-detailed-balance) case. If A is decaying slowly, $\lambda \approx 1$, and

$$S(p_\alpha(\cdot,\Delta t)|\bar{p}) - S(p_\alpha|\bar{p}) \sim \langle A^2\rangle_{\bar{p}}(1-\lambda). \tag{4.20}$$

Moreover, let us compare Eqs. (4.16) and (4.17) and let us assume that $\alpha\langle A^3\rangle_{\bar{p}} \approx 0$. Then eliminating $\lambda - 1$ from both equations, we obtain

$$\langle (A(\Delta t) - A(0))^2\rangle_{p_\alpha} = 2\langle A^2\rangle_{\bar{p}} \left| \frac{\langle A(\Delta t) - A(0)\rangle_{p_\alpha}}{\langle A(0)\rangle_{p_\alpha}} \right|. \tag{4.21}$$

This relation is, in our context, the analog of the standard fluctuation-dissipation theorem.

Example: The prototype of fluctuation-dissipation theory is obtained for a Langevin particle (of mass $m=1$) with the equation of motion

$$dx = v\ dt, \quad dv = -fv\ dt + dB(t),$$

where $dB(t)$ is the white noise force,

$$\langle dB(t)dB(s)\rangle = 2D\,\delta(t-s).$$

In the sense of our development, the "fast variables" are the sources of the noise. The variables x and v are "macroscopic" and "slow." The Fokker Planck equation is

$$L = \frac{\partial}{\partial v}\left(D\frac{\partial}{\partial v} + fv\right) - v\frac{\partial}{\partial x}.$$

This corresponds to a reduced operator (projected onto the slow variables) description of $I-R$ for the discrete time case. The space X consists of the configuration space (x,v) of the Langevin particle as well as the degrees of freedom that give rise to the white noise (that the Fokker–Planck equation absorbs into the diffusion coefficient). Take for A the function v. Then

$$L^* v = \left(D\frac{\partial^2}{\partial v^2} - fv\frac{\partial}{\partial v} + v\frac{\partial}{\partial x}\right) v = -fv,$$

and v is an eigenfunction of L^* with eigenvalue $-f$ (but not necessarily of the full operator that includes the sources of the noise). Now let us consider, instead of a state p_α, a state $\delta(v-v_0)$. Then

$$\langle v(\Delta t) - v_0\rangle_{\delta(v-v_0)} = \int (v-v_0)p(\Delta t, v|v_0)dv.$$

But

$$\begin{aligned}\lim_{\Delta t\to 0}\frac{\partial}{\partial \Delta t}\langle v(\Delta t) - v_0\rangle_{\delta(v-v_0)} &= \lim_{\Delta t\to 0}\int (v-v_0)L_v p(\Delta t, v|v_0)dv \\ &= \int L_v^*(v-v_0)\,\delta(v-v_0)dv \\ &= -fv_0.\end{aligned}$$

In the same way,

$$\lim_{\Delta t\to 0}\frac{\partial}{\partial \Delta t}\langle (v(\Delta t)-v(0))^2\rangle_{\delta(v-v_0)}=\int L_v^*(v-v_0)^2\delta(v-v_0)dv_0=2D.$$

Then our statement of the fluctuation-dissipation theorem [Eq. (4.21)] becomes

$$\lim_{\Delta t\to 0}\frac{\partial}{\partial \Delta t}\langle (v(\Delta t)-v(0))^2\rangle_{\delta(v-v_0)}$$

$$=2\langle v^2\rangle_{p_{\rm eq}}\left|\lim_{\Delta t\to 0} v\left(\frac{(\partial/\partial\Delta t)\langle v(\Delta t)-v_0\rangle_{\delta(v-v_0)}}{\langle v\rangle_{\delta(v-v_0)}}\right)\right|,$$

or

$$2D=2k_{\rm B}Tf,$$

which is the usual Einstein relation for $m=1$. So the relation (4.21) is the generalization of the standard fluctuation-dissipation theory in our context.

Remark: For initial conditions far from the stationary state, dissipation may be dominated by the friction coefficients, "f," irrespective of the fluctuations. As such, this way of calculating heat production, etc., will not involve the fluctuations. However, the validity of these mean field calculations (and use of f to derive heat production) does not contradict the fluctuation-dissipation theorem because it is a far from stationary-state situation.

5. The general case

In fact, Eqs. (4.16) and (4.17), even in the detailed balance case, are more precise than the fluctuation-dissipation theorem, because the fluctuation-dissipation theorem (in its usual statement) is the relation (4.21), relating these two quantities. In our case, we have separately derived each of the moments $\langle A(\Delta t)-A(0)\rangle_{p_\alpha}$ (the dissipation) and $\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}$ (the fluctuation) *separately*, and related them to the spectrum of the transfer matrix.

Equation (4.16) [for $\langle A(\Delta t)-A(0)\rangle_{p_\alpha}$] is straightforward.

Equation (4.17) [for $\langle (A(\Delta t)-A(0))\rangle_{p_\alpha}$)] contains a correction Γ of cubic order in A and first order in α,

$$\Gamma\equiv\alpha\sum_{x,y}A(x)^2A(y)(R_{xy}\tilde{p}(y)-R_{yx}\tilde{p}(x)).$$

Notice, also, that if we are away from criticality, both Γ and $\langle A^3\rangle_{\tilde{p}}$ would be close to 0.

E. Summary

In a sense, the standard fluctuation-dissipation theorem is a tautology; namely, the assertion that the stationary state is a solution of the stationary equation (see Ref. 12). In our situation, we say more because we compute separately the fluctuation *and* the dissipation in terms of the spectrum of the transfer matrix and then deduce the relation between the fluctuation and dissipation by eliminating the eigenvalue of the transfer matrix. Still, at our level of abstraction, all these identities can only be tautological. (The physics enters in judging the suitability of the stochastic description and the time scale separation.)

We now summarize our results concerning the fluctuation-dissipation theorem. Let A be a slow variable of the system, so $AR=\lambda R$ for a λ close to 1; it follows that $\langle A\rangle_{\tilde{p}}=0$. Let

$$p_\alpha(x)=\tilde{p}(x)\exp(\alpha A(x)+O(\alpha^2)), \tag{4.22}$$

be a neighboring state of the stationary state $\tilde{p}$, so that

$$\langle A\rangle_{p_\alpha}=\alpha\langle A^2\rangle_{\tilde{p}}+O(\alpha^2). \tag{4.23}$$

Then we have the two relations [see Eqs. (4.16) and (4.17)]:
dissipation,

$$\langle A(\Delta t)-A(0)\rangle_{p_\alpha}=\langle A\rangle_{p_\alpha}(\lambda-1); \tag{4.24}$$

fluctuation,

$$\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}=-2(\lambda-1)\langle A^2\rangle_{\tilde{p}}+O(\alpha A^3). \tag{4.25}$$

We thus obtain
fluctuation dissipation:

$$\langle (A(\Delta t)-A(0))^2\rangle_{p_\alpha}=\frac{2(A^2)_{\tilde{p}}}{\langle A\rangle_{p_\alpha}}\langle A(\Delta t)-A(0)\rangle_{p_\alpha}. \tag{4.26}$$

In this form we have a statement relating changes in a variable as it returns to the steady state ("dissipation") to its spontaneous fluctuations in that steady state. The term "dissipation," suggesting energy flow, may not apply in all applications of this theorem. There will nonetheless be inequalities relating this "dissipation" to changes in the relative entropy that we have defined.

Another (inequivalent) statement is that if

$$p(x)=\tilde{p}(x)\exp(\delta\varphi_1(x)+\cdots),$$

then

$$S(p(\cdot,\Delta t)|\tilde{p})-S(p|\tilde{p})\geqslant\tfrac{1}{2}\langle\delta^2\varphi_1^2\rangle_{\tilde{p}}(1-\mu_{\max}), \tag{4.27}$$

where $\mu_{\max}$ is the maximal (non-1) eigenvalue of the matrix ${}^tR(1/\tilde{p})\,R$ diag $(\tilde{p})$ [see Eqs. (1.11)–(1.12)].

F. Generalization to the case of *n* slow variables

1. Entropy

Let us now assume that one can find n slow variables $A_1,\ldots,A_n$ for the R dynamics. Take these such that

$$\langle A_j\rangle_{\tilde{p}}=0, \quad \text{for all } j.$$

For given values $a_1,\ldots,a_n$, we define

$$\tilde{P}(a_1,\ldots,a_n)=\sum_{\{x\in X|A_j(x)=a_j \text{ for all } j\}}\tilde{p}(x),$$

and we define a function

$$\Sigma(a_1,\ldots,a_n)=-\log\tilde{P}(a_1,\ldots,a_n).$$

As before (Sec. IV A), we have

$$\Sigma(a_1,\ldots,a_n)=-S(q_a|\tilde{p}),$$

where

$$q_a(x)=\frac{\tilde{p}(a,u)}{\tilde{P}(a)}\prod_j \delta(A_j(x)-a_j),$$

and Σ will be a Lyapunov function for the evolution of $a_1,...,a_n$.

2. General linear response theory

We generalize the results of Secs. IV C and IV D. Call $\alpha\equiv(\alpha_1,...,\alpha_n)$, where α_j is a conjugate variable for A_j and define as in Sec. IV C,

$$p_\alpha(x)=\frac{1}{Z_\alpha}\tilde{p}(x)\exp\left(\sum_{i=1}^{n}\alpha_i A_i+O(\alpha^2)\right).$$

It is immediate that

$$\langle A_i\rangle_{p_\alpha}=\sum_{j=1}^{n}\alpha_j\langle A_iA_j\rangle_{\tilde{p}}+O(\alpha^2).$$

One also proves

$$\langle A_i(\Delta t)-A_i(0)\rangle_{p_\alpha}=\sum_{j=1}^{n}\alpha_j\sum_{x,y}A_i(x)(R_{xy}-\delta_{xy})\tilde{p}(y)A_j(y)+O(\alpha^2)$$

and

$$\begin{aligned}\langle(A_i(\Delta t)-A_i(0))(A_j(\Delta t)-A_j(0))\rangle_{p_\alpha}&\\ =-\sum_{x,y}A_i(x)((R_{xy}-\delta_{xy})\tilde{p}(y)+(R_{yx}-\delta_{yx})\tilde{p}(x))A_j(y)+\sum_{k=1}^{n}\alpha_k\sum_{x,y}[-\tfrac{1}{2}A_i(x)((R_{xy}&\\ -\delta_{xy})\tilde{p}(y)A_k(y)+(R_{yx}-\delta_{yx})\tilde{p}(x)A_k(x))A_j(y)+\tfrac{1}{2}A_i(x)(A_j(x)R_{xy}\tilde{p}(y)A_k(y)&\\ -A_j(y)R_{yx}\tilde{p}(x)A_k(x))+\tfrac{1}{2}A_j(x)(A_i(x)R_{xy}\tilde{p}(y)A_k(y)-A_i(y)R_{yx}\tilde{p}(x)A_k(x))]+O(\alpha^2).&\end{aligned}$$

3. Choice of A_i as eigenvectors

We now choose the A_i to be left eigenvectors of R,

$$\sum A_i(x)R_{xy}=\lambda_iA_i(y).$$

Then, modulo $O(\alpha^2)$,

$$\langle A_i(\Delta t)-A_i(0)\rangle_{p_\alpha}=(\lambda_i-1)\sum_k\alpha_k\langle A_iA_k\rangle_{\tilde{p}}=(\lambda_i-1)\langle A_i(0)\rangle_{p_\alpha},$$

and modulo $O(\alpha A^3)$ terms,

$$\langle(A_i(\Delta t)-A_i(0))(A_j(\Delta t)-A_j(0))\rangle_{p_\alpha}=-\langle A_iA_j\rangle_{\tilde{p}}(\lambda_i+\lambda_j-2).$$

We can eliminate the eigenvalues $\lambda_i - 1$ and obtain modulo $O(\alpha A^3)$ terms,

$$\langle (A_i(\Delta t) - A_i(0))(A_j(\Delta t) - A_j(0)) \rangle_{p_\alpha}$$
$$= -\langle A_i A_j \rangle_{\tilde{p}} \left(\frac{\langle A_i(\Delta t) - A_i(0) \rangle_{p_\alpha}}{\langle A_i(0) \rangle_{p_\alpha}} + \frac{\langle A_j(\Delta t) - A_j(0) \rangle_{p_\alpha}}{\langle A_j(0) \rangle_{p_\alpha}} \right),$$

which is the fluctuation-dissipation statement in our general context.

Remark: If $(R, \tilde{p})$ satisfies detailed balance, $\tilde{p}$ vanishes nowhere and A is a left eigenvector of R with eigenvalue λ, then $\tilde{p}(x)A(x)$ is a right eigenvector of R with eigenvalue λ. When applied to the above equations and use is made of the orthogonality of the left and right eigenvectors of R, this implies a diagonal susceptibility matrix. In effect this says that choosing left eigenvectors of R as the slow variables chooses the macroscopic variables to be in a form diagonalizing the susceptibility.

G. Onsager reciprocity relations for nonequilibrium states

We consider a stationary state $\tilde{p}$ for a stochastic matrix R, and we consider the observables $A_1, \ldots, A_n$ with

$$\langle A_i \rangle_{\tilde{p}} = 0.$$

As usual, we define $p_\alpha = \tilde{p} \exp(\Sigma \alpha_i A_i + \cdots)$, and it follows that

$$\langle A_i \rangle_{\tilde{p}} = \sum_j \langle A_i A_j \rangle_{\tilde{p}} \alpha_j \tag{4.28}$$

(up to powers of α^2). We have seen that the relative entropy is

$$S(p_\alpha | \tilde{p}) = -\sum_{k,l} \langle A_k A_l \rangle_p^{-1} \langle A_k \rangle_{p_\alpha} \langle A_l \rangle_{p_\alpha}, \tag{4.29}$$

so that the corresponding forces F_k are given by

$$F_k \equiv -\frac{\partial S(p_\alpha | \tilde{p})}{\partial \langle A_k \rangle_{p_\alpha}} = \sum_l \langle A_k A_l \rangle_p^{-1} \langle A_l \rangle_{p_\alpha} = \alpha_k. \tag{4.30}$$

Moreover, the current (in one time step Δt) for A_k is given by

$$J_k \equiv \langle A_k(\Delta t) - A_k(0) \rangle_{p_\alpha} = \sum_j \alpha_j \sum_{xy} A_k(x)(R_{xy} - \delta_{xy}) \tilde{p}(y) A_j(y). \tag{4.31}$$

We can then write

$$J_k = \sum_j L_{kj} \alpha_j = \sum_j L_{kj} F_j, \tag{4.32}$$

where

$$L_{kj} = \sum_{xy} A_k(x)(R_{xy} - \delta_{xy}) \tilde{p}(y) A_j(y). \tag{4.33}$$

In general, the matrix L_{kj} is not symmetric. However, when $(R,\tilde{p})$ satisfies detailed balance, $R_{xy}\tilde{p}(y)=R_{yx}\tilde{p}(x)$, one can immediately see that $L_{kj}=L_{jk}$. Thus, an *absence* of detailed balance in R is manifested at the macroscopic level.

V. A PATH SUMMATION FORMULA FOR THE STATIONARY STATE

A. Expression of the stationary state in term of determinants

We consider a general $N\times N$ matrix A. Let us suppose that λ_0 is a nondegenerate eigenvalue of A and consider the right eigenvector u and the left eigenvector v of A of eigenvalue λ_0, so that

$$Au=\lambda_0 u, \quad vA=\lambda_0 v.$$

We normalize u,v so that

$$\sum_{i=1}^{N} u_i v_i=1. \tag{5.1}$$

Moreover, call $M(\lambda)_{ij}$ the minor of the element (i,j) in the matrix $\lambda I-A$ and $C(\lambda)$ the characteristic polynomial of A. Then one has the following identity:

$$u_i v_j=\frac{(-1)^{i+j}M_{ji}(\lambda_0)}{dC(\lambda_0)/d\lambda}. \tag{5.2}$$

This identity is derived in Appendix A, but can also be found in Ref. 26.

We apply this formula to a stochastic matrix R and to its eigenvalue 1, which we assume nondegenerate. The right eigenvector is the stationary state $\tilde{p}_j$ and the left eigenvector is $v_i=1$ for all i. The normalization condition of Eq. (5.1) is the normalization of the stationary state. Then Eq. (5.2) reduces to

$$\tilde{p}_i=\frac{(-1)^{i+j}M_{ji}(1)}{dC(1)/d\lambda}. \tag{5.3}$$

In particular, it is convenient to set $i=j$ and arrive at

$$\tilde{p}_i=\frac{M_{ii}(1)}{dC(1)/d\lambda}.$$

Using the normalization condition, we deduce the following identity:

$$\frac{dC(1)}{d\lambda}=\sum_{i=1}^{N} M_{ii}(1) \tag{5.4}$$

(this is a kind of partition function formula) and

$$\tilde{p}_i=\frac{M_{ii}(1)}{\sum_{j=1}^{N}M_{jj}(1)}. \tag{5.5}$$

B. Tree summation formula for the stationary state

We shall now state and derive a "tree integral" formula for the stationary state from Eq. (5.5). This result has been independently discovered on several occasions (including by us); we will

include our derivation, since the result is not known in the physics literature and is of interest for our statistical mechanics application. See Refs. 27–29, which may also be consulted for some of our tree-theory terminology.

We consider a stochastic matrix R of size N. Consider also the set of states $\{1,\ldots,N\}$ and among these points, we mark one point, say j_0, which we call the root. We can define a spanning tree of root j_0 as an oriented tree of root j_0 (the orientation going from the leaves to the root), such that any state $1 \leqslant k \leqslant N$ is a vertex of the tree.

We call T_j such a tree. Now, any edge (k,l) of T_j such that (k,l) is oriented from k to l is labeled by R_{lk}. The weight of the tree T_j is defined by

$$W(T_j)=\prod_{(k,l)\in T_j} R_{lk}. \tag{5.6}$$

We have the following result:
The minor M_{ij} of $-1+R_{jj}$ in $-I+R$ (for an $N\times N$ stochastic matrix R) is given by

$$M_{jj}=(-1)^{N-1}\sum_{T_j} W(T_j), \tag{5.7}$$

where the sum is taken over all spanning trees T_j with root j, as defined above. We shall prove this result in Sec. V D, but we can immediately make a number of comments: (i) Apart from the overall $(-1)^{N-1}$, M_{jj} is given by a sum of positive terms. (ii) M_{jj} is homogeneous of degree $N-1$ with respect to the $\{R_{kl}\}$ (for $k\neq l$). (iii) In a given term, $W(T_j)$, for a fixed T_j, a given R_{kl} does not appear twice. Moreover, one cannot have within a particular $W(T_j)$ a product of the type $R_{ik}R_{lk}$, but one can have terms like $R_{ki}R_{kl}$. Finally, one cannot have closed loops like $R_{i_1i_2}R_{i_2i_1}$.

The proofs of these statements are a direct consequence of the definition of a tree and of the weight W associated with it.

(i) Is obvious.

(ii) Is a consequence of the fact that a spanning tree for a set of N points has $N-1$ edges.

(iii) It is obvious that a given R_{kl} appears at most once in a $W(T_j)$. Moreover, since $W(T_j)$ is constructed by taking the product of the R_{lk}, starting from the leaves and following the edges up to the root j, it is clear that one cannot have a term $R_{ik}R_{lk}$ (this would mean that the vertex k has two fathers in the tree), but one can have $R_{ni}R_{nl}$ (when i and l are sons of the same father n).

Finally, we see that Eq. (5.5) has a natural meaning when we use the calculation of M_{jj} given in Eq. (5.7). The stationary probability of the state j is obtained by summing over all oriented paths leading from various points of the state space to the point j, quantities that are, for each path, the product of the elements R_{nl} that one encounters along the oriented path. Moreover, these paths may have several irreducible components leading to j, and they contain no loop. This is why such an oriented reducible path leading to j, is, in fact, a tree with root j.

C. The case of detailed balance

Detailed balance means that for all i,j,

$$\frac{R_{ij}}{R_{ji}}=\frac{\tilde{p}_i}{\tilde{p}_j}. \tag{5.8}$$

In particular, this implies that for any closed cycle $c=(x_1,x_2,x_3,\ldots,x_k,x_1)$ we have

$$\frac{R_{x_1x_k}R_{x_kx_{k-1}}\cdots R_{x_2x_1}}{R_{x_1x_2}R_{x_2x_3}\cdots R_{x_kx_1}}=1, \tag{5.9}$$

or with obvious notation,

$$\frac{\Pi_C R}{\Pi_{C^{-1}}R}=1,$$

where C^{-1} denotes the reverse cycle. On the other hand, we have proved above

$$\frac{\tilde{p}_i}{\tilde{p}_j}=\frac{M_{ii}}{M_{jj}},$$

in the general case.

Let us now assume that Eq. (5.9) holds for any closed cycle and prove that detailed balance holds. To fix everything, take $i=1$, $j=2$ and consider a tree of the type T_1. In this tree, there is a certain (unique) oriented path $\gamma(T_1)$ leading from 2 to 1, and this path has a certain length, $|\gamma(T_1)|$. Then

$$W(T_1)=\left[\prod_{\gamma(T_1)} R\right]W(T_1|\gamma(T_1)),$$

where $W(T_1|\gamma(T_1))$ is the product of all the elements of R along the edges of T_1 that are not on $\gamma(T_1)$.

Now, the edges of T_1 that are not on $\gamma(T_1)$ form a collection of $N-1-|\gamma(T_1)|$ edges [because $W(T_1)$ has $N-1$ edges, as we have seen]. These edges form a disjoint union of oriented trees with roots on the path $\gamma(T_1)$ and with their other vertices outside $\gamma(T_1)$ (this union of disjoint trees is a "forest").

Conversely, given a directed path γ leading from 2 to 1, and a forest F of trees having their roots in γ and their other vertices outside γ and with a total number of edges $N-1-|\gamma|$, the union of γ and F is a spanning oriented tree with root 1. This means that one can write

$$M_{11}=\sum_{\gamma \text{ from 2 to 1}}\left(\prod_{\gamma} R\right)\Phi(\gamma), \tag{5.10}$$

where $\Phi(\gamma)$ is defined to be

$$\Phi(\gamma)=\sum_{F} W(F), \tag{5.11}$$

where the sum is taken over all the oriented forests F having $N-1-|\gamma|$ edges, their roots on γ, and their other vertices outside γ, and $W(F)$ is the product of the R on all edges of F. In the same way,

$$M_{22}=\sum_{\gamma' \text{ leading from 1 to 2}}\left(\prod_{\gamma'} R\right)\Phi(\gamma'). \tag{5.12}$$

Now, for any path from 2 to 1, γ, one can find the inverse path γ^{-1} from 1 to 2 so that Eq. (5.12) can be rewritten,

$$M_{22}=\sum_{\gamma \text{ leading from 2 to 1}}\left(\prod_{\gamma^{-1}} R\right)\Phi(\gamma^{-1}).$$

But it is clear that

$$\Phi(\gamma)=\Phi(\gamma^{-1}),$$

and moreover, because of Eq. (5.9),

$$\frac{\Pi_\gamma R}{\Pi_{\gamma^{-1}} R}=\frac{R_{12}}{R_{21}},$$

so that

$$M_{11}=M_{22}\frac{R_{12}}{R_{21}},$$

from which we deduce Eq. (5.8).

D. Proof of Eq. (5.7)

We shall prove something slightly more general. In the following, Latin indices run from 1 to N and Greek indices run from $\underline{1}$ to $\underline{p}$. We consider $b_{\alpha i}$ and a_{ij} $(i\neq j)$ to be positive numbers. (For emphasis, numerical values taken by Greek indices are underlined. We make this distinction because at a later stage we will need to deal with switches between one sort of index and the other.)

We define the following determinant:

$$D_N(b,a)=\det\begin{vmatrix} -\sum_{\alpha=1}^{p} b_{\alpha 1}-\sum_{i=2}^{N} a_{i1} & a_{12} & \cdots & a_{1N} \\ a_{21} & -\sum_{\alpha=1}^{p} b_{\alpha 2}-\sum_{\substack{i=1\\ i\neq 2}}^{N} a_{i2} & \cdots & a_{2N} \\ \vdots & & \vdots & \vdots \\ \vdots & & \vdots & \vdots \\ a_{N1} & a_{N2} & \cdots & -\sum_{\alpha=1}^{p} b_{\alpha N}-\sum_{i\neq N} a_{iN} \end{vmatrix}.$$

It is obvious that if all bs are zero, this determinant is zero because the sum of all lines is zero.

Moreover, $D_N(b,a)$ is a homogeneous function of degree N of the bs and the as. We now consider the set $\{\underline{1},...,\underline{p}\}\cup\{1,...,N\}$, and for each α we consider an oriented tree T_α with root α and with other vertices in $\{1,...,N\}$. To any oriented edge (k,l) or (n,α) of T_α, we associate the label a_{lk} or $b_{\alpha n}$. We define

$$W(T_\alpha)=\left(\prod_{(k,l)\in T_\alpha} a_{lk}\right)\left(\prod_{(n,\alpha)\in T_\alpha} b_{\alpha n}\right). \tag{5.14}$$

We call a spanning forest a union of disjoint trees $\{T_\alpha\}$ for $\alpha=\underline{1},...,\underline{p}$ as before, such that all other vertices $1\leq i\leq N$ belong to a tree T_α of the forest (and then to a unique one),

$$W(F)=\prod_{F=\{T_\alpha\}} W(T_\alpha), \tag{5.15}$$

with the convention that, if T_α contains no edge, $W(T_\alpha)=1$. In particular, a spanning forest has N edges.

The main result of this section is that

$$D_N(b,a)=(-1)^N\sum_F W(F), \tag{5.16}$$

where the sum is taken on all spanning forests, as defined before. The proof of this statement is by induction on N.

First, for $N=2$ we have

$$D_2(b,a)=\det\begin{vmatrix} -\sum_{\alpha=\underline{1}}^{p} b_{\alpha 1}-a_{21} & a_{12} \\ a_{21} & -\sum_{\alpha=\underline{1}}^{p} b_{\alpha 2}-a_{12} \end{vmatrix} = \sum_{\alpha,\beta=\underline{1}}^{p} b_{\alpha 1}b_{\beta 2}+\sum_{\alpha=\underline{1}}^{p} b_{a1}a_{12}+\sum_{\alpha=\underline{1}}^{p} b_{\alpha 2}a_{21},$$

which obviously has the structure of Eq. (5.16).

We now assume that the statement is true for all determinants of size less than N, and we consider D_N as defined by Eq. (5.13). It is clear that D_N is symmetric by permutation of the Latin indices. As we have seen, it is a polynomial in the $b_{\alpha j}$ of degree $\leqslant N$ without a constant term [because $D_N(0,a)=0$]. Moreover, it is clear that in a given monomial, one cannot have products $b_{\alpha j}b_{\beta j}$, $a_{ik}a_{jk}$, because they correspond to terms in the same column of $D_N(b,a)$.

By symmetry, we can consider only the terms containing $b_{\underline{1}1}$ as a factor. Such a term is obviously

$$-b_{\underline{1}1}\det\begin{vmatrix} -\sum_{\alpha=\underline{1}}^{p} b_{\alpha 2}-\sum_{\substack{i=1\\ i\neq 2}}^{N} a_{i2} & a_{23} & \cdots & a_{2N} \\ a_{32} & -\sum_{\alpha=\underline{1}}^{p} b_{\alpha 3}-\sum_{\substack{i=1\\ i\neq 3}}^{N} a_{i3} & \cdots & a_{3N} \\ \vdots & & \vdots & \vdots \\ \vdots & & \vdots & \vdots \\ a_{N2} & a_{N3} & \cdots & -\sum_{\alpha=\underline{1}}^{p} b_{\alpha N}-\sum_{i\neq N} a_{iN} \end{vmatrix}.$$

But the determinant multiplying $-b_{\underline{1}1}$ is a determinant of the type $D_{N-1}(b',a')$, where now the new greek indices vary in the new set:

$$\{\underline{1},\ldots,\underline{p},\underline{p+1}\}\equiv\{\underline{1},\ldots,\underline{p},1\}$$

(i.e., $\underline{p+1}$ is the old latin index 1), and the set $\{1,\ldots,N\}$ has been replaced by $\{2,\ldots,N\}$ and with the identifications

$$b'_{\alpha i}=b_{\alpha i},\quad \underline{1}\leqslant\alpha\leqslant\underline{p},\quad 2\leqslant i\leqslant N,$$

$$b'_{\underline{p+1},i}=a_{1,i},\quad 2\leqslant i\leqslant N,$$

$$a'_{ij}=a_{ij},\quad 2\leqslant i\neq j\leqslant N.$$

But now, $D_{N-1}(b',a')$ is a sum over all spanning forests F' (with $N-1$ edges) of the weight of these forests. Each forest F' is a union of trees $\{T'_\alpha\}$ with roots in $\{\underline{1},...,\underline{p},1\}$ and with other vertices in $\{2,...,N\}$ and

$$W(F')=\left[\prod_{\alpha=\underline{1}}^{\underline{p}} W(T'_\alpha)\right]W(T'_1).$$

It may happen that some trees have no edge. Then we rewrite

$$b_{\underline{1}1}W(F')=(b_{\underline{1}1}W(T'_1)W(T'_{\underline{1}}))\prod_{\alpha=\underline{2}}^{\underline{p}} W(T'_\alpha).$$

But this is exactly

$$b_{\underline{1}1}W(F')=\prod_{\alpha=\underline{1}}^{\underline{p}} W(T_\alpha),$$

with

$$\begin{aligned} T_\alpha &= T'_\alpha, \quad \text{for } \alpha=\underline{2},\ldots,\underline{p}, \\ T_{\underline{1}} &= T'_{\underline{1}}\cup((1\,\underline{1})\cup T'_1), \end{aligned} \tag{5.17}$$

where $((\underline{1}1)\cup T_1)$ denotes a tree having as the root the point $\underline{1}$ and obtained by taking the edge $(1\underline{1})$ from 1 to $\underline{1}$ and attaching to it the tree T'_1 of root 1 (if it has some edge). Then

$$b_{\underline{1}1}W(F')=W(F),$$

where

$$F=\{T_{\underline{1}},T_{\underline{2}},\ldots,T_{\underline{p}}\}.$$

Conversely, any spanning forest F that contains the oriented edge, $(1\underline{1})$, is obviously a union of disjoint trees $T_{\underline{1}},\ldots,T_{\underline{p}}$ with $T_{\underline{1}}$ given as in Eq. (5.17), namely

$$T_{\underline{1}}=T'_{\underline{1}}\cup((1\,\underline{1})\cup T'_1),$$

with $T'_{\underline{1}}$ having its root in $\underline{1}$ and vertices in $\{2,...,N\}$ and T'_1 having its root in 1 and its vertices in $\{2,...,N\}$.

Thus, in $D_N(b,a)$ the terms containing $b_{\underline{1}1}$ are of the type $(-1)^N\Sigma_{1\underline{1}\in F}W(F)$. This proves Eq. (5.16).

Proof of Eq. (5.7) for M_{jj}: Eq. (5.7) is a particular case of Eq. (5.16). In this case, $p=\underline{1}=j$, the $N-2$ indices $l\neq j$ correspond to the Latin indices, and the Greek index α can take only the value j. Finally $b_{\alpha l}=a_{jl}$ and for $l,k\neq j$, $a_{lk}=R_{lk}$.

In this case a spanning forest for $\{\underline{1}\}\cup\{1,...,N-2\}$ having its root at j and other indices at points $k\neq j$ is exactly a spanning tree with root at j.

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APPENDIX: MATRIX FORMULAS

In this appendix, we derive the formulas of Sec. V A. We consider a matrix A and an eigenvalue λ_j of A. Call $\lambda_1,...,\lambda_p$ the other distinct eigenvalues. Then the matrix A can be decomposed in the Jordan normal form and we call E_{λ_j} the subspace associated with the eigenvalue λ_j in this decomposition. We call P_{λ_j} the projector on E_{λ_j} defined by

$$\begin{aligned} P_{\lambda_j}|_{E_{\lambda_j}} &= I, \\ P_{\lambda_j}|_{E_{\lambda_l}} &= 0, \quad j \neq l. \end{aligned} \tag{A1}$$

Take a contour γ_j in the complex plane surrounding λ_j once, but no λ_l for $l \neq j$. Then

$$P_{\lambda_j} = \frac{1}{2i\pi} \int_{\gamma_j} (z-A)^{-1}\, dz. \tag{A2}$$

Formula (A2) is obvious: take a vector u in the space E_{λ_l} for $l \neq j$. Then $(z-A)^{-1}u$ is holomorphic in a neighborhood of λ_j and

$$\int_{\gamma_j} (z-A)^{-1} u\, dz = 0.$$

Now, if u is in the space E_{λ_j}, we can write on E_{λ_j},

$$A|_{E_{\lambda_j}} = (\text{diag}\ \lambda_j) + T,$$

where T is an upper triangular matrix. Then

$$(z-A)^{-1}|_{E_{\lambda_j}} = \text{diag}(z-\lambda_j)^{-1} + T',$$

where T' is another upper triangular matrix (depending holomorphically on z in a neighborhood of λ_j). Then

$$\frac{1}{2i\pi} \int_{\gamma_j} (z-A)^{-1} u\, dz = u,$$

so that the Cauchy integral on the right-hand side of (A2) is given exactly by (A1). In particular, since $(z-A)^{-1}_{nl} = (-1)^{n+l}[M_{ln}(z)/C(z)]$, we have, from (A2),

$$(P_{\lambda_j})_{nl} = (-1)^{n+l}\ \text{Residue}_{\lambda_j}\left(\frac{M_{ln}(z)}{C(z)}\right).$$

If λ_j is a simple eigenvalue, then E_{λ_j} is generated by the right eigenvector u and the projector is simply given by

$$(P_{\lambda_j})_{nl} = u_n^* v_l,$$

with the normalization condition of Eq. (5.1).

In the case of a stochastic matrix R, if we take, say M_{11}, it is easy to see that $M_{11} = (-1)^{l+1} M_{l1}$ directly (write down M_{11} explicitly, then replace row number l by the sum of all rows of M_{11} and use the stochastic property $\Sigma_{l=1}^{N} R_{ln} = 1$). In general,

$$M_{nn}=(-1)^{n+l}M_{ln}.$$

But because $\det(I-R)=0$, we have

$$\sum_{j=1}^{N}(\delta_{lj}-R_{lj})(-1)^{j}M_{lj}=0, \quad l=1,\ldots,N,$$

which implies that the vector with components M_{jj} is an eigenvector of eigenvalue 1 of R.

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