Splitting of the rate matrix as a definition of time reversal in master equation systems

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Abstract
Motivated by recent progress in nonequilibrium fluctuation relations, we present a generalized time reversal for stochastic master equation systems with discrete states, which is defined as a splitting of the rate matrix into irreversible and reversible parts. An immediate advantage of this definition is that a variety of fluctuation relations can be attributed to different matrix splittings. Additionally, we find that the accustomed total entropy production formula and conditions of the detailed balance must be modified appropriately to account for the reversible rate part, which was previously ignored.

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(Some figures may appear in colour only in the online journal)

1. Introduction

Fluctuation theorems or fluctuation relations [1–14] are a variety of exact equalities about statistics of entropy production or dissipated work that are held even in far from equilibrium regimes. In the near-equilibrium region, these relations reduce to the famous fluctuation–dissipation theorems [2, 6, 15–17]. The discovery of these fluctuation relations significantly advances our understanding of nonequilibrium physics, and particularly the second law of thermodynamics of small systems [18].

Fluctuation relations are very relevant to the concept of time reversal [2–13, 19, 20]. For instance, under the framework of Markovian stochastic systems, previous work has proved that a majority of them can be derived by a ratio of probability densities observing a trajectory in an original system and the reversed trajectory in the time-reversed system. Very recently, Chetrite and Gawedzki [21] further elaborated this observation and presented a generalized time-reversal definition on continuous diffusion processes. Different from the conventional
definition of time reversal as simply changing the time parameter in a stochastic dynamics into minus, they explicitly defined time reversal as a splitting of the drift vector into irreversible and reversible parts which possess distinct rules under the transformation of $t \to -t$. Because of the freedom of the splitting, a variety of time reversal and corresponding fluctuation relations are obtained, e.g., the Hatano–Sasa equality [11] arising from a novel time reversal with nonzero reversible drift. The importance of the generalized time reversal was shown again when we understood the origin of a generalized integral fluctuation relation (GIFR) on general diffusion processes [22, 23].

In addition to the continuous diffusion process, another very typical Markovian process is the master equation with discrete states and continuous time [24]. Although the latter is more general than the former in principle, due to their highly formal analogy, many results and evaluations about fluctuation relations in diffusion processes could be established correspondingly in the master equation systems [6, 13, 25–28]. Nevertheless, whether a similar time-reversal definition exists and how to define it has not yet been investigated. We must emphasize that it is not trivial as one might think at first glance. Except for positive transition rates, in master equation systems there are no quantities such as drift vector and diffusion matrix that have intuitive rules of transformation as time is reversed [29]. Somewhat intriguingly, here we will show that a physically relevant answer indeed exists.

Before starting, we want to remind the reader that one purpose of this work is to reveal the essential role of a reversible part of the rate matrix when discussing time reversal. Although this concept is not unfamiliar in continuous diffusion processes, e.g. the Kramers equation, it is not widely acknowledged in master equation systems with discrete states. The reader will see that in the time-reversible system, having an equilibrium state, the reversible part nonvanishes only in the presence of both odd and even variables. However, almost all classical textbooks are limited to the cases with even variables only, e.g. particle numbers in chemical reactions [24, 29–31]. This paper is organized as follows. In section 2, we briefly review a GIFR in the master equation systems that we found very recently [27]. The reason that we use the GIFR rather than other famous fluctuation relations is its generality. Moreover, the necessity of extending the conventional time reversal will be brought forth naturally in deriving the GIFR. In order to interpret an unknown matrix in this relation, in section 3 we present a generalized time reversal in these systems as a splitting of the rate matrix into reversible and irreversible parts. The consequences of this new definition will be discussed in section 4, which includes a reinvestigation of the fluctuation relations from the point of view of the splitting, generalization of total entropy production and conditions of the detailed balance. Section 5 contains the summary.

2. A review of the GIFR in master equation systems

Assume that a Markovian process with discrete states and continuous time is described by the master equation

$$\frac{dp_n(t)}{dt} = [H(t)p(t)]_n,$$

(1)

where $n$ is the state index which may be a vector, e.g. $n = (n_1, n_2, \ldots)$, the $N$-dimensional column vector $p(t) = (p_1, \ldots, p_N)^T$ is the probability of the system at individual states at time $t$, and the matrix element $(H)_{mn} = H_{mn} > 0$ ($m \neq n$) is the time-dependent or -independent rate and $(H)_{nn} = -\sum_{m \neq n} H_{mn}$. Given a normalized positive column vector $f(t) = (f_1, \ldots, f_N)^T$ and an $N \times N$ matrix $A$ whose elements $(A)_{mn} = A_{mn}$ ($m \neq n$) satisfy the condition $H_{mn}f_n + A_{mn} > 0$ and $A_{nn} = -\sum_{m \neq n} A_{mn}$, we found that the inner product
\[ \mathbf{f}'(t')\mathbf{v}(t') \quad (t' < t) \] is time-\( t' \)-invariable if the vector \( \mathbf{v}(t') = (v_1, \ldots, v_N)^T \) satisfies a perturbed backward equation \[ \frac{d v_n(t')}{d t'} = - (H^T \mathbf{v})_n - f_n^{-1} (\partial_t \mathbf{f} - \mathbf{H} \mathbf{v})_n + f_n^{-1} [(\mathbf{A}^T)_n \mathbf{v}_n - (\mathbf{A}^T \mathbf{v})_n], \tag{2} \]

where the final condition \( v_n(t) = d_n \) and the \( N \)-dimensional column vector \( \mathbf{1} = (1, \ldots, 1)^T \).

This is easily proved by noting the time differential property and the transpose property of a matrix. Employing the Feynman–Kac and Girsanov formulas for discrete jump processes, the solution of (2) could be rewritten into a concise form \[ \text{f}_{n}(t') \mathbf{d}, \tag{3} \]

and the integrant in the functional is

\[ \mathcal{J}[\mathbf{f}, \mathbf{A}] = \int_{\mathbf{x}(\tau)} \mathbf{f}_{n}^{-1} [-\partial_t \mathbf{f} + \mathbf{H} \mathbf{v} + \mathbf{A} \mathbf{v}_{\mathbf{n}}] + \mathcal{Q}[\mathbf{x}(\tau) \mathbf{A}], \tag{4} \]

with

\[ \mathcal{Q}[\mathbf{B}] = - \mathbf{B} \mathbf{x}(\tau) \mathbf{x}(\tau) - \ln \left[ 1 + \frac{\mathbf{B} \mathbf{x}(\tau) \mathbf{x}(\tau)}{\mathbf{H} \mathbf{x}(\tau) \mathbf{x}(\tau)} \right] \sum_{j=1}^{k} \delta(\tau - \tau_j), \tag{5} \]

where \( m^{0}(\cdot) \) is the expectation over all trajectories \( \mathbf{x} \) generated from the system (1) with a fixed state at initial time 0, \( \mathbf{x}(\tau) \) is the discrete state of the system at instant time \( \tau \), and \( \mathbf{x}(\tau^-) \) represent the states just before and after a jump occurring at time \( \tau \), respectively, and we have assumed that the jumps occur \( k \) times for a trajectory. By selecting different \( \mathbf{f} \) and \( \mathbf{A} \), the GIFR (3) may be reduced to the different fluctuation relations in the literature [27].

The perturbed backward equation seems complicated. However, by multiplying both sides of (2) with \( f_n \) and introducing a normalized probability vector \( \mathbf{q}(s) \) whose elements are

\[ q_{n}(s) = [(\mathbf{f}(t') \mathbf{d})^{-1} f_{n}(t') v_{n}(t') \quad (s = t - t'), \tag{6} \]

equation (2) could be rewritten into a concise form [27],

\[ \frac{d q_{\bar{n}}(s)}{d s} = \left[ \hat{H}(s) \mathbf{q}(s) \right]_n, \tag{7} \]

where the elements of the matrix \( \hat{H}(s) \) are

\[ \hat{H}_{mn}(s) = \int_{t_n} f_{n}^{-1}(t') [H_{mn}(t') f_{m}(t') + A_{mn}(t')] \tag{8} \]

for \( m \neq n \) and \( \hat{H}_{nn}(s) = - \sum_{n \neq n} \hat{H}_{nn}(s) \) and \( \mathbf{n} = (\epsilon_1 n_1, \epsilon_2 n_2, \ldots) \) with \( \epsilon_i = +1 (-1) \) if the component is even (odd) under time reversal \( t \rightarrow -t \). Here we have assumed that the state space is large enough to include both \( n \) and \( \mathbf{n} \). Considering that the parameter \( s \) is analogous to a reversed time and \( \hat{H}_{mn}(s) = H_{mn}(t') \) when selecting \( \mathbf{A} \) to be a probability flux \( \mathbf{J}_{mn} = H_{mn} f_{m} - H_{mn} f_{n} \), we called equation (7) a generalized time reversal of the original system (1) [27]. However, for the general matrix \( \mathbf{A} \) such a “generalization” is bit odd. Hence, a reinvestigation seems essential.

3. Splitting rate matrix as a time reversal

Let us start with two general matrix identities:

\[ [\mathbf{M}(\mathbf{a}, \mathbf{b})]_n = [\mathbf{M}]_n b_n + a_n [\mathbf{M}^T \mathbf{b}]_n = [\mathbf{S}^T \mathbf{b}]_n + [\mathbf{S}]_n b_n, \tag{9} \]

\[ [\mathbf{M}(\mathbf{a}, \mathbf{b})]_n + [\mathbf{M}]_n b_n - a_n [\mathbf{M}^T \mathbf{b}]_n = [\mathbf{T}^T \mathbf{b}]_n - [\mathbf{T}]_n b_n, \tag{10} \]
where both \( \mathbf{a} \) and \( \mathbf{b} \) are \( N \)-dimensional vectors, \( \mathbf{M} \) is an \( N \times N \) matrix satisfying \( \sum_n M_{mn} = 0 \), \( (\mathbf{a}, \mathbf{b}) = (a_1 b_1, \ldots, a_N b_N)^T \) represents an array multiplication of the vectors and the matrices \( \mathbf{S} \) and \( \mathbf{T} \) are constructed by \( \mathbf{M} \) and \( \mathbf{a} \) as follows:

\[
(S)_{mn} = M_{mn} a_m + M_{nm} a_n, \quad (T)_{mn} = M_{nm} a_m - M_{mn} a_n.
\]

They are symmetric and antisymmetric, respectively. Proving equations (9) and (10) is straightforward.

Now we assume that the rate matrix can be split into a sum of ‘reversible’ and ‘irreversible’ matrices, namely \( \mathbf{H} = \mathbf{H}^{\text{rev}} + \mathbf{H}^{\text{irr}} \). Particularly, we require \( H^{\text{int}}_{mn} > H^{\text{rev}}_{mn} (m \neq n) \), the reason for which will be seen shortly. Obviously, \( H^{\text{int}}_{mn} \) is greater than zero for \( H^{\text{int}}_{mn} > 0 \). We further assume that under a time reversal \( \mathbf{H} \) is transformed into \( \mathbf{H} = \mathbf{H}^{\text{rev}} + \mathbf{H}^{\text{irr}} \) and specifically

\[
\mathbf{H}^{\text{int}}_{mn}(s) = H^{\text{int}}_{mn}(t'), \quad \mathbf{H}^{\text{rev}}_{mn}(s) = -H^{\text{rev}}_{mn}(t').
\]

We see that the time-reversed matrix \( \mathbf{H} \) still possesses a rate matrix interpretation. Using these definitions and identities we rewrite the right-hand side of (7) as

\[
[\mathbf{H}\mathbf{q}]_n = \frac{1}{\Gamma(t)} \mathbf{d}^T \left[ [\mathbf{H}^{\text{rev}}(\mathbf{f}, \mathbf{v})]_n + [\mathbf{H}^{\text{rev}}(\mathbf{f}, \mathbf{v})]_n \right]
= \frac{1}{\Gamma(t)} \mathbf{d}^T \left[ (\mathbf{H}^{\text{rev}})_{nm} f_m - (\mathbf{H}^{\text{rev}})_{nm} f_n \right],
\]

where

\[
(S^{\text{rev}})_{mn} = H^{\text{rev}}_{mn} f_m + H^{\text{rev}}_{nm} f_n,
\]

\[
(T^{\text{rev}})_{mn} = H^{\text{rev}}_{mn} f_m - H^{\text{rev}}_{nm} f_n.
\]

Comparing equation (13) with (2), we immediately find that

\[
(A)_{mn} = (T^{\text{rev}})_{mn} - (S^{\text{rev}})_{mn} = H^{\text{rev}}_{mn} f_m - H^{\text{rev}}_{nm} f_n - 2H^{\text{rev}}_{mn} f_m.
\]

Now we can explain why the matrix \( \mathbf{A} \) is almost arbitrary: the reversible matrix \( \mathbf{H}^{\text{rev}} \) to be determined is responsible for this freedom. In addition, we also note that the condition \( H^{\text{int}}_{mn} f_m + A_{mn} > 0 \) mentioned in the previous section is completely identical to the condition \( H^{\text{int}}_{mn} > H^{\text{rev}}_{mn} \) here.

In practice, we may first know the matrix \( \mathbf{A} \) or the time-reversed rate matrix \( \mathbf{H}^{\text{rev}} \). Under these circumstances, the splitting can be constructed conversely as

\[
H^{\text{rev}}_{mn} = (H^{\text{int}}_{mn} f_m - H^{\text{int}}_{mn} f_n - A_{mn})/2 f_m,
\]

\[
H^{\text{int}}_{mn} = (H^{\text{int}}_{mn} f_m + H^{\text{int}}_{mn} f_n + A_{mn})/2 f_m.
\]

Regardless of how a splitting of the rate matrix is achieved, substituting (16) into the integrand (4), its alternative form is obtained,

\[
\mathcal{J}[\mathbf{f}, \mathbf{H}^{\text{rev}}, \mathbf{H}^{\text{rev}}] = -\frac{d}{d\tau} \ln f_{\mathbf{v}(\tau)} (\tau) + \mathcal{J}[\mathbf{I}, \mathbf{H}^{\text{rev}}, \mathbf{H}^{\text{rev}}].
\]

\footnote{In the following we will alternatively use \( \mathcal{J}[\mathbf{f}, \mathbf{A}] \) and \( \mathcal{J}[\mathbf{f}, \mathbf{H}^{\text{int}}, \mathbf{H}^{\text{rev}}] \) without an explicit statement, which would not result in confusion because of the established equation (16).}
where the second term on the right-hand side is
\[
-2 \sum_{m \neq x} H_{mn}^{rev} - \ln \left[ \frac{H_{mn}^{irr}}{H_{mn}^{rev} + H_{mn}^{rev}} \right] \sum_{j=1}^{k} \delta(\tau - \tau_j). \tag{22}
\]
Equation (21) is a consequence of the arbitrariness of $f$ in equation (6), a brief explanation of which is given in appendix A. Note equation (22) also satisfies a fluctuation relation if the initial distribution of the system is uniform. Equations (21) and (22) are the central results of this work.

4. Discussion

Because the physical interpretation of the GIFR (3), its connection with the other fluctuation relations and its detailed version have been partially investigated [27], in the remainder of this paper we present only several new results obtained from the point of view of the splitting.

4.1. Total entropy production rate

If $f$ is the system’s probability vector, $p$, and the splitting is first known from physical consideration, e.g., time reversibility below, then (21) is the balance equation of trajectory entropy: the first term on the right-hand side is the change in trajectory system entropy, the second term is the change in trajectory environmental entropy along a specified trajectory and the left-hand side is the total trajectory entropy. This interpretation has been widely accepted [13, 25, 26, 28, 32]. To our knowledge, however, the expression of the trajectory environmental entropy (22) that involves both even and odd discrete variables is first given here. This new formula also reminds us that the average total entropy production for the master equation systems with both irreversible and reversible rate matrices is
\[
\langle J[p, H^{irr}, H^{rev}] \rangle = -2 \sum_{m \neq n} H_{mn}^{rev} p_m + \sum_{m \neq n} \sum_n \left( H_{mn}^{irr} + H_{mn}^{rev} \right) p_m \ln \left( \frac{H_{mn}^{irr} - H_{mn}^{rev}}{H_{mn}^{irr} + H_{mn}^{rev}} \right) p_n \geq 0. \tag{23}
\]
The inequality may be proved easily using $(x - 1) \geq \ln x$ or the Jensen inequality for the GIFR (3). We note that equation (23) is significantly distinct from the classical entropy production formula given by Schnakenberg [33] that involves only even variables.

4.2. Conditions of the detailed balance

For a time-independent master equation system that has equilibrium state $p^{eq}$, we may physically require it to be invariable under time reversal, namely, $\tilde{H} = H$. Then, according to equations (19) and (20) a unique splitting is
\[
H_{mn}^{rev} = (H_{mn} - H_{mn}^{d})/2, \quad H_{mn}^{irr} = (H_{mn} + H_{mn}^{d})/2. \tag{24}
\]
Under this circumstance the irreversible and reversible parts have distinctive properties:
\[
H_{mn}^{irr} = -H_{nm}^{rev}, \quad H_{mn}^{rev} = H_{nm}^{irr}. \tag{25}
\]
Obviously, if the discrete master equation system involves only even variables, which is exclusively the object investigated in numerous references [24, 29–31], the reversible rate matrix must vanish. Because the entropy production is zero when the system is in the equilibrium state, according to equation (23), all terms on its right-hand side must vanish. Hence, we obtain the conditions of detailed balance on the rate matrix and the state that are
\[
(S^{eq})_{mn} = H_{mn}^{rev} p_{mn} + H_{mn}^{rev} p_{nm} = 0, \tag{26}
\]
respectively. We must emphasize that, except for the last equation (28) which is from the steady-state requirement and was ignored in previous literature [34], the former two equations are fully equivalent with the conventional detailed balance condition [35]

\[ H_{\text{irr}}^{mn} = H_{\text{irr}}^{nm} p_{eqm} - H_{\text{irr}}^{nm} p_{eqn} = 0, \]

(27)

\[ \sum_{m \neq n} H_{\text{rev}}^{mn} = 0, \]

(28)

One may easily check it using equation (24). We see an unusual feature of a time-reversible master equation system with nonvanishing \( H_{\text{rev}} \): the probability flux \( J_{mn} \) between two states \( m \) and \( n \) may not be zero even if the system is in an equilibrium state; both the irreversible and reversible rate matrices have contributed to the total entropy production. The latter point can be seen more clearly when we rewrite equation (23) as

\[ \langle J[p, H_{\text{irr}}, H_{\text{rev}}] \rangle = - \sum_{m \neq n} \sum_{n} H_{\text{irr}}^{mn} p_{mn} \ln \frac{H_{\text{irr}}^{mn} p_{mn}}{H_{\text{irr}}^{nm} p_{nm}} - \sum_{m \neq n} \sum_{n} H_{\text{rev}}^{mn} p_{mn} \ln \left(- \frac{H_{\text{rev}}^{mn} p_{mn}}{H_{\text{rev}}^{nm} p_{nm}}\right) \]

(30)

by taking equations (26)–(28) into account. To the best of our knowledge, there are few stochastic jump processes with both even and odd variables in the literature. In appendix B we present a simple mathematical model to exemplify their intriguing features.

4.3. Jarzynski and Hatano–Sasa equalities

For a time-dependent master equation system, we may select the simplest case with \( A = 0 \). Then equations (17) and (18) become

\[ H_{\text{rev}}^{mn} = \frac{1}{2f_{m}} (H_{mn} f_{m} - H_{nm} f_{n}), \]

(31)

\[ H_{\text{irr}}^{mn} = \frac{1}{2f_{m}} (H_{mn} f_{m} + H_{nm} f_{n}). \]

(32)

We use a new notation \( \mathcal{H} \) to indicate the speciality of this splitting. Under this circumstance, equation (21) is

\[ \mathcal{J}[f, 0] = - \frac{d}{d\tau} \ln f_{\mathcal{X}(\tau)}(\tau) + \mathcal{J}[1, H_{\text{irr}}, H_{\text{rev}}], \]

(33)

and

\[ \mathcal{J}[1, H_{\text{irr}}, H_{\text{rev}}] = - \frac{1}{f_{\mathcal{X}(\tau)}} \sum_{m \neq \mathcal{X}(\tau)} [H_{\mathcal{X}(\tau)m} f_{m} - H_{m\mathcal{X}(\tau)} f_{\mathcal{X}(\tau)}] - \ln \frac{f_{\mathcal{X}(\tau)}}{f_{\mathcal{X}(\tau)}} \sum_{i=1}^{k} \delta(\tau - \tau_{i}). \]

(34)

If one further selects \( f \) to be the instant equilibrium solution \( p_{eq}(t) \) or the instant nonequilibrium steady-state solution \( p_{ss}(t) \) of the master equation, then the GIFR is respectively the famous Jarzynski equality about the dissipated work [7, 8] or Hatano–Sasa equality about the excess entropy [11]. Note that in both cases, the first term on the right-hand side of the above equation vanishes because of the steady-state condition. It is worth pointing out that for the former equality, the detailed balance conditions (26) and (27) imply that such a splitting is

Haken may have been the first to investigate the stationary solution of the master equation for systems with both even and odd variables in detailed balance [34], but the conditions that must be satisfied by the rates and the total entropy production were not further pursued by him.
trivial because of $\mathcal{H}_{\text{irr}}^{\text{ev}}(t) = H_{\text{irr}}^{\text{ev}}(t)$ and $\mathcal{H}_{\text{int}}^{\text{int}}(t) = H_{\text{int}}^{\text{int}}(t)$, and (34) has other three different but equivalent expressions:

$$
\ln \frac{P_{\text{eq}}(t^-)}{P_{\text{eq}}(t^+)} = \ln \frac{H_{\text{irr}}^{\text{int}}(t^-)}{H_{\text{irr}}^{\text{int}}(t^+)} = \ln \left[ -\frac{H_{\text{irr}}^{\text{ev}}(t^-)}{H_{\text{irr}}^{\text{ev}}(t^+)} \right] \\
= \ln \frac{H_{\text{irr}}^{\text{int}}(t^-)}{H_{\text{irr}}^{\text{int}}(t^+)} - H_{\text{irr}}^{\text{ev}}(t^-) + H_{\text{irr}}^{\text{ev}}(t^+),
$$

(35)

On the other hand, if $A_1 = 0$, the integrand (4) can be decomposed into

$$
\mathcal{J}[f, A] = \mathcal{J}[f, 0] + Q [f_{X(t)}^{-1}, A],
$$

(36)

This relationship is very intriguing. In addition to the fact that the three terms above satisfy the integral fluctuation relation [27] simultaneously, all of them have important physical implications in the nonequilibrium steady-state thermodynamics [11, 14, 28, 32, 36]. Here we do not repeat previous interpretations but present a simple understanding from the splitting viewpoint. We assume that the nonequilibrium master equation system possesses only an irreversible rate part, namely, $H_{\text{irr}}^{\text{int}}(t) = H(t)$, and the system always has a unique steady-state $p^\text{ss}(t)$ as the external time-dependent parameter is fixed. Obviously, the corresponding instant steady-state current $(J^\text{ss})_{nm} = H_{nm} p^\text{ss}_m - H_{mn} p^\text{ss}_n$ satisfies $J^\text{ss} \mathbf{1} = 0$. On the basis of equations (21), (36) and (33), we have

$$
\mathcal{J}[p, J] = -\frac{d}{dt} \ln \frac{p(t)}{p^\text{eq}(t)} + \mathcal{J}[p^\text{ss}, J^\text{ss}]
$$

(37)

$$
= -\frac{d}{dt} \ln \frac{p(t)}{p^\text{eq}(t)} + \mathcal{J}[p^\text{ss}, 0] + Q \left[ \frac{J^\text{ss}}{p^\text{ss}(t)} \right]
$$

(38)

$$
= -\frac{d}{dt} \ln p(t) + \mathcal{J}[1, H_{\text{int}}, H_{\text{ev}}] + Q \left[ \frac{J^\text{ss}}{p^\text{ss}(t)} \right].
$$

(39)

Equation (38) implies that trajectory total entropy is the sum of trajectory relative entropy, trajectory excess entropy [11] and trajectory housekeeping entropy [14]. Particularly, the sum of the first two terms in equation (39), which was called nonadiabatic trajectory entropy [28], is just $\mathcal{J}[p, H_{\text{int}}, H_{\text{ev}}]$, the ensemble average of which is

$$
\langle \mathcal{J}[p, H_{\text{int}}, H_{\text{ev}}] \rangle = -\sum_{m \neq n} \sum_n H_{nm} p_n \ln \frac{p_m p_n}{p_n^\text{eq} p_m^\text{eq}} \geq 0
$$

(40)

according to (23). In [32] this quantity was also called free energy dissipation.

5. Conclusion

Motivated by the important idea of defining a time reversal as a splitting of the drift vector in the continuous diffusion process, we present the same effort in the master equation system with discrete states. Different from the former, we define the time reversal in this system as a splitting of the rate matrix into irreversible and reversible parts. Even so, we find that very analogous formulas and results are revealed in these two systems, e.g. (22) corresponds to (7.6) in the work of Chetrite and Gawedzki [21] or (28) in our work [23]. The advantages of introducing this definition are obvious. First, we explain the origin of the matrix $A$ in the GIFR, which was somewhat mysterious to us before starting this work. Second, a variety of fluctuation relations in the master equation systems are unified into various rate matrix
splittings. This point was not acknowledged previously. Additionally, the relationships among these fluctuation relations become very clear from this splitting viewpoint, e.g. equations (38) and (39). Finally, this definition also reminds us of the importance of the reversible part of the rate matrix. For instance, the expression of the total entropy production must be modified appropriately. To our knowledge, its existence and implications in the master equation systems were almost ignored for a very long time.

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Appendix A. Derivation of equations (21) and (22)

For simplicity in notation, we study a specific case of (2) with a final condition \( d_n = 1 \). Under this circumstance the perturbed backward master equation has the time-reversal explanation (7) and

\[
\hat{q}_n(s) = f_n(t') u_n(t').
\]

Note that the initial condition of \( \hat{q}_n \) is now \( f_n(t) \). It is worth emphasizing that we can replace the vector \( f \) above by other normalized positive vectors, e.g. \( c = (1/N, \ldots, 1/N) \). Then we have a new relation

\[
\hat{q}_n(s) = c_n u_n(t'),
\]

where \( u_n \) satisfies the same (2) except that \( f_n \) therein including those in the matrix \( A \) (16) are substituted by \( c_n \) and the final condition becomes \( u_n(t) = f_n(t)/c_n \). According to (A.1), (A.2) and the path integral representation (3), we immediately obtain

\[
n_0,0 \langle e^{-\int_0^t J[f,A](x(\tau)) d\tau} \rangle = n_0,0 \langle e^{-\int_0^t J[c,A_c](x(\tau)) d\tau} \frac{f_{x(t)}(t)}{f_{x(0)}} \rangle,
\]

where \( A_c \) is the matrix of (16) but all \( f_n \) are substituted by \( c_n \). Because of apparent \( J[c,A_c] = J[1,A_1] \) equation (21) is proved given a prior splitting. An alternative proof may use a total derivative formula about the jump function \( f_{x(t)}(t) \); see (9) in [27].

Appendix B. A discrete jump model with both even and odd variables

To show the unusual features of jump processes with both even and odd variables, we construct a simple mathematic model; see figure B1(a). Assuming that the variable \( i = 1 \) and \( 2 \) are even and \( \uparrow \) and \( \downarrow \) are odd, under the time reversal \( (t \to -t) \) we have \( (i, \uparrow) = (i, \downarrow) \) and \( (i, \downarrow) = (i, \uparrow) \). Although this model has eight rate parameters, they would not to be independent of each other if we required the model to have a steady-state solution satisfying the detailed balance principle. Using equations (26)–(28) and performing a simple calculation, we find these rates must obey the following three constraints:

\[
\begin{align*}
lr - lr' &= 0, \\
ul - ul' &= 0, \\
ul' - ul &= 0.
\end{align*}
\]
The first equation is a consequence of (26) or (27), while the last two equations are from (28).

The equilibrium solutions can be easily obtained, which are

\[ p_{(1,\uparrow)}^{\text{eq}} = p_{(1,\downarrow)}^{\text{eq}} = (l + l')/2(l + l' + r + r'), \]

\[ p_{(2,\uparrow)}^{\text{eq}} = p_{(2,\downarrow)}^{\text{eq}} = (r + r')/2(l + l' + r + r'). \]

In addition, the total entropy production (30) is

\[ \langle \mathcal{J} \rangle = [d_1 p_{(1,\uparrow)} - u_1 p_{(1,\downarrow)}] \ln \frac{p_{(1,\uparrow)}}{p_{(1,\downarrow)}} + [d_2 p_{(2,\uparrow)} - u_2 p_{(2,\downarrow)}] \ln \frac{p_{(2,\uparrow)}}{p_{(2,\downarrow)}} + [r p_{(1,\uparrow)} - l p_{(2,\uparrow)}] \ln \frac{(r + r') p_{(1,\uparrow)}}{(l + l') p_{(2,\uparrow)}} + [r' p_{(1,\downarrow)} - l' p_{(2,\downarrow)}] \ln \frac{(r + r') p_{(1,\downarrow)}}{(l + l') p_{(2,\downarrow)}}, \]

where \( p_{(i,\uparrow)} \) and \( p_{(i,\downarrow)} \) are the transient probabilities of the model at distinct states starting from an initial distribution. The reader may easily check that, if \( p = p^{\text{eq}} \), the probability currents \( \mathcal{J} \) or the terms before these logarithmic functions above are nonzero.

It would be interesting to compare these results with those obtained in conventional jump processes with only even variables, e.g. figure B1(b). Its states are respectively marked by the variables 1, 2, 1', and 2', all of which are even under time reversal. Because of \( p^{\text{eq}}(i, \uparrow) = p^{\text{eq}}(i, \downarrow) \) in the model (a), here we additionally require \( p^{\text{eq}}(i) = p^{\text{eq}}(i') \). Hence, we obtain three constraints on the rates given by

\[ d_1 - u_1 = 0, \]
\[ d_2 - u_2 = 0, \]
\[ l' r' - l r = 0, \]

the equilibrium solutions

\[ p_{1}^{\text{eq}} = p_{1'}^{\text{eq}} = l/2(r + l), \]
\[ p_{2}^{\text{eq}} = p_{2'}^{\text{eq}} = r/2(r + l), \]

and the classical total entropy production [33]

\[ \langle \mathcal{J} \rangle_c = (d_1 p_1 - u_1 p_1') \ln \left( \frac{d_1 p_1}{u_1 p_1'} \right) + (d_2 p_2 - u_2 p_2') \ln \left( \frac{d_2 p_2}{u_2 p_2'} \right) \]
\[ + r p_1 - l p_2 \ln \left( \frac{r p_1}{l p_2} \right) + r' p_1 - l' p_2 \ln \left( \frac{r' p_1}{l' p_2} \right), \]

where \( p_1 \) and \( p_2 \) (\( i = 1, 2 \)) are the transient probabilities of the model (b) at individual states starting from an initial distribution. Obviously, in the model (a) if one did not take the odd

![Figure B1.](image-url)

(a) A discrete jump process with both even (1 and 2) and odd (\( \uparrow \) and \( \downarrow \)) variables, (b) jump process with only even variables (1', 2 and 2'). The letters \( r, l, r', l', u_1, d_1, u_2, d_2 (>0) \) are respective rates.
Figure B2. (a) Time evolution of the probabilities at the four states in figure B1(a) and (b) the entropy production rates calculated by the classical Schnakenberg formula (B.12) and (30) proposed in this work. We choose \( l = 1 \, \text{s}^{-1}, r = 0.2 \, \text{s}^{-1}, j_1 = 0.4 \, \text{s}^{-1}, j_2 = 0.5 \, \text{s}^{-1}; d_1 = 0.7 \, \text{s}^{-1}, w_1 = 0.4 \, \text{s}^{-1}, d_2 = 0.2 \, \text{s}^{-1} \) and \( w_2 = 0.8 \, \text{s}^{-1} \), which satisfy (B.1)–(B.3). The initial conditions are \( p_{(1,\uparrow)} = 1.0 \) and the others vanish.

variable into account and naively used (B.12), namely replacing \( p_i \) and \( p_i' \) therein by \( p_{(i,\uparrow)} \) and \( p_{(i,\downarrow)} \), respectively, he or she would find that the classical formula would not vanish as the system reaches the equilibrium states (B.4) and (B.5). A numerical example confirming these results is shown in figure B2.

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