Large deviation function and fluctuation theorem for classical particle transport

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We analytically evaluate the large deviation function in a simple model of classical particle transfer between two reservoirs. We illustrate how the asymptotic long-time regime is reached starting from a special propagating initial condition. We show that the steady-state fluctuation theorem holds provided that the distribution of the particle number decays faster than an exponential, implying analyticity of the generating function and a discrete spectrum for its evolution operator.

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I. INTRODUCTION

The second law of thermodynamics has two major ingredients: the existence, in an equilibrium state, of a state function called the entropy and the increase of the total entropy in a spontaneous transition between two equilibrium states. In recent years, equilibrium thermodynamic concepts have been extended to nonequilibrium situations. In particular, several relations associated with the distribution of fluctuations in entropy production and related dissipative processes have been reported for systems driven far from equilibrium. These are collectively known as fluctuation relations [1–3].

The fluctuation theorem (in its simplest form) states that the probability distribution function \( P(\Delta S) \) of the fluctuations in the total entropy production during a nonequilibrium evolution in a given time interval obeys the symmetry property

\[
\frac{P(\Delta S)}{P(-\Delta S)} = \exp\left\{ \frac{\Delta S}{k_B} \right\}.
\]

This equality implies the second law inequality \( \langle \Delta S \rangle \geq 0 \) for the average entropy production.

The earliest thermodynamic results for systems not in equilibrium were focused on the dissipated work in isolated driven systems [4,5]. Similar results were later reported for the entropy production in deterministic closed thermostatted systems [6] and for open systems described by stochastic dynamics [7–12]. The discovery of the fluctuation theorem entails a double departure from the standard formulation of the second law. Entropy is defined in nonequilibrium states, and it is even defined for a single realization in a nonextensive system, where it is a stochastic quantity which can increase as well as decrease with time.

Prior to the proper definition of a stochastic entropy [13], fluctuation theorems were discovered in “asymptotic versions.” These early so-called steady-state fluctuation theorems are valid for times much longer than the time scales associated with the relaxation of the system to a steady state. In this version, one only focuses on the entropy production in idealized reservoirs. For a heat reservoir \( i \) at temperature \( T^{(i)} \), the entropy change is given by \( \Delta S^{(i)} = Q^{(i)}/T^{(i)} \), where \( Q^{(i)} \) is the net amount of heat transferred from the system to reservoir \( i \). Note that \( Q^{(i)} \) and hence \( \Delta S^{(i)} = Q^{(i)}/T^{(i)} \) are well defined even though the system need not be at equilibrium. Furthermore, in the case of a small system, the amount of heat \( Q^{(i)} \) will differ from one realization to another, so that this is a genuine stochastic quantity.

Coming back to the fluctuation theorem, one notes that the total entropy change is the sum of all the contributions: \( \Delta S_{\text{tot}} = \sum_i \Delta S^{(i)} + \Delta S \), which includes the entropy change \( \Delta S \) of the system. Since the entropy productions in the reservoirs are easier to monitor than the nonequilibrium stochastic system entropy \( \Delta S \) (where, for simplicity, we also avoid a discussion of the possible contribution to the entropy coming from interaction terms between system and reservoirs), it is of interest to identify situations in which the latter contribution is negligible. A good candidate is the asymptotic time regime for systems operating under steady-state nonequilibrium conditions. Indeed one intuitively expects that the heat (and/or particle) flows will be proportional to time, while the (stochastic) entropy of the system, being in a steady state, should remain more or less constant. Hence the latter contribution becomes negligible in the long-time limit. This was indeed proven to be the case for a class of systems with bounded energy.

One can then write

\[
\frac{P\left( \sum_i \Delta S^{(i)} \right)}{P\left( -\sum_i \Delta S^{(i)} \right)} \sim e^{\Delta S^{(i)}/k_B},
\]

known as the steady-state fluctuation theorem [1].

This asymptotic version of the fluctuation theorem can, however, break down when the energy of the system is unbounded [14]. A well documented example is that of a Brownian particle in contact with one or several heat reservoirs [15–19]. The large deviation function probes exponentially unlikely events for the heat evacuated to the reservoirs. Such an event can, however, be the result of an exponentially unlikely

\[
\frac{P\left( \sum_i \Delta S^{(i)} \right)}{P\left( -\sum_i \Delta S^{(i)} \right)} = \exp\left\{ \sum_i \Delta S^{(i)}/k_B \right\}.
\]
initial energy of the system. In this case the entropy contribution of the system is no longer negligible and a fluctuation theorem in terms of reservoir entropies alone breaks down.

Particle transport (both classical and quantum) between reservoirs connected through a finite system has attracted a great deal of interest more recently. In quantum systems, this particle exchange leads to an effective interaction between particles [12,20,21]. Thus, for instance, two fermions (e.g., electrons) avoid each other even if the Coulomb interaction between them is turned off. This property has been mapped (approximately) onto classical transport by modeling the particle dynamics within the symmetric exclusion process using hard-core interactions [22].

Here, we analyze noninteracting classical particle transport through a system in contact with particle reservoirs [23]. We evaluate analytically the large deviation function for the particle flux. We find that the asymptotic fluctuation theorem is satisfied under “natural” conditions, which are typical for the distribution of the number of particles. More precisely, whereas the Boltzmann factor allows for large energies, albeit in an exponentially unlikely way, the distribution \( P(n) \) for having \( n \) particles in a system typically decays faster than exponential, due to the indistinguishability property giving rise to a \( n! \) contribution in the denominator. As a result the generating function \( F(s) = \sum_n s^n P(n) \) is analytic in \( s \) in the entire complex plane, and the stochastic operator describing the particle exchange has a discrete spectrum. Under this condition, we are able to prove the validity of the steady-state fluctuation theorem. We note that our model is similar (except for a few details) to the one studied in Ref. [24], where it was shown that the steady-state fluctuation theorem for the particle flux breaks down. As discussed above, an exponential (Boltzmann) initial condition under certain conditions may contribute to the large deviations. This leads to the so-called breakdown of the fluctuation theorem. However, as shown in the following, due to particle indistinguishability the “natural” initial condition decays faster than an exponential and one thus recuperates the steady-state fluctuation relation for the particle flux.

When the system is coupled to particle reservoirs, a cumulated particle flux develops between the system and the reservoirs. The probability distribution of this flux typically depends on time in a complicated way. One may wonder whether there exist special initial conditions for which the functional form of this distribution does not change with time. We will identify such an initial distribution for the model under consideration. Note that, from the point of view of a detector is the number of single particle states with energy \( \epsilon \) decays more quickly than exponentially, due to the indistinguishability of particles, we expect this to be a genuine feature of a particle distribution. This leads to the so-called breakdown of the fluctuation theorem. However, as shown in the following, due to particle indistinguishability the “natural” initial condition decays faster than an exponential and one thus recuperates the steady-state fluctuation relation for the particle flux.

We start with a number of remarks. First, when in contact with a single bath \( i \), the steady-state probability solution of Eq. (2) reduces to the Poissonian equilibrium distribution \( P^{eq}(n) \):

\[
P^{eq}(n) = \frac{\beta^i_n}{n!} e^{-\beta^i},
\]

with a corresponding average number of particles \( \rho_i \) given by

\[
\rho_i = \frac{k_+^{(i)}}{k_-^{(i)}}.
\]

We mention for further use the relation between this equilibrium density and the reservoir properties \((\beta^i) = 1/k_B T^i\):

\[
\rho_i = g e^{-\beta^i (\epsilon - \mu_i)}.
\]

where \( g \) is the number of single particle states with energy \( \epsilon \) in the system and \( \mu_i \) is the chemical potential of the \( i \)th bath. Since we are dealing with the classical limit, \( g \) should be much larger than \( n \) and the exponential in Eq. (6) is much smaller than unity. Equation (6) is the detailed balance condition for the transport between the system and the \( i \)th reservoir.

Second, due to the combinatorial factor \( n! \), the probability distribution \( P^{eq}(n) \) decays more quickly than exponentially for large \( n \). Since there is a compelling physical reason for this factor, namely, the indistinguishability of particles, we expect this to be a genuine feature of a particle distribution function, and we will assume below a faster than exponential decay for the probability distribution even when operating under nonequilibrium conditions.
Third, we mention the following exact time-dependent solution of Eq. (2), namely, a propagating Poisson distribution:

\[ P(n; t) = \frac{[\rho(t)]^n e^{-\rho(t)}}{n!} \]  

(7)

with the mean number of particles in the system at time \( t \) given by

\[ \rho(t) = \frac{k_+}{k_-} + \left( \rho(0) - \frac{k_+}{k_-} \right) e^{-k_- t}. \]  

(8)

At steady state, \( \rho_{st} = k_+/k_- \). Hence, the particle distribution maintains a Poissonian equilibriumlike shape, even though it is in a nonequilibrium state.

We next turn to our main quest, namely, the study of the fluctuation theorem. We first identify the entropy change \( \Delta S^{(i)} \) in bath (i), for a given total elapsed time \( t \):

\[ \Delta S^{(i)} = \frac{Q^{(i)}}{T^{(i)}} = -\frac{(\epsilon - \mu^{(i)})N_i}{T^{(i)}} = k_B N_i \ln \frac{\rho_i}{g}. \]  

(9)

Here \( N_i \) is a register that adds (subtracts) 1 whenever a particle crosses from heat bath \( i \) to the system (from the system to heat bath \( i \)). Thus, \( N_i \) is the net number crossing from bath \( i \) to the system between time zero and \( t \). In going from the first to the second line, we have used the conservation of energy, with the change in bath energy \( -\epsilon N_i \) being equal to heat plus chemical energy, \( Q^{(i)} - \mu^{(i)} N_i \). Transition to the third line is based on Eq. (6).

The evaluation of the stochastic bath entropies is thus reduced to that of the number of particles \( N_i \) (\( N_i \in \mathbb{Z}, i = 1, 2 \)) transferred from baths to the system in time \( t \). Since these numbers are deterministic functions of the system dynamics, the enlarged set of variables \( n, N_1, N_2 \) again defines a Markov jump process, and the joint probability \( P(n, N_1, N_2; t) \) to find \( n \) particles in the system while having a cumulative transfer of \( N_1 \) and \( N_2 \) particles in time \( t \) evolves according to the following master equation:

\[ \frac{\partial}{\partial t} P(n, N_1, N_2; t) = k_+^{(1)} P(n - 1, N_1 - 1, N_2; t) + k_+^{(2)} P(n - 1, N_1, N_2 - 1; t) + (n + 1)k_+^{(1)} P(n + 1, N_1 + 1, N_2; t) + (n + 1)k_+^{(2)} P(n + 1, N_1, N_2 + 1; t) - (k_+ + nk_-) P(n, N_1, N_2; t). \]  

(10)

As we proceed to show, it is possible to find an exact time-propagating solution of this equation, which allows us to find the asymptotic large time properties, in particular those of the stochastic entropy. This solution furthermore illustrates how this asymptotic regime is reached in the course of time.

We note that, due to particle conservation, the following identity holds at all times:

\[ n(t) = N_1(t) - N_1(0) + N_2(t) - N_2(0) + n(0), \]  

(11)

where \( n(0), N_1(0), \) and \( N_2(0) \) are, respectively, the number of particles in the system and the number of particles on registers 1 and 2 at time \( t = 0 \). We make two observations, the significance of which will be revealed when discussing the time-propagating solution of Eq. (10). First, while it is quite natural and tempting, from the observer’s point of view, to choose \( N_1(0) = N_2(0) = 0 \), the choice of \( N_1(0) \) and \( N_2(0) \) is in principle free and could even be stochastic. Second, it follows from Eq. (11) that the condition \( n(0) = N_1(0) + N_2(0) \) propagates in time; i.e., it implies that \( n(t) = N_1(t) + N_2(t) \) holds at all times.

Finally, we note that, in view of Eq. (11), we can write \( P(n, N_1, N_2; t) = P(N_1 + N_2 - M(0), N_1, N_2; t) \), where \( M(0) = N_1(0) + N_2(0) - n(0) \). At this point, one might be tempted to express \( P(n, N_1, N_2; t) \) simply as \( P(N_1, N_2; t) \). However this is not correct, as it would imply integration over all possible values of \( M(0) \).

### III. Generating Function

The solution of the master equation is facilitated by switching to the following generating function:

\[ F_s(n, t) = \sum_{N_1, N_2 = -\infty}^{\infty} \sum_{n=0}^{\infty} e^{\lambda_s N_1 + \beta_s N_2} s^n P(n, N_1, N_2; t). \]  

(12)

The parameters \( \lambda = \{\lambda_1, \lambda_2\} \) are so-called counting parameters that keep track of the net number of particles transferred between the system and the corresponding reservoirs. We shall use \( s \) to denote a dependence on \( \lambda_1 \) and \( \lambda_2 \).

By combination with Eq. (10) we find

\[ \frac{\partial}{\partial t} F_s(n, t) = \mathcal{L}_s F_s(n, t), \]  

(13)

where the operator \( \mathcal{L} \) is defined as

\[ \mathcal{L}_s = k_+ \alpha_s s + k_- \beta_s \frac{\partial}{\partial s} - k_- \frac{\partial}{\partial s} - k_+. \]  

(14)

with

\[ \alpha_s = \frac{k_+^{(1)} e^{\lambda_1} + k_+^{(2)} e^{\lambda_2}}{k_-} \]  

(15)

\[ \beta_s = \frac{k_-^{(1)} e^{-\lambda_1} + k_-^{(2)} e^{-\lambda_2}}{k_-}. \]  

(16)

Since the dependence on the counting parameters in Eq. (13) is parametric, it suffices to evaluate the eigenvectors and eigenvalues of \( \mathcal{L} \) as operators with respect to the variable \( s \). Let \( \Psi_s(s) \) be an eigenvector of \( \mathcal{L}_s \), s \) with eigenvalue \( \xi_s \):

\[ \mathcal{L}_s(s) \Psi_s(s) = \xi_s \Psi_s(s). \]  

(17)

With the expression Eq. (14) for the operator, the eigenfunctions are found by straightforward integration:

\[ \Psi_s(s) = (s - \beta_s)^{\alpha_s} \exp \left[ \alpha_s (s - \beta_s) \right]. \]  

(18)

where

\[ g_s = \alpha_s \beta_s - \frac{k_+ + \xi_s}{k_-}. \]  

(19)
We now make the following crucial assumption, already mentioned in the introduction: we request that the eigenfunctions be analytic in the variable $s$ in the entire complex plane. Analyticity imposes two restrictions on the exponent $g_k$: it must be greater than or equal to zero, $g_k \geq 0$, and it must be an integer. Hence, setting $g_k = l$ with $l \in \mathbb{N}$ we find from Eq. (19) for the eigenspectrum of the operator $L_k$ that

$$\xi_k^{(l)} = k_- \alpha_k \beta_k - k_+ - lk_-, \quad l \in \mathbb{N}. \quad (20)$$

The corresponding eigenfunction can now be written in the following compact way:

$$\Psi_k^{(l)}(s) = \frac{\partial^l}{\partial \alpha_k^l} \exp \left[ \alpha_k (s - \beta_k) \right]. \quad (21)$$

At this point, we make two observations. First, the eigenvalue $\xi_k^{(l)}$ depends on $\lambda$ only via $\lambda = \lambda_1 - \lambda_2$ and is invariant under the following interchanges:

$$\lambda \leftrightarrow -\lambda - \ln \frac{\rho_1}{\rho_2}, \quad (22)$$

$$\lambda_i \leftrightarrow -\lambda_i - \ln (\rho_i) \quad \text{for} \quad i = 1 \text{ and } 2. \quad (23)$$

The eigenfunctions themselves, however, do not obey this symmetry. This symmetry arises due to the constraint imposed by Eq. (5) on the rates with which particles can enter and exit the system. This condition is not arbitrary, and together with Eq. (6) it is required to have a unique steady state. This symmetry property will be crucial to verify the steady-state fluctuation theorem.

Second, the above set of eigenfunctions is complete in the sense that any analytic function of $s$ can be expanded in terms of this basis. Furthermore, the expansion is unique as it corresponds to a Taylor expansion around the point $\beta_k$. Hence we obtain the following explicit expression for the generating function (assumed to be analytic in $s$) obeying Eq. (13):

$$F_k(s; t) = \sum_{l=0}^{\infty} a_k^{(l)} \Psi_k^{(l)}(s) e^{\gamma_k^{(l)}}, \quad (24)$$

where $a_k^{(l)}$ is an expansion coefficient of the eigenfunction $\Psi_k^{(l)}(s)$ for the initial function $F_k(s; 0)$. In the sequel, we will focus on a particular simple initial condition, corresponding to $a_k^{(0)} = \delta_k^{(0)}$, where $\delta_k$ is the Kronecker delta. One reason is obvious: the corresponding eigenfunction is dominating the long-time limit, since it has the lowest eigenvalue [see Eq. (20)]. The other reason is that such an initial condition corresponds, for an appropriate choice of the coefficient $a_k^{(0)}$, to a genuine probability distribution. The explicit expression for $\Psi_k^{(l)}(s)$ suggests the following choice for $a_k^{(0)}$:

$$a_k^{(0)} = e^{\alpha_k \beta_k - k_- / k_+}, \quad (25)$$

where the subtraction of $k_- / k_+$ guarantees normalization. Referring to Appendix A for the calculation of the inverse, we find that it leads to the following initial probability distribution:

$$P(n, N_1, N_2; t = 0) = e^{-\rho s} \frac{k_+^{(1)} N_1 \kappa_+^{(2)} N_2}{N_1! N_2!} \times \delta_{n, N_1 + N_2} \Theta(N_1) \Theta(N_2), \quad (26)$$

where $\Theta(x)$ is a Heaviside theta function. The Kronecker delta in Eq. (26) imposes the condition that initially $n(0) = N_1(0) + N_2(0)$. This condition was to be expected since, as mentioned earlier, it propagates in time with $n = N_1 + N_2$ at all times. The corresponding reduced distribution for the number of particles in the system, $P^R(n)$, is, as expected, the steady-state distribution [see Eq. (7)]:

$$P^R(n) \equiv P(n; t = 0) = \frac{\rho^n e^{-\rho}}{n!}, \quad (27)$$

with $\rho = k_+ / k_-$. The reduced distribution for the initial cumulated particle transfer $P(N_1, N_2; t = 0)$ (see also Fig. 1) is obtained by summation of Eq. (26) over $n$. The summation only affects the Kronecker delta; hence $P(N_1, N_2; t = 0)$ is obtained from Eq. (26) by the replacement of the Kronecker delta $\delta_{n, N_1 + N_2}$ with $\Theta(N_1 + N_2)$, but this factor is superfluous due to the presence of $\Theta(N_1) \Theta(N_2)$. We conclude that the reduced initial distribution $P(N_1, N_2; t = 0)$ is a product of two independent Poissonian distributions. One can verify that the “natural initial condition” $P(N_1, N_2; t = 0) = \delta_{N_1, 0} \delta_{N_2, 0}$ does not lead to a time propagating solution; that is, it cannot be expressed solely in terms of the eigenvector $\Psi_k^{(l)}(s)$. This could have been anticipated from the fact that the propagating condition $n(0) = N_1(0) + N_2(0)$ would then imply $n(0) = 0$, which is incompatible with the “propagating” steady-state statistics $P^R(n)$ for $n$.

IV. PROPAGATING SOLUTION AND LARGE DEVIATION FUNCTION

Our main focus is the evaluation of the joint reduced distribution, $P(N_1, N_2; t)$. Its generating function $F_k(t)$ is
found by setting \( s = 1 \) in Eq. (24):

\[
F_{\lambda}(t) = \sum_{N_1, N_2 = -\infty}^{\infty} e^{\lambda(N_1+\lambda_2 N_2)} P(N_1, N_2; t)
\]

\[
= \sum_{l=0}^{\infty} d_{\lambda}^{(l)} \Psi_{\lambda}^{(l)}(s = 1) e^{\lambda t_0}.
\]

The \( l = 0 \) term dominates the series in Eq. (24) for asymptotically long times \( t \to \infty \). Alternatively, this term corresponds to the full solution at all times for the initial condition identified in the previous section. We henceforth consider this case and can thus write

\[
F_{\lambda}(t) = d_{\lambda}^{(0)} e^{\lambda t_0} \Psi_{\lambda}^{(0)}(s = 1),
\]

(30)

with \( d_{\lambda}^{(0)} \) given by Eq. (25). The corresponding joint probability \( P(N_1, N_2; t) \) is computed by taking the inverse transform of Eq. (30):

\[
P(N_1, N_2; t) = \sum_{m=0}^{\infty} \frac{1}{m!} \frac{1}{k_-^m} \oint \frac{d\lambda_1}{2\pi i} \oint \frac{d\lambda_2}{2\pi i} e^{\lambda(N_1+\lambda_2 N_2)}
\]

\[
\times d_{\lambda}^{(0)} \Psi_{\lambda}^{(0)}(s = 1).
\]

(31)

Switching to the integration variable \( \lambda = \lambda_1 - \lambda_2 \), and expanding the exponential of \( \alpha_j \) which appears inside \( \Psi_{\lambda}^{(0)}(s = 1) \) [see Eq. (21)], Eq. (31) can be rewritten as

\[
P(N_1, N_2; t) = \sum_{m=0}^{\infty} \frac{1}{m!} \frac{1}{k_-^m} \oint \frac{d\lambda}{2\pi i} (k_-^{(1)} + k_-^{(2)} e^{-\lambda} \rho e^{\lambda(N_1+\lambda_2 N_2)}) e^{-k_-/k_-}.
\]

(32)

Since \( \xi_{\lambda}^{(0)} \) and \( \alpha_j \beta_\lambda \) are functions of only \( \lambda \), the integral over \( \lambda \) reduces to the Kronecker delta \( \delta_{k_-, N_1+N_2} \). Expanding the exponential in \( \alpha_j \beta_\lambda \), the remaining integral over \( \lambda \) can be performed. Following the same steps which led to Eq. (26), we obtain the following propagating solution for the joint distribution function:

\[
P(N_1, N_2; t) = e^{-t \left( k_+^{(1)} + k_-^{(2)} \rho \right) / k_-} e^{-k_-/k_-}
\]

\[
\times \frac{1}{(N_1+N_2)!} \frac{k_-^{(1)}}{k_-^{(2)}} \frac{N_1}{N_2}
\]

\[
\times \frac{N_1+N_2}{N_1+N_2} \sum_{m=0}^{\infty} \binom{m}{m} \frac{k_-^{(2)}}{k_-^{(1)}} \left( k_-^{(1)} k_-^{(2)} \rho \right) e^{-k_-/k_-}
\]

\[
\times I_{N_1+N_2} (x t) \Theta(N_1+N_2),
\]

(33)

where

\[
x = \frac{2}{k_-} \sqrt{k_-^{(1)} k_-^{(2)} k_-^{(1)} k_-^{(2)}}
\]

(34)

and \( I_n(y) \) is the modified Bessel function of the first kind of order \( n \). It is straightforward to check that for \( t = 0 \) this reduces to the product of Poissonians [see Eq. (26)]. The distribution function Eq. (33) is shown in Fig. 2 for different values of \( t \).

\[\text{FIG. 2. (Color online) Probability distribution function, } P(N_1, N_2; t), \text{ for different times, } t = 1, 10, \text{ and } 100 \text{ (top to bottom). As time increases, the distribution becomes more peaked around the second diagonal, illustrating the convergence of } j_1 = N_1/t \text{ to } -j_2 = -N_2/t.\]

We next focus on the large \( t \) limit. Since the particle fluxes \( N_1 \) and \( N_2 \) diverge for \( t \to \infty \), we introduce the fluxes per unit time \( j_1 = N_1/t \). In this limit, an additional simplification takes place as one finds asymptotically that the stochastic quantities \( j_1 \) and \( j_2 \) become identical (see Fig. 2). Hence, the statistics in the long-time limit are expressed in terms of a single flux \( j_1 = -j_2 = j \). The corresponding probability distribution function for \( j \) has the typical shape from large deviation theory, namely,

\[
P(j; t) \sim e^{-t \mathcal{L}(j)},
\]

(35)

with the large deviation function

\[
\mathcal{L}(j) = -\lim_{t \to \infty} \frac{1}{t} \ln P(j; t)
\]

(36)

given by the following convex non-negative function (see Appendix B):

\[
\mathcal{L}(j) = \frac{k_-^{(1)} k_-^{(2)} + k_-^{(2)} k_-^{(1)}}{k_-} - \sqrt{x^2 + j^2}
\]

\[
+ \frac{j}{2} \ln \left( \rho_2 \sqrt{x^2 + j^2} + j \right) \left( \rho_1 \sqrt{x^2 + j^2} - j \right)
\]

(37)

From Eq. (33), the probability \( P(N, t) \) to have \( N_1 = -N_2 = N \) at time \( t \) takes a simple form:

\[
P(N; t) = e^{-t \left( k_+^{(1)} + k_-^{(2)} \rho \right) / k_-} \left( \frac{k_-^{(1)}}{k_-^{(2)}} \right)^{N/2} e^{-k_-/k_-} I_N (x t).
\]

(38)
The extremum is found at
\[ \lambda(j) = \frac{1}{2} \ln \left( \frac{\rho_2 \sqrt{j^2 + \lambda^2 + j}}{\rho_1 \sqrt{j^2 + \lambda^2 - j}} \right). \]  
(43)

Substituting this result in Eq. (42), we recover Eq. (37).

The cumulant generating function \( G_\lambda \) allows for a swift calculation of the cumulants of the current. For large times, the \( n \)-th cumulant \( \kappa_n(N_1) \) of \( N_1 \), the net number of particles transferred between the system and the reservoir by time \( t \), is obtained from \( G_\lambda \) as
\[ \kappa_n(N_1) = t \frac{\partial^n G_\lambda}{\partial \lambda^n} \bigg|_{\lambda=0}. \]  
(44)

Thus, according to Eq. (44), all cumulants \( \kappa_n(N_1) \) vary linearly with times. For particle current \( j \), the average and the variance are given by
\[ \kappa_1(j) = \frac{1}{k_-}(k_+^{(1)}k_-^{(2)} - k_+^{(2)}k_-^{(1)}), \]  
(45)
\[ \kappa_2(j) = \frac{1}{2k_-}(k_+^{(1)}k_-^{(3)} + k_+^{(3)}k_-^{(1)}). \]  
(46)

Furthermore, all odd and even cumulants of \( j \) are proportional to the first and second cumulants, respectively:
\[ \kappa_n(j) = \frac{\kappa_2(j)}{t^{n-2}}, \quad n \text{ even}, \]  
(47)
\[ \kappa_n(j) = \frac{\kappa_1(j)}{t^{n-1}}, \quad n \text{ odd}. \]  
(48)

V. FLUCTUATION THEOREM

From Eq. (9), we find that the total entropy production in the reservoirs (divided by \( k_B \)) is asymptotically given by
\[ \frac{\Sigma_i \Delta S^{(i)}}{k_B} = \Sigma_i N_i \ln \frac{\rho_i}{\rho} \sim t j \ln \frac{\rho_1}{\rho_2}, \]  
(49)
where we used the fact that asymptotically \( j = j_1 = -j_2 \) \( (N_i = t j_i) \). The steady-state fluctuation theorem then requires that
\[ \frac{P(j)}{P(-j)} \sim e^{t j \ln \frac{\rho_1}{\rho_2}}, \]  
(50)
or, more precisely, that the large deviation function, Eq. (37), obeys the symmetry relation,
\[ \mathcal{L}(j) - \mathcal{L}(-j) = j \ln \frac{\rho_2}{\rho_1}, \]  
(51)
which is easily verified by Eq. (37). This symmetry of the large deviation function implies an analogous symmetry for the cumulant generating function (see Fig. 4):
\[ G_\lambda = G_{-\lambda - \ln(\rho_2/\rho_1)} \]  
(52)

We recall that the analyticity of the generating function for the particle number \( n \) is an essential assumption in the above derivation of the steady-state fluctuation theorem. This requires that the corresponding probability \( P(n) \) decay faster.
than an exponential in $n$. This property is verified by the steady-state Poisson distribution, Eq. (7), which decays (for large $n$) logarithmically faster than the exponential: $P(n) \sim e^{-\alpha n}$, for $n \gg 1$. It is therefore quite natural to assume that the initial condition satisfies the same property, that is, that it decays faster than an exponential. Without this assumption, exponentially rare fluctuations in the initial particle distribution will lead to a breakdown of the fluctuation theorem.

We finally mention that the large deviation function Eq. (37) is identical to that for a random walker on a line with jump rates $j_{1}^{(1)}k^{(2)}/k_{-}$ to the right and $k^{(2)}j_{-}/k_{-}$ to the left. The physical interpretation is clear. Since the probability distribution of the number of particles contained in the system decays faster than an exponential, the large deviation statistics is essentially described by the transfer statistics between the reservoirs only [cf. the asymptotic identities of $j_{1}$ and $-j_{2}$]. It is intuitively clear that this long-time process will be identical to the asymptotic properties of a random walk. An identical result is obtained for the effusion of particles between two reservoirs connected through a small opening [25].

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**APPENDIX A: DERIVATION OF EQ. (26)**

In order to compute the initial probability distribution for which only the term with $l = 0$ in the series Eq. (24) survives, we need to inverse transform $a_{0}^{(0)}(s_{\alpha})\Psi_{\alpha}(s)$, where we choose $a_{0}^{(0)} = e^{-\lambda_{1}k_{-}/k_{+}}$. Note that, for $\lambda = 0$, $a_{0}^{(0)} = 1$ to preserve the normalization of the probability distribution. This is not the only possible choice for $a_{0}^{(0)}$, however this choice leads to a simple natural initial Poissonian distribution [see Eq. (26)], which propagates in time. Thus we have

\[
P(n, N_{1}, N_{2}; t = 0) = \int \frac{ds}{2\pi i} s^{n-1} \int \frac{d\lambda_{1}}{2\pi i} e^{-\lambda_{1}N_{1}}
\]

\[
\times \int \frac{d\lambda_{2}}{2\pi i} e^{-\lambda_{2}N_{2}} a_{0}^{(0)}(s)\Psi_{\lambda}(s).
\]  

(A1)

Substituting for $\Psi_{\lambda}(s)$ from Eq. (21), we get

\[
P(n, N_{1}, N_{2}; t = 0) = \int \frac{ds}{2\pi i} s^{n-1} \int \frac{d\lambda_{1}}{2\pi i} e^{-\lambda_{1}N_{1}}
\]

\[
\times \int \frac{d\lambda_{2}}{2\pi i} e^{-\lambda_{2}N_{2}} e^{s\lambda_{1}} e^{-\lambda_{2}k_{-}/k_{+}}.
\]  

(A2)

Next we expand the exponential which contains the variable $s$. This allows us to perform the $s$ integral and gives the Kronecker delta function $\delta_{n,m}$. We find

\[
P(n, N_{1}, N_{2}; t = 0) = \frac{1}{n!} \int \frac{d\lambda_{1}}{2\pi i} e^{-\lambda_{1}N_{1}}
\]

\[
\times \int \frac{d\lambda_{2}}{2\pi i} a_{0}^{(0)} e^{-\lambda_{2}N_{2}} e^{-\lambda_{1}k_{-}/k_{+}}.
\]  

(A3)

Using $\alpha_{\lambda}$ from Eq. (15), and expanding $\alpha_{\lambda}^{e}$ using binomial expansion, we obtain

\[
P(n, N_{1}, N_{2}; t = 0) = e^{-\frac{1}{\lambda_{1}}\left(\sum_{l=0}^{n} \frac{(\lambda_{1})^{l}}{l!} \left(\frac{k_{+}}{k_{-}}\right)^{l}\right)}
\]

\[
\times \int \frac{d\lambda_{1}}{2\pi i} e^{\lambda_{1}(s-l-N_{1})} \int \frac{d\lambda_{2}}{2\pi i} e^{\lambda_{2}(s-l-N_{2})}.
\]  

(A4)

The integrals over $\lambda_{1}$ and $\lambda_{2}$ give Kronecker deltas, $\delta_{n,N_{1}}$ and $\delta_{N_{2}}$, respectively. Using these in Eq. (A4), we get (for $n = N_{1} + N_{2}$) Eq. (26).

An alternative method to obtain the probability distribution is to expand the generating function $F_{\lambda}$ and compare it term by term with the definition Eq. (12). Here we present this method to recover Eq. (26). At $t = 0$, the generating function $F_{\lambda}$ is (keeping only the $l = 0$ term)

\[
F_{\lambda}(s; 0) = a_{0}^{(0)}(s)\Psi_{\lambda}^{(0)}(s).
\]  

(A5)

Substituting for $a_{0}^{(0)}(s)$ and $\Psi_{\lambda}^{(0)}(s)$, we can re-express it as

\[
F_{\lambda}(s; 0) = e^{-\frac{1}{\lambda_{1}}s}\sum_{n=0}^{\infty} \frac{s^{n}}{n!} a_{0}^{(0)}(s)
\]

\[
= e^{-\frac{1}{\lambda_{1}}s}\sum_{n=0}^{\infty} \frac{s^{n}}{n!} \left(\frac{k_{+}}{k_{-}}\right)^{n}
\]

\[
\times \sum_{m=0}^{n} \frac{n}{m} \left(\frac{k_{+}}{k_{-}}\right)^{m} e^{(s-m)\lambda_{1}N_{1}} e^{m\lambda_{2}}.
\]  

(A6)

Since $m$ and $n$ are dummy variables, we can rewrite the last line as

\[
F_{\lambda}(s; 0) = e^{-\frac{1}{\lambda_{1}}s}\sum_{n=0}^{\infty} \sum_{N_{1}=0}^{\infty} \frac{s^{n}}{n!} \left(\frac{k_{+}}{k_{-}}\right)^{n}
\]

\[
\times \left(\frac{n}{N_{2}}\right) e^{N_{2}\lambda_{1}N_{1}} e^{N_{2}\lambda_{2}}.
\]  

(A7)

In order to put it in a convenient form which will allow an easy comparison with Eq. (12), we introduce a Kronecker delta $\delta_{n-N_{1},N_{2}}$. This allows us to rewrite Eq. (A7) as

\[
F_{\lambda}(s; 0) = e^{-\frac{1}{\lambda_{1}}s}\sum_{n=0}^{\infty} \sum_{N_{1}=0}^{\infty} \sum_{N_{2}=0}^{\infty} \frac{s^{n}}{n!} \left(\frac{k_{+}}{k_{-}}\right)^{n}
\]

\[
\times \left(\frac{n}{N_{2}}\right) e^{N_{2}\lambda_{1}N_{1}} e^{N_{2}\lambda_{2}} \delta_{n-N_{1},N_{2}}.
\]  

(A8)

Finally, rearranging the Kronecker delta and using the fact that due to the binomial coefficients all terms for $N_{2} < 0$ and $n < N_{2}$ vanish, we can recast this expression as

\[
F_{\lambda}(s; 0) = e^{-\frac{1}{\lambda_{1}}s}\sum_{n=0}^{\infty} \sum_{N_{1}=0}^{\infty} \sum_{N_{2}=0}^{\infty} \frac{s^{n}}{N_{1}!N_{2}!} \left(\frac{k_{+}}{k_{-}}\right)^{n}
\]

\[
\times \left(\frac{n}{N_{2}}\right) e^{N_{2}\lambda_{1}N_{1}} e^{N_{2}\lambda_{2}} \delta_{n,N_{1}+N_{2}}.
\]  

(A9)
Comparing this with Eq. (12), we recover Eq. (26). Similar steps can be followed to obtain the time-dependent joint distribution function given in Eq. (33).

**APPENDIX B: CALCULATION OF THE LARGE DEVIATION FUNCTION, EQ. (37)**

The large deviation function is defined in Eq. (36). We thus need to take the long-time limit \( t \to \infty \) of Eq. (37) after substituting \( N_1/t = -N_2/t = j \). This gives

\[
\mathcal{L}(j) = \frac{k_2^{(1)}}{k_1^{(1)}} + \frac{k_2^{(2)}}{k_1^{(2)}} - \frac{j}{2} \ln \left( \frac{k_2^{(1)} k_2^{(2)}}{k_1^{(1)} k_1^{(2)}} \right) - \lim_{t \to \infty} \ln I_{j,t}(x). \tag{B1}
\]

In order to compute the limit of the Bessel function, it is useful to express the Bessel function \( I_p(y) \) as

\[
I_p(y) = \sum_{k=0}^{\infty} e^{f_p(y,k)}, \tag{B2}
\]

where

\[
f_p(y,k) = \ln \left( \frac{(y/2)^{2k+p}}{k!(k+p)!} \right). \tag{B3}
\]

We note that, for large \( y \), the function \( f_p(y,k) \) has a maximum at \( k \gg 1 \). This maximum occurs at

\[
k^* = \frac{j}{2} \sqrt{x^2 + y^2 - p}. \tag{B4}
\]

As \( y \) increases, the sum over \( k \) in Eq. (B2) is dominated by the term \( f_p(y,k^*) \). Then, as \( y \to \infty \), we can expand \( f_p(y,k) \) around \( k = k^* \) and replace the summation with an integral over \( k \) [for large \( k^* \), the function \( f_p(y,k) \) can be approximated as a continuous function of \( k \)]. Assuming that the function decays quickly as we move away from \( k = k^* \), we can keep only up to quadratic terms in the Taylor expansion. This results in the following approximate expression for the Bessel function (for large \( p \)):

\[
I_p(y) \approx e^{f_p(y,k^*)} \frac{\pi}{2|f_p'(y,k^*)|} \times \left[ 1 + \text{erf} \left( k^* \sqrt{\frac{|f_p'(y,k^*)|}{2}} \right) \right], \tag{B5}
\]

where \( f_p'(y,k^*) = \frac{\partial^2 f_p(y,k)}{\partial k^2} |_{k=k^*} \) and \( \text{erf}(z) \) is the Gaussian error function.

In Eq. (B1), \( p = jt \) and \( y = xt \), both vary linearly with \( t \). In the limit \( t \to \infty \), \( k^* \to \infty \) linearly in time and the error function \( \to 0 \). Thus, in the \( t \to \infty \) limit, we get

\[
\lim_{t \to \infty} \frac{1}{t} \ln I_{j,t}(x) = \frac{1}{t} f_{j,t}(xt,k^*). \tag{B6}
\]

Substituting \( k^* \) and \( y = xt \) in Eq. (B3), we get

\[
f_{j,t}(xt,k^*) = (2k^* + tj) \ln \left( \frac{xt}{2} \right) - k^* \ln(k^*) - (k^* + tj) \ln(k^* + tj) + 2k^* + tj. \tag{B7}
\]

Using Eqs. (B6)–(B8), we get

\[
\lim_{t \to \infty} \frac{1}{t} \ln I_{j,t}(x) = \sqrt{j^2 + x^2} + \frac{j}{2} \ln \left( \frac{\sqrt{j^2 + x^2} - j}{\sqrt{j^2 + x^2} + j} \right). \tag{B9}
\]

Substituting this in Eq. (B1), we recover Eq. (37).