Fluctuation relation for heat exchange in Markovian open quantum systems

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A fluctuation relation for the heat exchange of an open quantum system under a thermalizing Markovian dynamics is derived. We show that the probability that the system absorbs an amount of heat from its bath, at a given time interval, divided by the probability of the reverse process (releasing the same amount of heat to the bath) is given by an exponential factor which depends on the amount of heat and the difference between the temperatures of the system and the bath. Interestingly, this relation is akin to the standard form of the fluctuation relation (for forward-backward dynamics). We also argue that the probability of the violation of the second law of thermodynamics in the form of the Clausius statement (i.e., net heat transfer from a cold system to its hot bath) drops exponentially with both the amount of heat and the temperature differences of the baths.

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I. INTRODUCTION

The irreversibility of dynamics is a ubiquitous feature of macroscopic systems, which appears despite microscopic reversibility in classical and quantum systems [1–4]. The relation between these macroscopic and microscopic features can be captured by "fluctuation relations" (FRs) [5–10]. For example, for a Markovian process, where the initial and final states of the system are thermal (with inverse temperature $\beta = 1/T$, taking the Boltzmann constant $k_B \equiv 1$), the probability of doing work $\mathbb{W}$ on the system in the *forward* path is related to the corresponding probability of doing work $-\mathbb{W}$ in the *reverse* path, through [5]

$$\frac{P_{\mathrm{F}}(+\mathbb{W})}{P_{\mathrm{R}}(-\mathbb{W})} = e^{\beta(\mathbb{W}-\Delta\mathbb{F})}, \tag{1}$$

where $\Delta\mathbb{F}$ is the difference in the free energy of the system. A similar relation has been shown to govern heat exchange $\mathbb{Q}$ in the forward and reverse paths between two weakly coupled systems S and B (from B to S), with the difference $\Delta\beta = \beta_S - \beta_B$ in their inverse temperatures [10–14],

$$\frac{P_{\mathrm{F}}(+\mathbb{Q})}{P_{\mathrm{R}}(-\mathbb{Q})} = e^{\mathbb{Q}\Delta\beta}. \tag{2}$$

The distribution function of any quantity in the forward and reverse paths depends on two factors. One is the probability of the initial state of the system, which is determined by preparation; the other is the probability of the path, which is determined by dynamics. For a *closed* system (where the associated dynamics is generated completely by a Hamiltonian through the Schrödinger equation), due to the time reversibility of the dynamics, the path probabilities are equal in the forward and reverse cases such that they cancel each other out when we calculate the ratio of the distributions [10]. As a result, dynamics seems to play no explicit role in deriving FRs.

The dynamics of an *open* system (where the system is coupled to an environment which affects its behavior [15]) is not necessarily time reversible, so the path probabilities in the forward and reverse paths are not the same. But in what follows we show that a similar FR (in some sense) is attainable. A principal question for open systems is what a *reverse* dynamics physically means. Although there has been some progress towards defining reverse dynamics [16–18]), an unambiguous definition has been elusive thus far. In order to avoid this issue, here we simply replace the notion of reverse *dynamics* with reverse *process*. If in the forward dynamics, the system releases heat $\mathbb{Q}$, the reverse process corresponds to absorbing heat $\mathbb{Q}$. This setting helps us express our finding fully for the forward path.

Such a relation may have its own appeal and advantages. For example, a prototypical example for the irreversibility inherent within the second law is that one cannot unbreak a broken egg. In this example there seems to be no explicit mention of backward dynamics; even if we wait for a long time (i.e., in the forward direction of time), we will almost never see the reverse event. This observation implies that (forward) dynamics does hardly feature a violation of the second law of thermodynamics. What we do in this paper is to provide a rigorous formulation of this observation for a fairly general class of open-system quantum dynamics.

For a system S prepared in a thermal state of temperature $T_S = 1/\beta_S$, and then put in contact for time τ with a heat bath of temperature $T_B = 1/\beta_B$, where the dynamics of the system is given by a thermalizing Lindblad equation (i.e., the dynamics drives the system to become thermal with the bath in a sufficiently long time), we can show that the following FR holds,

$$\frac{P_{\mathrm{F}}(+\mathbb{Q},\tau)}{P_{\mathrm{F}}(-\mathbb{Q},\tau)} = e^{\mathbb{Q}\Delta\beta}, \tag{3}$$

where $P_{\mathrm{F}}(\pm\mathbb{Q},\tau)$ is the probability that the system *absorbs* (*releases*) heat $\mathbb{Q}$ from (to) the heat bath in the time interval τ in the forward path (see Fig. 1). An interesting feature of our result is that although both $P_{\mathrm{F}}(+\mathbb{Q},\tau)$ and $P_{\mathrm{F}}(-\mathbb{Q},\tau)$ are time-dependent and transient expressions, our FR relation indicates that their ratio is indeed time independent. In the

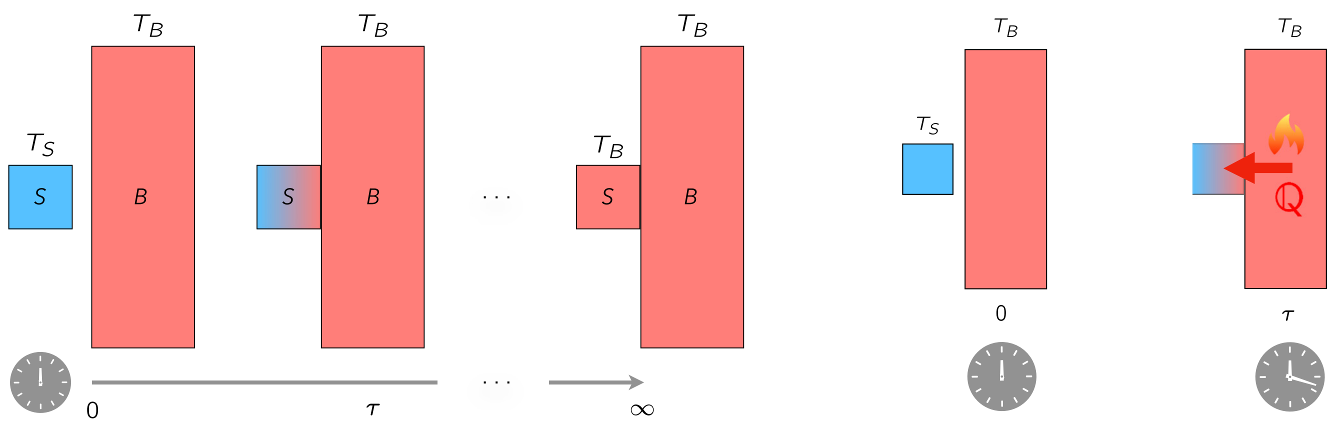

FIG. 1. Left: Schematic of thermalization of system S (with initial temperature T_S) in contact with a bath B with temperature T_B. Right: Schematic of the process where at time τ system S absorbs heat $\mathbb{Q}$ from bath B.

following sections, after reviewing a fairly general model for thermalizing Markovian dynamics, we give the proof of our FR. In what follows, we omit "F" hereon in order to simplify the notation.

II. THERMALIZING DYNAMICS OF AN OPEN QUANTUM SYSTEM

Consider a quantum system with a D-dimensional Hilbert space, and let H_0 be its free Hamiltonian and the set $\{|m\rangle\}$ indicates the eigenvectors corresponding to the discrete and nondegenerate eigenvalues $E_m^{(0)}$ ($H_0|m\rangle = E_m^{(0)}|m\rangle$), ordered increasingly as $E_1^{(0)} < E_2^{(0)} < \cdots < E_D^{(0)}$. Assume that the system is then put in contact with a heat bath (reservoir or environment) of inverse temperature β_B and that the dynamics of the system is described (within the weak-coupling and Markovian approximation) by the Lindblad equation [19,20]

$$\frac{d\varrho}{d\tau} = -i[H,\varrho] + \sum_a \left(L_a \varrho L_a^\dagger - \frac{1}{2}\{L_a^\dagger L_a, \varrho\}\right), \quad (4)$$

where we have set $\hbar \equiv 1$, $\varrho(\tau)$ is the density matrix of the system, H representing an effective Hamiltonian (usually different from H_0), and L_a's are the quantum jump operators induced by the interaction with the bath (whose number can always be reduced to $D^2 - 1$).

Conditions under which a Lindbladian dynamics can yield a stationary state which is a thermal equilibrium state in a Gibbsian form have been studied extensively in the literature [3,4,21–23]. Here, we in particular follow the formalism and the framework laid out recently in Ref. [21]. The structure of the evolution (4) should meet the following conditions: (i) Consider

$$H = \sum_m E_m |m\rangle\langle m|, \quad (5)$$

that is, the eigenvectors of the isolated system $\{|m\rangle\}$ are the eigenvectors of the effective Hamiltonian, too. (ii) Choose L_a's as the jump operators among all different energy levels of the isolated system, i.e.,

$$L_{mn} = l_{mn}|m\rangle\langle n|, \quad (6)$$

for $m \neq n$ (implying forbidden $|n\rangle \to |n\rangle$ transitions), where l_{mn}'s fulfill the "detailed balance" condition

$$|l_{mn}|^2 = C_{mn} e^{-\beta_B (E_m^{(0)} - E_n^{(0)})/2}, \quad (7)$$

in which $C_{mn} = C_{nm} > 0$ and C_{mn}'s depend on the interaction of the system and the heat bath. These conditions—along with the nondegeneracy of the spectrum of H_0—have been shown to guarantee the existence of a unique stationary solution of thermal (Gibbsian) form $\varrho^{(\text{eq})} = e^{-\beta_B H_0}/\text{Tr}[e^{-\beta_B H_0}]$.

Defining the vector $|\mathbf{v}(\tau)\rangle = \sum_m \varrho_{mm}(\tau)|m\rangle$, we have the following evolution:

$$|\mathbf{v}(\tau)\rangle = e^{-\tau A}|\mathbf{v}(0)\rangle, \quad (8)$$

where $A = \sum_{mn} A_{mn}|m\rangle\langle n|$ is defined as

$$A_{mn} = \begin{cases} \sum_{j\neq m} |l_{jm}|^2, & m = n, \\ -|l_{mn}|^2, & m \neq n. \end{cases} \quad (9)$$

The matrix A is diagonalizable with non-negative real eigenvalues, with the minimum value being zero and nondegenerate (valid for typical nondegenerate H_0's). Additionally, it can also be seen that the off-diagonal elements of ϱ evolve independently as

$$\varrho_{mn}(\tau) = e^{-(i\omega_{mn}+\gamma_{mn})\tau}\varrho_{mn}(0), \quad (10)$$

where $\omega_{mn} = E_m - E_n$ is the gap of the effective Hamiltonian and $\gamma_{mn} = (1/2)\sum_j (|l_{jm}|^2 + |l_{jn}|^2) \geqslant 0$ represents the decay rate. Thus we obtain

$$\varrho(\tau) = \sum_{nm} \varrho_{nn}(0)\langle m|e^{-\tau A}|n\rangle |m\rangle\langle m| + \sum_{m\neq n} e^{-(i\omega_{mn}+\gamma_{mn})\tau}\varrho_{mn}(0)|m\rangle\langle n|. \quad (11)$$

III. FORWARD FLUCTUATION RELATION

Consider a system prepared initially in a thermal state of inverse temperature β_S, $\varrho(0) = e^{-\beta_S H_0}/\text{Tr}[e^{-\beta_S H_0}]$, which is brought into contact with a heat bath of inverse temperature β_B. Suppose that the dynamics of the system is given by the Lindblad equation (4) and after a sufficiently long time it reaches a thermal state of the inverse temperature

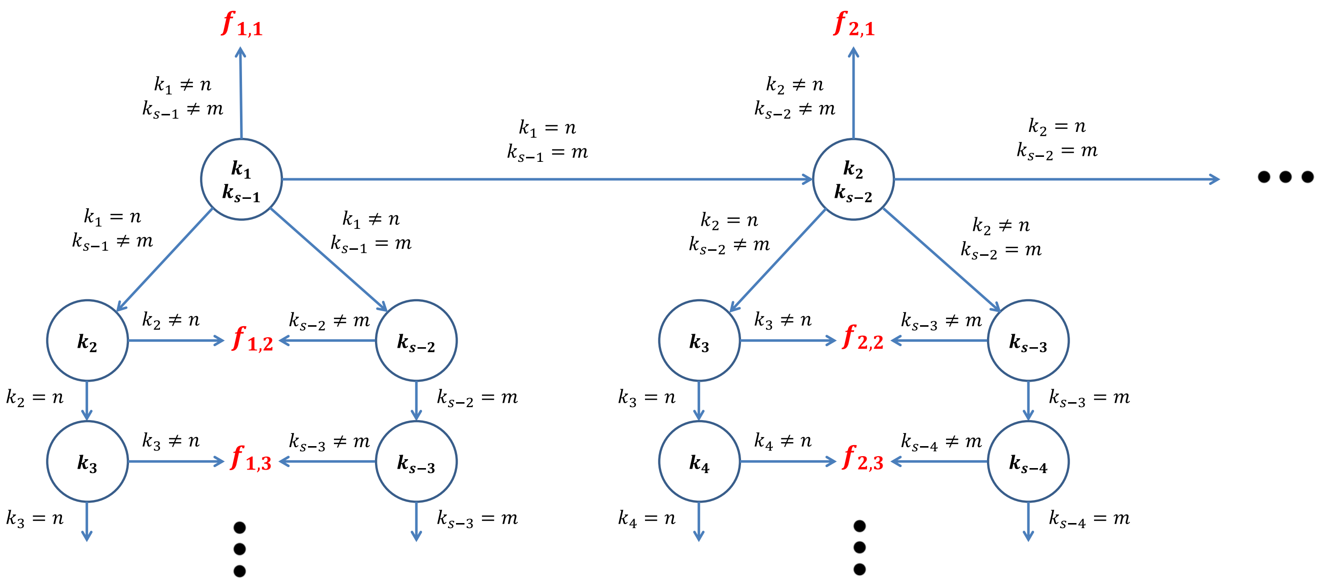

FIG. 2. Schematic construction of the functions $f_{i,j}$.

β_B, $\varrho(\infty) = e^{-\beta_B H_0}/\mathrm{Tr}[e^{-\beta_B H_0}]$. Because the initial state is diagonal in the eigenbasis of the original Hamiltonian H_0, it is evident from Eq. (10) that the state of the system remains diagonal in time.

The probability that the system *absorbs* heat $\mathbb{Q}$ from the bath, in the time interval $(0,\tau)$, is given by

$$P(+\mathbb{Q},\tau) = \sum_{mn} p_m \; p(n,\tau|m,0)\delta\big(\mathbb{Q} - [E_n^{(0)} - E_m^{(0)}]\big). \quad (12)$$

Here, p_m is the probability that the system is initially in the state $|m\rangle$,

$$p_m = \mathrm{Tr}[\varrho(0)|m\rangle\langle m|] = \frac{e^{-\beta_S E_m^{(0)}}}{\mathrm{Tr}[e^{-\beta_S H_0}]}, \quad (13)$$

and $p(n,\tau|m,0)$ is the probability that the system reaches the state $|n\rangle$ at time τ, if it starts from the state $|m\rangle$,

$$p(n,\tau|m,0) = \mathrm{Tr}[\varrho(\tau;m)|n\rangle\langle n|] = \langle n|e^{-\tau A}|m\rangle, \quad (14)$$

where $\varrho(\tau;m)$ is the state of the system at time τ, if it started from the state $|m\rangle$ at time 0 (i.e., $\varrho(0) = |m\rangle\langle m|$). In addition, following Ref. [10], we have defined heat as the energy needed to induce a transition between two energy levels of the original system [10],

$$\mathbb{Q} = E_n^{(0)} - E_m^{(0)}. \quad (15)$$

See also the remark below Eq. (16).

If in the transition $|m\rangle \to |n\rangle$ the system absorbs heat $\mathbb{Q}$ from the bath, in the reverse transition $|n\rangle \to |m\rangle$ it releases the same amount to the bath. Hence the probability that the system *releases* heat $\mathbb{Q}$ to the bath in the time interval $(0,\tau)$ is given by

$$P(-\mathbb{Q},\tau) = \sum_{mn} p_n \; p(m,\tau|n,0)\delta\big(\mathbb{Q} - \big[E_n^{(0)} - E_m^{(0)}\big]\big). \quad (16)$$

Remark. The concepts of "heat" and "work" have been defined in various ways in the literature (see, e.g., Refs. [7,24]). However, for simplicity, here we have adopted the commonly used definition for heat as the change of the energy of the isolated system [Eq. (15)], a definition which sounds plausible within the weak-coupling and Markovian regime when the system Hamiltonian does not vary in time.

In order to prove Eq. (3), we show that

$$p_m \; p(n,\tau|m,0) = e^{\mathbb{Q}\Delta\beta} p_n \; p(m,\tau|n,0). \quad (17)$$

First, we note that from Eq. (13) we have

$$\frac{p_m}{p_n} = e^{\beta_S(E_n^{(0)} - E_m^{(0)})} = e^{\beta_S \mathbb{Q}}. \quad (18)$$

The rest (and main part) of the proof hinges on the following relation:

$$\frac{p(n,\tau|m,0)}{p(m,\tau|n,0)} = \frac{\langle n|e^{-\tau A}|m\rangle}{\langle m|e^{-\tau A}|n\rangle} = e^{-\beta_B \mathbb{Q}}. \quad (19)$$

We give the detailed proof of this in the Appendix. In brief, the idea is as follows. We first write $\langle n|e^{-\tau A}|m\rangle$ as a Taylor series expanding $e^{-\tau A}$. Next, in this series expansion, we show that each expression in the form $\langle n|A^s|m\rangle$ can be recast as $e^{-\beta_B(E_n^0 - E_m^0)/2} f(n,m)$, therefore,

$$\langle n|e^{-\tau A}|m\rangle = e^{-\frac{\beta_B}{2}\left(E_n^{(0)} - E_m^{(0)}\right)} \sum_{s=0} \sum_{i,j=1} a_s(\tau) f_{i,j}(n,m;s). \quad (20)$$

We next argue by a term-by-term analysis and by examining its construction (diagrammatically represented in Fig. 2) that f is a symmetric function of n and m, i.e., $f(n,m) = f(m,n)$.

Combining Eqs. (18) and (19) completes the proof.

Having Eq. (3), similarly to Ref. [10], one can also obtain an upper bound on the (accumulative) probability of a heat transfer from a cold system to a hot bath ($T_S < T_B$ or $\Delta\beta > 0$)—a violation of the Clausius statement of the second law of thermodynamics. This can be seen as follows. Take $\mathbb{Q}$ to be equal to some given (but arbitrary) value q. Then we have

$$\int_{-\infty}^{q} P(\mathbb{Q},\tau)d\mathbb{Q} = \int_{-\infty}^{q} P(-\mathbb{Q},\tau)e^{\mathbb{Q}\Delta\beta}\, d\mathbb{Q} \leqslant e^{q\Delta\beta}. \quad (21)$$

If we now assume that $q \leqslant 0$, the above relation implies that the total probability of a heat transfer of amount $\geqslant |q|$ from a cold system to a hot bath drops *exponentially* with both the amount of the transferred heat $|q|$ and the temperature difference $\Delta\beta$.

IV. SUMMARY

We have derived a quantum fluctuation relation for the heat transfer from a system (in its thermal state) to its bath, when they are interacting such that the system would reach a unique thermal state (characterized by the temperature of the bath) through a weak-coupling, Markovian (Lindbladian) master equation. Unlike the usual fluctuation relations, where the time-reverse dynamics is also assumed valid (microreversibility), here our relation is given by a heat transfer process and its reverse—obviating the need to define a reverse dynamics. In this sense, our relation (although in a form similar to but) differs from the existing fluctuation relations where the probability of absorbing an amount of heat in the forward path is divided by the probability of releasing the same amount of heat in the backward dynamics. We have shown that in the forward dynamics, given by a fairly general class of quantum Markovian evolutions, the Clausius statement of the second law of thermodynamics may be violated negligibly with an exponentially small probability.

It will be interesting to see how far one may extend our analysis to non-Markovian dynamics and see whether some sort of similar fluctuation relation may be obtained.

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APPENDIX: PROOF OF EQ. (19)

We have

$$\langle n|e^{-\tau A}|m\rangle = \sum_{s=0}^{\infty}(1/s!)(-\tau)^s\langle n|A^s|m\rangle. \quad (A1)$$

Let us expand

$$\langle n|A^s|m\rangle = \sum_{k_1,k_2,\ldots,k_{s-1}} \langle n|A|k_1\rangle\langle k_1|A|k_2\rangle\langle k_2|\cdots|k_{s-2}\rangle \times \langle k_{s-2}|A|k_{s-1}\rangle\langle k_{s-1}|A|m\rangle. \quad (A2)$$

In the above sum, we discern four possibilities for the values of indices k_1 and k_{s-1}:

(a) $k_1 \neq n \wedge k_{s-1} \neq m$,
(b) $k_1 = n \wedge k_{s-1} \neq m$,
(c) $k_1 \neq n \wedge k_{s-1} = m$,
(d) $k_1 = n \wedge k_{s-1} = m$.

For case (a), according to Eqs. (7) and (9), the right-hand side (RHS) of the above equation becomes

$$e^{-\beta_B\left(E_n^{(0)}-E_m^{(0)}\right)/2} f_{1,1}(n,m;s), \quad (A3)$$

where

$$f_{1,1}(n,m;s) = \sum_{k_1\neq n,k_2,\ldots,k_{s-2},k_{s-1}\neq m} C_{nk_1}C_{mk_{s-1}}e^{\beta_B(E_{k_1}^{(0)}-E_{k_{s-1}}^{(0)})/2} \times \langle k_1|A|k_2\rangle\langle k_2|\cdots|k_{s-2}\rangle\langle k_{s-2}|A|k_{s-1}\rangle. \quad (A4)$$

We note that the function $f_{1,1}(n,m;s)$ is symmetric under $n \leftrightarrow m$, i.e., $f_{1,1}(n,m;s) = f_{1,1}(m,n;s)$.

In case (b), we consider index k_2. There are two possibilities for the value of k_2, $k_2 = n$ and $k_2 \neq n$, so

(b1) $k_1 = n \wedge k_{s-1} \neq m \wedge k_2 \neq n$,
(b2) $k_1 = n \wedge k_{s-1} \neq m \wedge k_2 = n$.

For case (b1) the RHS of Eq. (A2) becomes

$$e^{-\beta_B(E_n^{(0)}-E_m^{(0)})/2}\langle n|A|n\rangle \sum_{k_2\neq n,k_3,\ldots,k_{s-2},k_{s-1}\neq m} C_{nk_2}C_{mk_{s-1}} \times e^{\beta_B(E_{k_2}^{(0)}-E_{k_{s-1}}^{(0)})/2}\langle k_2|A|k_3\rangle\langle k_3|\cdots|k_{s-2}\rangle\langle k_{s-2}|A|k_{s-1}\rangle. \quad (A5)$$

Before getting to case (b2), we consider case (c) and later combine these cases, as explained below.

In case (c), we look at index k_{s-2}. There are two possibilities for the value of k_{s-2}, $k_{s-2} = m$ and $k_{s-2} \neq m$, so

(c1) $k_1 \neq n \wedge k_{s-1} = m \wedge k_{s-2} \neq m$,
(c2) $k_1 \neq n \wedge k_{s-1} = m \wedge k_{s-2} = m$.

For case (c1) the RHS of Eq. (A2) becomes

$$e^{-\beta_B(E_n^{(0)}-E_m^{(0)})/2}\langle m|A|m\rangle \sum_{k_1\neq n,k_2,\ldots,k_{s-1},k_{s-2}\neq m} C_{nk_1}C_{mk_{s-2}} \times e^{\beta_B(E_{k_1}^{(0)}-E_{k_{s-2}}^{(0)})/2}\langle k_1|A|k_2\rangle\langle k_2|\cdots|k_{s-3}\rangle\langle k_{s-3}|A|k_{s-2}\rangle. \quad (A6)$$

Now, adding up the results of cases (b1) and (c1)—Eqs. (A5) and (A6)—yields

$$e^{-\beta_B(E_n^{(0)}-E_m^{(0)})/2} f_{1,2}(n,m;s), \quad (A7)$$

where

$$f_{1,2}(n,m;s) = (\langle n|A|n\rangle + \langle m|A|m\rangle) \sum_{k_1\neq n,k_2,\ldots,k_{s-1},k_{s-2}\neq m} C_{nk_1} \times C_{mk_{s-2}}e^{\beta_B(E_{k_1}^{(0)}-E_{k_{s-2}}^{(0)})/2}\langle k_1|A|k_2\rangle\langle k_2|\cdots| \times k_{s-3}\rangle\langle k_{s-3}|A|k_{s-2}\rangle. \quad (A8)$$

We note that, similarly to $f_{1,1}(n,m;s)$, the function $f_{1,2}(n,m;s)$ is symmetric, as $f_{1,2}(n,m;s) = f_{1,2}(m,n;s)$.

In a similar fashion, for cases (b2) and (c2), we consider, respectively, indices k_3 and k_{s-3}. The possibilities are as follows:

(b2.1) $k_1 = n \wedge k_{s-1} \neq m \wedge k_2 = n \wedge k_3 \neq n$,
(b2.2) $k_1 = n \wedge k_{s-1} \neq m \wedge k_2 = n \wedge k_3 = n$,
(c2.1) $k_1 \neq n \wedge k_{s-1} = m \wedge k_{s-2} = m \wedge k_{s-3} \neq m$,
(c2.2) $k_1 \neq n \wedge k_{s-1} = m \wedge k_{s-2} = m \wedge k_{s-3} = m$.

The combination of the terms corresponding to cases (b2.1) and (c2.1) in the RHS of Eq. (A2) can be written as

$$e^{-\beta_B(E_n^{(0)}-E_m^{(0)})/2} f_{1,3}(n,m;s), \tag{A9}$$

where

$$f_{1,3}(n,m;s) = (\langle n|A|n\rangle^2 + \langle m|A|m\rangle^2) \sum_{k_1\neq n,k_2,\ldots,k_{s-2},k_{s-3}\neq m} C_{nk_1} \times C_{mk_{s-3}} e^{\beta_B(E_{k_1}^{(0)}-E_{k_{s-3}}^{(0)})/2} \langle k_1|A|k_2\rangle \times \langle k_2|\cdots|k_{s-3}\rangle\langle k_{s-4}|A|k_{s-3}\rangle. \tag{A10}$$

We can continue this procedure for the remaining possibilities.

For case (d), the RHS of Eq. (A2) becomes

$$\langle n|A|n\rangle\langle m|A|m\rangle \sum_{k_2,k_3,\ldots,k_{s-2}} \langle n|A|k_2\rangle\langle k_2|A|k_3\rangle\langle k_3|\cdots|k_{s-3}\rangle \times \langle k_{s-3}|A|k_{s-2}\rangle\langle k_{s-2}|A|m\rangle. \tag{A11}$$

The above sum is the same as the sum in Eq. (A2) except with two less indices. Thus the same steps (a)–(d) can be carried out here again. Figure 2 summarizes the whole scenario. Combining all pieces, the matrix elements of A^s are then given by

$$\langle n|A^s|m\rangle = e^{-\beta_B(E_n^{(0)}-E_m^{(0)})/2}\sum_{i,j} f_{i,j}(n,m;s), \tag{A12}$$

where

$$f_{i,j}(n,m;s) = (1/2)^{\delta_{j,1}}(\langle n|A|n\rangle\langle m|A|m\rangle)^{i-1}(\langle n|A|n\rangle^{j-1} + \langle m|A|m\rangle^{j-1}) \sum_{k_1\neq n,k_2,\ldots,k_{s-j-2(i-1)}\neq m} C_{nk_1}C_{mk_{s-j-2(i-1)}} \times e^{\beta_B(E_{k_1}^{(0)}-E_{k_{s-j-2(i-1)}}^{(0)})/2}\langle k_1|A|k_2\rangle\langle k_2|\cdots| \times k_{s-j-2i+1}\rangle\langle k_{s-j-2i+1}|A|k_{s-j-2(i-1)}\rangle. \tag{A13}$$

Note that each $f_{i,j}(n,m;s)$ has the $m\leftrightarrow n$ symmetry, $f_{i,j}(n,m;s) = f_{i,j}(m,n;s)$. Substituting Eq. (A12) in Eq. (A1) yields

$$\langle n|e^{-\tau A}|m\rangle = e^{-\beta_B(E_n^{(0)}-E_m^{(0)})/2}\sum_{s=0}\sum_{i,j=1} a_s(\tau) f_{i,j}(n,m;s). \tag{A14}$$

Since $f_{i,j}(n,m;s)$'s are symmetric under the $n\leftrightarrow m$ transformation, one can conclude that

$$\frac{\langle n|e^{-\tau A}|m\rangle}{\langle m|e^{-\tau A}|n\rangle} = e^{-\beta_B(E_n^{(0)}-E_m^{(0)})} = e^{-\beta_B\mathbb{Q}}. \tag{A15}$$

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