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An operator derivation of the Feynman–Vernon theory, with applications to the generating function of bath energy changes and to an-harmonic baths

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Abstract

We present a derivation of the Feynman–Vernon approach to open quantum systems in the language of super-operators. We show that this gives a new and more direct derivation of the generating function of energy changes in a bath, or baths. As found previously, this generating function is given by a Feynman–Vernon-like influence functional, with only time shifts in the kernels coupling the forward and backward paths. We further show that the new approach extends to an-harmonic and possible non-equilibrium baths, provided that the interactions are bi-linear, and that the baths do not interact between themselves. Such baths are characterized by non-trivial cumulants. Every non-zero cumulant of certain environment correlation functions is thus a kernel in a higher-order term in the Feynman–Vernon action.

Keywords: stochastic thermodynamics, quantum thermodynamics, quantum thermal power operators, super-operator formalism, higher-order Feynman–Vernon influence functionals

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

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1. Introduction

When a quantum system interacts with an environment (open quantum system or OQS), the state of the system is influenced by the environment in a fundamental manner. Decoherence, for example, is a consequence of quantum entanglement between the system and the environment [1]. The understanding of effects on the system induced by the environments is essential to the quantum information technology [2] and quantum thermodynamics [3]. A direct inclusion of the environment in a first principle investigation is usually not practically feasible. On the other hand, the structure and details of a large environment can only partially be reflected in the dynamics of the system. In the super-operator approach going back to Nakajima and Zwanzig [4, 5] one starts from equations of motion (von Neumann–Liouville equations) of the total density matrix of the system and the environment, and projects that to an effective dynamics for the system density matrix [6–8]. In the alternative approach of Feynman and Vernon the influence of an environment on the system is expressed in terms of an influence functional [9]. The development of the reduced density matrix of the system is then given by a double path integral, where the influence functional couples the two paths. If used exactly, both approaches agree and specify completely positive dynamic maps describing the evolution of the system. Once these are obtained, the system dynamics can be investigated without the knowledge of environment dynamics [7, 8].

This paper is about the relation between the two approaches, and how important extensions consequences obtained only recently are much more easily derived in the super-operator approach. We will also show that the super-operator approach yields new higher-order corrections to Feynman–Vernon. Before proceeding to the main argument, we note that even after one has found a closed form expression of a dynamical map or the influence functional, calculating the time-evolution of the system using them is technically challenging. Tractable methods using various approximations have been developed. Quantum master equations (Lindblad equations) [10, 11] based on the Born-Markovian approximation are the most popular, and can be derived in both approaches [7, 12, 13]. Non-Markovian methods are also developed [14–16]. For the 'spin-boson' problem of one two-state system (a qubit) interacting with a bosonic bath the non-interacting blip approximation (NIBA) was developed [17], and shown to be equivalent to relaxation after a polaron transform [18, 19]. Many numerical algorithms have been developed to treat the spin-boson problem with one or several bath including the hierarchical equation of motion (HEOM) [15, 20–24], the quasi-adiabatic propagator path integral (QuAPI) [25, 26], the multi-configuration time-dependent Hartree (MCTDH) approach [27], the Stochastic Liouvillian algorithm [28], and other Monte Carlo approaches [29].

The thermodynamics of an OQS describes how a quantum system exchanges energy, particles and other quantities with one or several reservoirs [30]. While in quantum Markov dynamics the energy interchanged with a reservoir, which we call *heat*, can be expressed in terms of Lindblad operators acting on the system density matrix [31], in general that is not so. Nevertheless, it has recently been shown by several groups that the generating function of heat can be computed by the path integral technique in a Feynman–Vernon-like approach [32–35]. In this formulation appears a new influence functional depending on the generating function parameters. However, the terms in this new influence functional are in fact the same as in Feynman–Vernon, with only a time shift in the argument in some of the kernels as announced previously in [36]. We will in this work derive the same result in the super-operator

formalism where it emerges in a straight-forward manner without cancellations in intermediate steps of the calculation.

Let us emphasize that the super-operator approach itself is well known: the expression for the evolution operator of the reduced density matrix of the system (which we will derive as equation (10)) is given, in the Schrödinger picture, as equation (3.508) on page 187 in the monograph of Breuer and Petruccione [7]. Nevertheless, we feel that this result is not as appreciated as it should be. In particular, it deserves to be better known that the super-operator approach extends both generating function of heat and to environments beyond ideal Bose gases. As we will show, cumulants of specific bath correlation functions (to be discussed below) enter as kernels in higher-order order terms in the Feynman–Vernon action. For instance, a non-zero third-order bath correlation function gives the kernel of a third-order term in the Feynman–Vernon action. This result have to our knowledge not previously been obtained in either the path integral formulation or in the super-operator formalism.

We present the equivalence of super-operator and Feynman–Vernon approach in a coherent manner through the generating function of heat as an example, and the extension to an-harmonic baths. The paper is organized as follows. Section 2 contains a brief summary of the path integral and super-operator approaches to the development of the system density matrix, and section 3 extends the discussion to generating function of heat. Section 4 contains a systematic and general derivation of the super-operator approach, and section 5 the extension to an-harmonic baths. Section 6 sums up and discussed the results. Appendix A defines the super-operator time-ordering used in the main body of the paper, and appendix B contains the details of the pair correlation functions in harmonic baths. Appendix C shows the third- and fourth-order kernels in high-order influence functional that result from non-zero third-order and fourth-order cumulants in the bath. Appendix D gives for completeness an outline of the path integral derivation the result of which was previously announced in [36].

2. Theory of open quantum systems: a brief summary

Consider a system S in contact with an environment B. Their Hamiltonians are denoted as H_s and H_b, respectively and they are coupled through an interaction Hamiltonian V_sb. The whole system is assumed to be initially in a product state $\rho(t_i) = \rho_\mathrm{s}(t_i) \otimes \rho_\mathrm{b}(t_i)$ and evolves by a unitary transformation:

$$\rho(t) = U(t;t_i)\rho(t_i)U^\dagger(t;t_i) \tag{1}$$

where ρ is the density operator of the whole system and $U(t;t_i) = \mathrm{e}^{-\mathrm{i}H(t-t_i)}$ is a usual time-evolution operator with the total Hamiltonian $H = H_\mathrm{s} + H_\mathrm{b} + V_\mathrm{sb}$. Units of time are chosen such that $\hbar = 1$.

When considered together with a time-constant interaction the assumption of an initial product state limits the analysis to weak system-bath interaction. One way around that problem is to take the system-bath interaction time-dependent and small only initially, as was allowed for in the original treatment by Feynman and Vernon [9]. Other approaches were discussed in [13], and have some advantages for the analysis of the quantum state. When considering thermodynamic quantities the question is more involved. Even in classical mesoscopic systems interacting strongly with an environment the concept of heat is controversial, and has been vigorously debated in the recent literature [37–40]. As discussed by one of us [41] different proposals for strong-coupling heat can be understood as different types of control

and joint initial conditions of bath and baths. On the quantum side, aspects of some of these issues were discussed some time ago in exactly solvable models [42, 43]. Here we will follow the option made available in [9] and assume that the system-bath interaction vanishes at the beginning and the end of a process, but can be arbitrarily strong in between. Heat can then be identified by the energy change in a bath. Heat has been calculated in this scenario using equation (10) in [44].

Returning to the previous thread, the state of the system is defined as a reduced density $\rho_{\rm s} = {\rm tr}_{\rm b}\rho(t)$ where ${\rm tr}_{\rm b}$ traces out the degrees of freedom of the environments. The theory of OQS seeks a completely positive operation $\mathcal{M}(t;t_i)$ (dynamical map, or quantum map) defined by [11]

$$\rho_{\rm s}(t) = {\rm tr}_{\rm b}\{U(t;t_i)\rho(t_i)U^{\dagger}(t;t_i)\} \equiv \mathcal{M}(t,t_i)\rho_{\rm s}(t_i) \tag{2}$$

Alternatively, a quantum map can be considered as the given, and then one of the many possible couplings to an environment giving rise to same map after tracing out the environment is called an environmental representation of that map [45]. In either case, once the map is obtained, we can evaluate any quantity associated with the system. For example the transition probability from an initial pure state of the system $|i\rangle$ to a final pure state of the system $|f\rangle$ is given by

$$P_{if} = {\rm tr}_{\rm s}\{|f\rangle\langle f|\,\mathcal{M}(t_i,t_f)\left(|i\rangle\langle i|\right)\}. \tag{3}$$

In the path integral approach the dynamical map (2) is written in coordinate basis as

$$\rho_{\rm s}(Q_f,\tilde{Q}_f;t) = \iint {\rm d}Q_i\,{\rm d}\widetilde{Q}_i\mathcal{M}(Q_f,\widetilde{Q}_f;Q_i,\widetilde{Q}_i;t_f,t_i)\rho_{\rm s}(Q_i,\widetilde{Q}_i,t_i) \tag{4}$$

where Q and $\tilde{Q}$ are coordinates of the system at the initial time t_i and final time t_f. Feynman and Vernon wrote $\mathcal{M}$ in a path integral form

$$\mathcal{M}(Q_f,\tilde{Q}_f;Q_i,\tilde{Q}_i;t_f,t_i) = \iint \mathcal{D}Q\,\mathcal{D}\widetilde{Q}\,{\rm e}^{{\rm i}S[Q]}\,F(Q,\widetilde{Q};t_f,t_i)\,{\rm e}^{-{\rm i}S[\widetilde{Q}]} \tag{5}$$

where $S[Q]$ is the classical action of a system trajectory without interactions with the environment, and $Q(t)$ and $\tilde{Q}(t)$ are forward and time-reversed trajectories. The effects of the environment are fully included in the *influence functional* $F[Q,Q';t_f,t_i]$ [9, 46]. General properties of influence functionals were discussed in [9], and we will return to those below.

Exact expressions can be obtained when the functional integrals over the environment variables can be done in closed form. In practice this means that integrals have to be Gaussian, which is the case when the environment is a Bose gas with Hamiltonian $H_{\rm b} = \sum_k \left(p_k^2/2m_k + m_k\omega_k^2 q_k^2/2\right)$ initially in Gibbs states $\rho_{\rm b}(t_i) = {\rm e}^{-\beta H_{\rm b}}/Z_{\rm b}$, and the coupling takes a bi-linear form $V_{\rm sb} = X_{\rm s}\otimes(\sum_x c_k q_k)$ where $X_{\rm s}$ is an operator of the system, and c_k is the coupling strength. Such an environment is called a harmonic bath at inverse temperature β. We will later discuss the case when the environment consists of two or more harmonic baths, each with its own temperature.

Under the above assumptions the influence functional can be written in exponential form $F[Q,\tilde{Q};t_f,t_i] = {\rm e}^{{\rm i}\Phi[Q,\tilde{Q};t_f,t_i])}$ where

$$i\Phi[Q,\tilde{Q};t_f,t_i] = i\int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}s' \left[(Q(s)-\tilde{Q}(s))(Q(s')+\tilde{Q}(s'))\kappa_i\left(s,s'\right)\right] - \int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}s' \left[(Q(s)-\tilde{Q}(s))(Q(s')-\tilde{Q}(s'))\kappa_r\left(s,s'\right)\right] \tag{6}$$

where Φ is known as the influence action or Feynman–Vernon action, and $\kappa_r(\tau)$ and $\kappa_i(\tau)$ are known as dissipation and noise kernel. These functions are

$$\kappa_i(s,s') = \sum_k \frac{c_k(s)c_k(s')}{2m_k\omega_k} \sin\ \omega_k(s-s') \tag{7}$$

$$\kappa_r(s,s') = \sum_k \frac{c_k(s)c_k(s')}{2m_k\omega_k} \coth\left(\frac{\omega_k\beta}{2}\right) \cos\ \omega_k(s-s') \tag{8}$$

Except close to the initial and final times the time-dependence of the coupling coefficients c_k can be ignored, and the kernels κ_i and κ_r then only depend on the time difference $\tau = s - s'$.

In the super-operator approach one instead directly evaluates equation (1). Rewriting equation (1) using the Liouville operator $\mathcal{L}(t)\bullet = -i[V_{\mathrm{sb}}(t),\bullet]$ in the interaction picture, the map (in the interaction picture) can be written as

$$\mathcal{M}(t_f,t_i) = \mathrm{tr_b}\left\{\overleftarrow{\mathcal{T}}\ \exp\left[\int_{t_i}^{t_f} \mathcal{L}(s)\mathrm{d}s\right](I\otimes\rho_{\mathrm{b}}(t_i))\right\} \tag{9}$$

where $\overleftarrow{\mathcal{T}}$ is time-ordering super-operator which chronologically orders the super-operators (see appendix A.) The symbol $\bullet$ in above and in the following is the 'slot' on which the super-operator acts, and represents any operator, including density operator. Using Wick's theorem, we find the map $\mathcal{M}(t_f,t_i) = \overleftarrow{\mathcal{T}}\mathrm{e}^{\mathrm{i}\Phi(t_f,t_i)}$ with super-operator

$$i\Phi(t_f,t_i) = i\int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}s' \left[\left(\mathcal{X}_{\mathrm{s}}^{+}(s)+\mathcal{X}_{\mathrm{s}}^{-}(s)\right)\left(\mathcal{X}_{\mathrm{s}}^{+}(s')-\mathcal{X}_{\mathrm{s}}^{-}(s')\right)\kappa_i(s-s')\right] - \int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}s' \left[\left(\mathcal{X}_{\mathrm{s}}^{+}(s)+\mathcal{X}_{\mathrm{s}}^{-}(s)\right)\left(\mathcal{X}_{\mathrm{s}}^{+}(s')+\mathcal{X}_{\mathrm{s}}^{-}(s')\right)\kappa_r(s-s')\right] \tag{10}$$

where $\mathcal{X}_{\mathrm{s}}^{+}(t)\bullet = X_{\mathrm{s}}(t)\bullet$ and $\mathcal{X}_{\mathrm{s}}^{-}(t)\bullet = -\bullet X_{\mathrm{s}}(t)$ The two super-operators together can be expressed with commutator and anti-commutator as $(\mathcal{X}_{\mathrm{s}}^{+}\pm\mathcal{X}_{\mathrm{s}}^{-})\bullet = [X_{\mathrm{s}},\bullet]_{\mp}$. The kernels κ_r and κ_i in (10) are the same as those in equation (6), and will be shown to be the equilibrium pair correlation functions of the ideal Bose gas.

While the two methods use different mathematical objects, one with paths $Q(t)$ and $\tilde{Q}(t)$, and the other with super-operators $\mathcal{X}_{\mathrm{s}}^{+}(t)$ and $\mathcal{X}_{\mathrm{s}}^{-}(t)$, equations (6) and (10) clearly show similarity. They are the same if two quantities are replaced as $Q(t)\leftrightarrow\mathcal{X}_{\mathrm{s}}^{+}(t)$ and $\tilde{Q}(t)\leftrightarrow -\mathcal{X}_{\mathrm{s}}^{-}(t)$.

Extension of these methods to a system interacting with multiple environments is straightforward if the environments do not interact between themselves. Indeed, *General property of influence functionals 2* of Feynman and Vernon states that 'if a number of (environments) act on (the system) and if F^k is the influence of the k'th (environment) alone, then the total influence of all (the environments) is given by the product of the individual influences' [9]. In the super-operator approach the same statement follows from the observation that if the Liouville operator is a sum, say $\mathcal{L}(t)\bullet = -i[V_{\mathrm{sh}}(t),\bullet] - i[V_{\mathrm{sc}}(t),\bullet]$, and if the environment

operators in $V_{\rm sh}(t)$ and $V_{\rm sc}(t)$ commute and act on parts of the environment that start in a product state (different baths), then the time ordering of environment operators in (9) can be done separately.

3. Generating function of heat

Once the dynamical map is found, we know the state of the system precisely. However, the information on the state of environments is completely buried in the map. If we want to investigate any quantity associated with the environments or correlation between the system and environments, the knowledge of the system density alone is not enough. In order to make our story concrete, we consider a system interacting with a hot and a cold bath. Their Hamiltonians are denoted as $H_{\rm s}$, $H_{\rm h}$, and $H_{\rm c}$, respectively, and the interaction Hamiltonians between the system and the baths are $V_{\rm sh}$ and $V_{\rm sc}$. As is well known, for harmonic baths the interaction Hamiltonian are accompanied by the Caldeira–Leggett counter-terms [12] which redefine the system Hamiltonian $H_{\rm s}$.

The initial state of the whole system is assumed to be a product state $\rho(t_i) = \rho_{\rm s}(t_i) \otimes \rho_{\rm h}(t_i) \otimes \rho_{\rm c}(t_i)$ and the baths are at thermal equilibrium

$$\rho_\ell(t_i) = \frac{1}{Z_\ell}\sum_n |E_\ell(n)\rangle\, {\rm e}^{-\beta_\ell E_\ell(n)} \langle E_\ell(n)|\,, \quad \ell = {\rm h}, {\rm c} \tag{11}$$

where Z_ℓ is a partition function. $E_\ell(n)$ and $|E_\ell(n)\rangle$ are eigenvalue and the corresponding eigenket of H_ℓ. The system is initially in an arbitrary state $\rho_s(t_i) = \sum_i \mu_i |i\rangle \langle i\rangle$ where μ_i and $|i\rangle$ are eigenvalues and eigenkets of the density.

Now we want know the change in the energy of the cold bath, $\Delta E_{\rm c}$, over time period $t_f - t_i$. The probability distribution of $\Delta E_{\rm c}$ may be written as

$$P(\Delta E_{\rm c}) = \sum_f \sum_i \mu_i P_{if}(\Delta E_{\rm c}) \tag{12}$$

where

$$P_{if}(\Delta E_{\rm c}; t_f) = \sum_{m,m'}\sum_{n,n'} |\langle f, m', n'| U(t_f, t_i) |i, m, n\rangle|^2 \delta(\Delta E_{\rm c} - E_{\rm c}(n') + E_{\rm c}(n)) \times \frac{{\rm e}^{-\beta_{\rm h} E_{\rm h}(m)}}{Z_{\rm h}} \frac{{\rm e}^{-\beta_{\rm c} E_{\rm c}(n)}}{Z_{\rm c}}$$

μ_i and $|i\rangle$ are the eigenvalue and eigenket of $\rho_{\rm s}(t_i)$ and the final states $|f\rangle$ can be any basis set.

The generating function of heat is the Fourier transform of parameter ν of the probability distribution $P(\Delta E_{\rm c}; t_f)$ with respect to variable $\Delta E_{\rm c}$. One finds

$$G(\nu; t_f) = \int_{-\infty}^{\infty} P(q, t_f) {\rm e}^{{\rm i}\nu q}\, {\rm d}q = {\rm tr}_{\rm s}\, \Gamma(\nu; t_f) \tag{13}$$

where

$$\Gamma(\nu, t_f) = {\rm tr}_{\rm h}{\rm tr}_{\rm c} \left\{ {\rm e}^{{\rm i}\nu H_{\rm c}} U(t_f, t_i) {\rm e}^{-{\rm i}\nu H_{\rm c}} \rho(0) U^\dagger(t_f, t_i) \right\}. \tag{14}$$

is an operator in the Hilbert space of the system. Direct comparison of equations (2) and (14) shows that $\Gamma(\nu, t)$ is quite similar to $\rho_{\rm s}(t)$. In fact, when $\nu = 0$, they coincide. The only difference is that one of the time evolution operators in equation (14) is rotated by ${\rm e}^{{\rm i}\nu H_{\rm c}}$.

The resemblance suggests that the generating function can be computed with the methods developed for OQS. Following the procedure discussed in the previous section, we first write Γ with a map as

$$\Gamma(\nu, t_f) = \mathcal{M}(\nu; t_f, t_i)\rho_{\rm s}(t_i). \tag{15}$$

where

$$\mathcal{M}(\nu; t_f, t_i) = {\rm tr_h tr_c}\left\{\left(I_{\rm s}\otimes I_{\rm h}\otimes \overleftarrow{\mathcal{T}}\,{\rm e}^{{\rm i}\nu H_{\rm c}}\right)\exp\left[\int_{t_i}^{t_f}\mathcal{L}(s)\,{\rm d}s\right]\left(I_{\rm s}\otimes\rho_{\rm h}(t_i)\otimes {\rm e}^{-{\rm i}\nu H_{\rm c}}\rho_{\rm c}(t_i)\right)\right\} \tag{16}$$

We generally assume that the system interacts separately with the hot and the cold bath, and thus the Liouville operator is split to two parts, $\mathcal{L}_{\rm x} = -i[V_{\rm sx}(t), \cdot], \quad {\rm x} = {\rm c}, {\rm h}$. If the system parts of the two operators (in the interaction picture) always commute the expression further factorizes into the product of the traces over the two baths separately.

The energy change associated with a particular transition from $|i\rangle$ to $|f\rangle$ can be expressed like the transition probability (3):

$$G_{if}(\nu, t) = {\rm tr_s}\{|f\rangle\langle f|\,\mathcal{M}(\nu; t_f, t_0)\left(|i\rangle\langle i|\right)\} \tag{17}$$

In order to use the path integral approach, we express equation (15) in the coordinate representation in the same way as equation (4),

$$\Gamma(\nu; Q_f, \tilde{Q}_f, ; t_f, t_i) = \iint {\rm d}Q_i\,{\rm d}\widetilde{Q}_i\mathcal{M}(\nu; Q_f, \widetilde{Q}_f; Q_i, \widetilde{Q}_i; t_f, t_i)\rho_{\rm s}(Q_i, \widetilde{Q}_i, t_i). \tag{18}$$

The map has been derived using the path integral [47] for the baths of ideal Bose gases and expressed with a new influence functional $F[\nu; Q, \tilde{Q}; t_f, t_i] = {\rm e}^{{\rm i}\Phi_{\rm h}[Q,\tilde{Q};t_f,t_i]+{\rm i}\Phi_{\rm c}[\nu;Q,\tilde{Q};t_f,t_i]}$. For the hot bath, $\Phi_{\rm h}$ remains exactly the same as equation (6) but for the cold bath, $\Phi_{\rm c}$ is slightly changed to

$$\begin{aligned} i\Phi_{\rm c}[\nu; Q, \tilde{Q}; t_f, t_i] = &-\int_{t_i}^{t_f}{\rm d}s\int_{t_i}^{s}{\rm d}s'\left[Q(s)Q(s') + \tilde{Q}(s)\tilde{Q}(s')\right]\kappa_r(s-s') \\ &+ i\int_{t_i}^{t_f}{\rm d}s\int_{t_i}^{s}{\rm d}s'\left[Q(s)Q(s') - \tilde{Q}(s)\tilde{Q}(s')\right]\kappa_i(s-s') \\ &+ \int_{t_i}^{t_f}{\rm d}s\int_{t_i}^{t_f}Q(s)\tilde{Q}(s')\left[\kappa_r(s-s'+\nu) + i\kappa_i(s-s'+\nu)\right]. \end{aligned} \tag{19}$$

where for simplicity we write the kernels as they are away from the initial and final times. When $\nu = 0$, equation (19) is back to equation (6). The time shift ν in the cross correlation between forward and backward trajectories contains all information about $\Delta E_{\rm c}$. In the path integral approach (19) emerges from rather complicated intermediate results after cancellations and using properties of hyperbolic and trigonometric functions. For completeness we provide in appendix D an outline of these results previously announced in [36].

Based on the correspondence between the path integral method and the super operator method, we expect that the map defined in equation (16) is given by $\mathcal{M}(\nu; t_f, t_i) = {\rm e}^{{\rm i}\Phi_{\rm h}(t_f,t_i)+{\rm i}\Phi_{\rm c}(\nu;t_f,t_i)}$ with

$$
\begin{aligned}
i\Phi_{\mathrm{c}}(\nu;t_f,t_i) = &-\int_{t_i}^{t_f}\mathrm{d}s\int_{t_i}^{s}\mathrm{d}s'\left[\mathcal{X}^{+}(s)\mathcal{X}^{+}(s')+\mathcal{X}^{-}(s)\mathcal{X}^{-}(s')\right]\kappa_r(s-s')\\
&+i\int_{t_i}^{t_f}\mathrm{d}s\int_{t_i}^{s}\mathrm{d}s'\left[\mathcal{X}^{+}(s)\mathcal{X}^{+}(s')-\mathcal{X}^{-}(s)\mathcal{X}^{-}(s')\right]\kappa_i(s-s')\\
&-\int_{t_i}^{t_f}\mathrm{d}s\int_{t_i}^{t_f}\mathcal{X}^{+}(s)\mathcal{X}^{-}(s')x\left[\kappa_r(s-s'+\nu)+i\kappa_i(s-s'+\nu)\right]
\end{aligned}
\tag{20}
$$

$\Phi_{\mathrm{h}}(t_f;t_i)$ remains the same as equation (10). In the next section, we derive equation (20), and show that it appears more directly in the super-operator approach.

4. Super-operator approach for the generating function

We derive equation (20) by evaluating the super-operator expression of map

$$
\mathcal{M}_{\mathrm{c}}(\nu;t_f,t_i) = \mathrm{tr}_{\mathrm{c}}\left\{\mathrm{e}^{\mathrm{i}\nu H_{\mathrm{c}}}\overleftarrow{\mathcal{T}}\ \exp\left[\int_{t_i}^{t_f}\mathcal{L}_{\mathrm{c}}(s)\,\mathrm{d}s\right]I_{\mathrm{s}}\otimes\mathrm{e}^{-\mathrm{i}\nu H_{\mathrm{c}}}\rho_{\mathrm{c}}(t_i)\right\}. \tag{21}
$$

For simplicity we have assumed that the system parts of the interaction Hamiltonians commute, which allow us to focus on the trace over the cold bath.

First we rewrite H_{b} with creation and annihilation operators, $a_k^{\dagger}$ and a_k:

$$
H_{\mathrm{b}} = \sum_k \omega_k a_k^{\dagger}a_k \tag{22}
$$

and the interaction Hamiltonian $V_{\mathrm{c}} = X_{\mathrm{s}}\otimes Y_{\mathrm{c}}$ with

$$
Y_{\mathrm{c}} = \sum_k c_k'(a_k^{\dagger}+a_k) \tag{23}
$$

where $c_k' = c_k/\sqrt{2m_k\omega_k}$ is coupling strength. The system part of the coupling X_s is arbitrary.

Using the interaction picture, the Liouville super-operator is defined by

$$
\mathcal{L}_{\mathrm{c}}(t)\bullet = -i[X_{\mathrm{s}}(t)\otimes Y_{\mathrm{b}}(t),\bullet] = -i\sum_{d=\pm}\left\{\mathcal{X}_{\mathrm{s}}^{d}(t)\otimes\mathcal{Y}_{\mathrm{c}}^{d}(t)\right\}\bullet \tag{24}
$$

where $X_{\mathrm{s}}(t) = \mathrm{e}^{\mathrm{i}H_{\mathrm{s}}(t-t_i)}X_{\mathrm{s}}\mathrm{e}^{-\mathrm{i}H_{\mathrm{s}}(t-t_i)}$ and $\mathrm{e}^{\mathrm{i}H_{\mathrm{b}}(t-t_i)}Y_{\mathrm{b}}^{-\mathrm{i}H_{\mathrm{b}}(t-t_i)}$ and for mathematical convenience, we introduced the following super-operators

$$
\mathcal{X}_{\mathrm{s}}^{+}\bullet = X_{\mathrm{s}}\bullet,\quad \mathcal{X}_{\mathrm{s}}^{-}\bullet = -\bullet X_{\mathrm{s}},\quad \mathcal{Y}_{\mathrm{c}}^{+}\bullet = Y_{\mathrm{c}}\bullet,\quad \mathcal{Y}_{\mathrm{c}}^{-}\bullet = \bullet Y_{\mathrm{c}}. \tag{25}
$$

Expanding the exponential function in equation (21)

$$
\begin{aligned}
\mathcal{M}_c(\nu;t_f,t_i) &= \sum_n\frac{1}{n!}\int_{t_i}^{t_f}\mathrm{d}t_1\cdots\int_{t_i}^{t_f}\mathrm{d}t_n\mathrm{tr}_{\mathrm{c}}\left\{\mathrm{e}^{\mathrm{i}\nu H_{\mathrm{c}}}\overleftarrow{\mathcal{T}}\mathcal{L}(t_1)\ \cdots\ \mathcal{L}(t_n)\left(I_{\mathrm{s}}\otimes\mathrm{e}^{-\mathrm{i}\nu H_{\mathrm{c}}}\rho_{\mathrm{b}}(t_i)\right)\right\}\\
&= \sum_n\frac{(-i)^n}{n!}\int_{t_i}^{t_f}\mathrm{d}t_1\cdots\int_{t_i}^{t_f}\mathrm{d}t_n\sum_{d_1}\ \cdots\ \sum_{d_n}\overleftarrow{\mathcal{T}}\left(\mathcal{X}_{\mathrm{s}}^{d_1}(t_1)\cdots\mathcal{X}_{\mathrm{s}}^{d_n}(t_n)\right)\\
&\quad\times\ C^{d_1,\ldots,d_n}(\nu;t_1,\ldots,\ t_n)
\end{aligned}
\tag{26}
$$

where multi-time correlation functions of the environment are defined as

$$C^{d_1,\ldots,d_n}(\nu;t_1,\ldots,\ t_n) = \mathrm{tr_c}\left[\overleftarrow{\mathcal{T}}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{d_1}(t_1)\ \cdots\ \mathcal{Y}_\mathrm{c}^{d_n}(t_n)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\right\}\rho_\mathrm{c}(t_1)\right]$$
$$= \langle\overleftarrow{\mathcal{T}}[\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{d_1}(t_1)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}]\ \cdots\ [\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{d_n}(t_n)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}]\rangle_{t_i} \tag{27}$$

where $\langle\cdots\rangle_{t_i}$ indicates expectation value $\mathrm{tr_c}\{\cdots\rho_\mathrm{c}(t_i)\}$. Since H_c is quadratic in a and $a^\dagger$, all odd order correlation functions vanish. For the even order terms, we apply the Wick's theorem for operators $\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{d_1}(t)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}$

$$C^{d_1,\cdots d_{2n}}(t_1,\ldots,\ t_{2n}) = \sum_{\substack{\text{all possible}\\ \text{pairing}}}\ \prod_{\text{all pairs}} C^{d_j,d_k}(t_j,t_k) \tag{28}$$

where $\sum_{\text{all pairing}}$ indicates the sum of all possible combinations of pairs. The map is now expressed with the pair correlation functions as

$$\mathcal{M}_\mathrm{c}(\nu;t_f,t_i) = \overleftarrow{\mathcal{T}}\sum_n\frac{(-i)^{2n}}{2^n n!}\left[\int_{t_i}^{t_f}\mathrm{d}t_1\int_{t_i}^{t_f}\mathrm{d}t_2\sum_{d_1}\sum_{d_2}\mathcal{X}_\mathrm{s}^{d_1}(t_1)\mathcal{X}_\mathrm{s}^{d_2}(t_2)C^{d_1,d_2}(\nu;t_1,t_2)\right]^n$$
$$= \overleftarrow{\mathcal{T}}\ \exp\left[-\frac{1}{2}\int_{t_i}^{t_f}\mathrm{d}t_1\int_{t_i}^{t_f}\mathrm{d}t_2\sum_{d_1}\sum_{d_2}\mathcal{X}_\mathrm{s}^{d_1}(t_1)\mathcal{X}_\mathrm{s}^{d_2}(t_2)C^{d_1,d_2}(\nu;t_1,t_2)\right] \tag{29}$$

The four pair correlation functions C^{++}, C^{+-}, C^{-+}, and C^{--} can be expressed with the standard pair correlation function $\kappa(\tau)$ as shown in figure 1. (See appendix B.) The correlation functions between the two times on the same branch are

$$C^{++}(t_1,t_2) = \kappa_r(\tau) - i\,\mathrm{sign}(\tau)\kappa_i(\tau), \quad C^{--}(t_1,t_2) = \kappa_r(\tau) + i\,\mathrm{sign}(\tau)\kappa_i(\tau) \tag{30}$$

where $\tau = t_1 - t_2$. The cross correlation functions are

$$C^{+-}(t_1,t_2) = \kappa_r(\tau+\nu) + i\kappa_i(\tau+\nu) \quad C^{-+}(t_1,t_2) = \kappa_r(\tau-\nu) - i\kappa_i(\tau-\nu) \tag{31}$$

where we have again stated the form these kernels take away from the initial and final time. Substituting these correlation functions into (29) we obtain equation (20). Note that only the difference between the map for ρ derived and discussed in section 2 and the map for the generating function is the cross correlations.

5. An-harmonic baths and cluster expansions

A second advantage of the super-operator formulation is in the derivation of corrections to the Feynman–Vernon theory. The starting point is then the dynamical map (26) with the multi-time correlation functions of the environment (27), but without assuming Wick's theorem. The outcome will be that multi-time cumulants of the environment (discussed below) translate into kernels of higher-than-quadratic contributions to the Feynman–Vernon action.

For ordinary operator correlation functions, successive orders of cumulants are defined inductively as

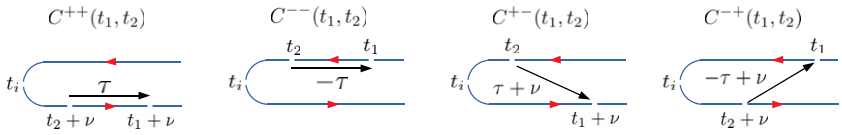

Figure 1. Time-line diagrams for two-time correlation for $\tau = t_1 - t_2 > 0$. The upper (lower) branch shows anti-chronological (chronological) time line. The time on chronological branch shifts by ν. For $C^{++}(t_1,t_2)$, both times shift by the same amount and thus the time difference is not affected by the shift. The situation for $C^{--}(t_1,t_2)$ is identical to the normal correlation. For the cross correlation $C^{+-}(t_1,t_2)$, only t_1 on the chronological branch shifts and thus the time difference also shifts. The situation for $C^{-+}(t_1,t_2)$ is similar to $C^{+-}(t_1,t_2)$ except that t_2 shifts by ν instead of t_1. For $t_1 < t_2$, the direction of τ is reversed for $C^{++}(t_1,t_2)$ and $C^{--}(t_1,t_2)$. However, the sign of $t_1 - t_2$ does not affect the situation of $C^{+-}(t_1,t_2)$ and $C^{-+}(t_1,t_2)$.

$$\begin{aligned}
G_1(t_1) &= C(t_1)\\
G_2(t_1,t_2) &= C(t_1,t_2) - G_1(t_1)G_1(t_2)\\
G_3(t_1,t_2,t_3) &= C(t_1,t_2,t_3) - G_1(t_1)G_1(t_2)G_1(t_3) - G_1(t_1)G_2(t_2,t_3)\\
&\quad - G_1(t_2)G_2(t_1,t_3) - G_1(t_3)G_2(t_1,t_2)\\
&\;\;\vdots
\end{aligned} \tag{32}$$

Owing to the time-ordering super-operator $\overleftarrow{\mathcal{T}}$ and indexes d_j, $C^{d_1,\ldots,d_n}(t_1,\ldots,\ t_n)$ defined in equation (27) behaves like an ordinary multi-time correlation function and the relations (32) hold. Hence,

$$C^{d_1,\cdots d_N}(t_1,\ldots,\ t_N) = \sum_{\substack{\text{all possible}\\ \text{groupings}}} \prod_{\substack{\text{groups of}\\ \text{one time}}} G_1^{d_1}(t_1) \prod_{\substack{\text{groups of}\\ \text{two times}}} G_2^{d_1,d_2}(t_2,t_3) \quad \cdots \tag{33}$$

where N can be even or odd. The first order cumulant (G_1^d) can be set to zero by a shift. The first non-trivial cumulant is then

$$\begin{aligned}
G_4^{d_1,d_2,d_3,d_4}(t_1,t_2,t_3,t_4) &= C^{d_1,d_2,d_3,d_4}(t_1,t_2,t_3,d_4) - C^{d_1,d_2}(t_1,t_2)C^{d_3,d_4}(t_3,t_4)\\
&\quad - C^{d_1,d_3}(t_1,t_3)C^{d_2,d_4}(t_2,t_4) - C^{d_1,d_4}(t_1,t_4)C^{d_2,d_3}(t_2,t_3)
\end{aligned} \tag{34}$$

where we have retained the super-operator notation on the right-hand side. For a bath that satisfies Wick's theorem, this cumulant and all others beyond $G_2^{d_1,d_2}$ vanish.

The second step is to count the number of groupings in (33) with n_1 groups of one element, n_2 groups of two elements (pairs), n_3 groups of three elements, etc. There are $\frac{N!}{n_1!n_2!(2!)^{n_2}n_3!(3!)^{n_3}\cdots}$ such groupings. The correlation functions appear inside the time integral and index sums in (26) and the indices and time variables can therefore be renamed in any way. Each grouping of the same type (same $n_1, n_2, \ldots$) hence contributes the same, and the quantum map can be summed in an analogous way to section 4.

Introducing $Q(t)$ as the coordinate representation of $\mathcal{X}_s^+(t)$ and $-\tilde{Q}(t)$ the coordinate representation of $\mathcal{X}_s^-(t)$ one can show that the contribution to the Feynman–Vernon action from n number of X and m number Y is

$$S^{(n,m)} = (-i)^n (i)^m \int_{t_0}^{t} \mathrm{d}s_1 \int_{t_0}^{s_1} \mathrm{d}s_2 \cdots \int_{t_0}^{t} \mathrm{d}u_1 \int_{t_0}^{u_1} \mathrm{d}u_2 \ \cdots \ Q(s_1)Q(s_2)\cdots Q(s_n)$$
$$\times \tilde{Q}(u_1)\tilde{Q}(u_2) \ \cdots \ \tilde{Q}(u_m) G_{n+m}(u_m, \ldots, \ u_1, s_1, \ldots, \ s_n) \tag{35}$$

where the last term is the cumulant of the operator correlation function with the times ordered as required in the super-operator cumulant. One can further sum all contributions of the same order and express them in terms of time-ordered sums $\zeta_+(t) = Q(t) + \tilde{Q}(t)$ and differences $\zeta_-(t) = Q(t) - \tilde{Q}(t)$. The most important general result one can find this way is for the largest time, the dependence in only through the difference $\zeta_-(t)$ as also follows from Feynman and Vernon's *General property of influence functionals 5* [9]. Ultimately this is a consequence of the super-operator correlation function $C^{d_1,\ldots,d_n}(t_1, \ldots, \ t_n)$ being independent of the symbol connected to the largest time. Otherwise the general expressions are somewhat unwieldy, and we will here only quote the result to third order

$$\sum_{n+m=3} S^{(n,m)} = \frac{i}{4} \int_{t_0}^{t} \mathrm{d}s \zeta_-(s) \int_{t_0}^{s} \mathrm{d}u \int_{t_0}^{u} \mathrm{d}v$$
$$\times \left(\zeta_+(u)\zeta_+(v)A + \zeta_+(u)\zeta_-(v)B + \zeta_-(u)\zeta_+(v)C + \zeta_-(u)\zeta_-(v)D \right) \tag{36}$$

where A, B, C and D are combinations of third order bath correlation functions given in appendix C.

6. Discussion

In this paper we have compared the path integral and super-operator approaches to the theory of open quantum system (OQS). We have pointed out that both approaches lead to equivalent descriptions of a system interacting with one or several harmonic oscillator baths, but that the routes to the result are qualitatively different. In the super-operator approach the kernels in the description are found to be certain pair correlation functions of the bath (or baths), and the main assumption is Wick's theorem, reducing any correlation function to sums or products of pair correlation functions. In the path integral approach, the result on the hand follow from integrating over the initial and final points of the propagator of an harmonic oscillator (one of the degrees of freedom of the bath) acted upon by a linear drive (a linear interaction with the system), and after a fair amount of cancellation.

We have here shown that same holds for the generating function of heat: both approaches give the same result, but the super-operator approach is more direct. In particular, the fact that the generating function of heat can be expressed with the same kernels as for the system density matrix (Feynman–Vernon theory), with only a time shift in the terms mixing the forward and time-revered paths, follows in a much more straight-forward manner in the super-operator approach.

We have also shown that the super-operator approach extends in a natural way to interactions with environments where Wick's theorem does not hold. Cumulants of correlation functions of the environment, which vanish when Wick's theorem holds, hence translate to kernels in higher-order terms in the Feynman–Vernon action. In the text we have discussed that the resulting higher-order theory of the influence functional satisfies the general properties stated by Feynman and Vernon. Considerations of when the higher-order terms are comparable or more important than the Feynman–Vernon terms are left for future work.

Apart from the foundational contribution [7] discussed in Introduction, we also mention two more recent contributions that use a similar plus/minus (left/right) representation of the super-operator [48, 49]; the latter paper also extends the analysis beyond harmonic baths, though in a different manner than we do. Time shifts in kernels describing a statistics of heat have also appeared earlier in the theory of heat transport through a Josephson junction [50], though for a partially classical model. We hope to have put these earlier results in a coherent whole and in the context of current concerns in quantum thermodynamics.

We end by summarize the assumptions that go and do not go into the new higher-order theory we have developed here. First, we assume that the system and the environment start out in a product state. Second, we assume that it is possible to write the system-environment interaction as $V_{\rm sb} = \sum_k X_{\rm s}^k \otimes B_{\rm b}^k$, where $X_{\rm s}^k$ and $B_{\rm b}^k$ are operators on respectively the system and the environment, and where all the $B_{\rm b}^k$ commute. Third, we assume that the initial state of the environment is a product state compatible with the interaction. By the latter we mean that if the full environment Hilbert space is a product space $\mathcal{H}_{\rm b} = \mathcal{H}_{\rm b}^1 \otimes \mathcal{H}_{\rm b}^2 \cdots$ and the operators $B_{\rm b}^j$ act on $\mathcal{H}_{\rm b}^j$, then the initial environment density matrix factorizes as as $\rho_{\rm b} = \rho_{\rm b}^1 \otimes \rho_{\rm b}^2 \cdots$ where $\rho_{\rm b}^j$ is a unit trace positive Hermitian operator on $\mathcal{H}_{\rm b}^j$. One class of models that fulfill the above is when the system interacts with one or several baths which start out independent, and which do not interact between themselves. In the other direction, in each bath the environmental degrees of freedom can be either Bosonic or Fermionic (or both), and the Hamiltonians can be arbitrary. The initial state of each bath does not even have to be in equilibrium. We suspect that such a general-looking result will find applications also outside the current realm of theory of open quantum system.

Acknowledgments

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Appendix A. Unitary time evolution of a density operator and time-ordering super-operator

We consider first unitary time-evolution of a ket $|\psi(t)\rangle$ and a bra $\langle\psi(t)|$ under a Hamiltonian $H = H_0 + V$ where H_0 is an unperturbed Hamiltonian and V a perturbation. Using the interaction picture $V(t) = {\rm e}^{{\rm i}H_0 t} V {\rm e}^{-{\rm i}H_0 t}$ the time evolution of the ket and bra can be expressed with a time evolution operator.

$$|\psi(t)\rangle = U(t,t_i)\,|\psi(t_i)\rangle\,, \quad \langle\psi(t)| = \langle\psi(t_i)|\,U^\dagger(t,t_i) \tag{A.1}$$

where the forward and backward evolution operators are defined by

$$U(t,t_i) = \overleftarrow{T}\ \exp\left[-i\int_{t_i}^{t} V(s)\,{\rm d}s\right] \tag{A.2a}$$

$$U^\dagger(t,t_i) = \overrightarrow{T}\ \exp\left[i\int_{t_i}^{t} V(s)\,{\rm d}s\right] \tag{A.2b}$$

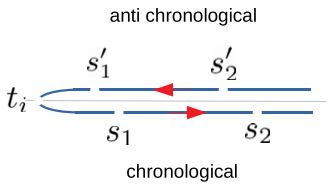

Figure A1. Time evolution of density operator, the operators $\widetilde{V}(s)$ are ordered along the time line from t to t_i (anti-chronological order) and then from t_i to t (chronological order). This particular diagram shows the case of $V(s_2)V(s_1)\rho(t_i)V(s_1')V(s_2')$.

where $\overleftarrow{T}$ and $\overrightarrow{T}$ are chronological and anti-chronological time ordering operator.

The evolution of a density operator involves both forward and backward evolution operators as

$$\rho(t) = U(t, t_i)\rho(t_i)U^{\dagger}(t, t_i). \tag{A.3}$$

Managing the order of operators is a bit complicated due to the presence of two evolutions. There is a simpler expression using the time line shown in figure A1. We note that the evolution of the density operator is determined by the Liouville–von Neumann equation

$$\frac{\mathrm{d}\rho(t)}{\mathrm{d}t} = \mathcal{L}(t)\rho(t) \tag{A.4}$$

where the Liouville super-operator is defined by $\mathcal{L}(t) = -i[V(t), \bullet]$. Then,

$$\rho(t) = \overleftarrow{\mathcal{T}}\ \exp\left[\int_{t_i}^{t} \mathcal{L}(s)\mathrm{d}s\right]\rho(t_i) \tag{A.5}$$

where the time-ordering super-operator $\overleftarrow{\mathcal{T}}$ orders super-operators such as $\mathcal{L}(t)$ chronologically. It automatically orders regular operators along the time line shown in figure A1. As an example, consider $t_1 > t_2$,

$$\begin{aligned}\overleftarrow{\mathcal{T}}\mathcal{L}(t_2)\mathcal{L}(t_1)\rho(t_i) &= \mathcal{L}(t_1)\mathcal{L}(t_2)\rho(t_i)\\ &= V(t_1)V(t_2)\rho(t_i) - V(t_1)\rho(t_i)V(t_2) - V(t_2)\rho(t_i)V(t_1) + \rho(t_i)V(t_2)V(t_1)\end{aligned} \tag{A.6}$$

which automatically orders $V(t)$ chronologically if it is on the left of $\rho(t_i)$ and anti-chronologically on the right.

Appendix B. Pair correlation functions

Now, we evaluate the four pair correlation functions C^{++}, C^{+-}, C^{-+}, and C^{--} and express them with an ordinary correlation function

$$C(t_1, t_2) = \langle Y_{\mathrm{c}}(t_1)Y_{\mathrm{c}}(t_2)\rho_{\mathrm{c}}(t_i)\rangle = \kappa_r(t_1 - t_2) - i\kappa_i(t_1 - t_2) \tag{B.1}$$

where $\kappa_r(\tau)$ and $\kappa_i(\tau)$ are shown in equation (7).

For the diagonal ones, we find the exactly the same correlation functions as those in the influential function as follows:

$$
\begin{aligned}
C^{++}(t_1,t_2) &= \mathrm{tr_c}\left\{\overleftarrow{\mathcal{T}}\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{+}(t_1)\mathcal{Y}_\mathrm{c}^{+}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} \\
&= \begin{cases} \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}Y_\mathrm{c}(t_1)Y_\mathrm{c}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} & t_1 > t_2 \\ \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}Y_\mathrm{c}(t_2)Y_\mathrm{c}(t_1)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} & t_1 < t_2 \end{cases} \\
&= \begin{cases} \langle Y_\mathrm{c}(t_1)Y_\mathrm{b}(t_2)\rangle = C(t_1,t_2) & t_1 > t_2 \\ \langle Y_\mathrm{c}(t_2)Y_\mathrm{b}(t_1)\rangle = C^*(t_1,t_2) & t_1 < t_2 \end{cases}
\end{aligned} \tag{B.2}
$$

$$
\begin{aligned}
C^{--}(t_1,t_2) &= \mathrm{tr_c}\left\{\overleftarrow{\mathcal{T}}\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}Y_\mathrm{c}^{<}(t_1)Y_\mathrm{c}^{<}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} \\
&= \begin{cases} \mathrm{tr_c}\left\{\rho_\mathrm{c}(t_i)Y_\mathrm{c}(t_2)Y_\mathrm{c}(t_1)\right\} & t_1 > t_2 \\ \mathrm{tr_c}\left\{\rho_\mathrm{c}(t_i)Y_\mathrm{c}(t_1)Y_\mathrm{c}(t_2)\right\} & t_1 < t_2 \end{cases} \\
&= \begin{cases} \langle Y_\mathrm{c}(t_2)Y_\mathrm{c}(t_1)\rangle = C(t_1,t_2)^* & t_1 > t_2 \\ \langle Y_\mathrm{c}(t_1)Y_\mathrm{c}(t_2)\rangle = C(t_1,t_2) & t_1 < t_2 \end{cases}
\end{aligned} \tag{B.3}
$$

where we used $[\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}, \rho_\mathrm{c}(t_i)] = 0$. A standard correlation function $C(t_1,t_2) = \langle Y_\mathrm{b}(t_1)Y_\mathrm{b}(t_2)\rangle$ and its complex conjugate $C^*(t_1,t_2) = \langle Y_\mathrm{b}(t_2)Y_\mathrm{b}(t_1)\rangle$ are used in the final expression. Notice that these two correlation functions are exactly the same as ones in the influence functional.

However, the off-diagonal ones are different

$$
\begin{aligned}
C^{+-}(t_1,t_2) &= \mathrm{tr_c}\left\{\overleftarrow{\mathcal{T}}\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{>}(t_1)\mathcal{Y}_\mathrm{c}^{<}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} \\
&= \begin{cases} \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{>}(t_1)\mathcal{Y}_\mathrm{c}^{<}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} & t_1 > t_2 \\ \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{<}(t_2)\mathcal{Y}_\mathrm{c}^{>}(t_1)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} & t_2 > t_1 \end{cases} \\
&= \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}Y_\mathrm{c}(t_1)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\,\rho_\mathrm{c}(t_i)\,Y_\mathrm{c}(t_2)\right\} \\
&= \langle Y_\mathrm{c}(t_2)Y_\mathrm{c}(t_1+\nu)\rangle = C(t_2,t_1+\nu)
\end{aligned} \tag{B.4}
$$

$$
\begin{aligned}
C^{-+}(t_1,t_2) &= \mathrm{tr_c}\left\{\overleftarrow{\mathcal{T}}\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{<}(t_1)\mathcal{Y}_\mathrm{c}^{>}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} \\
&= \begin{cases} \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{<}(t_1)\mathcal{Y}_\mathrm{c}^{>}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} & t_1 > t_2 \\ \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}\mathcal{Y}_\mathrm{c}^{>}(t_2)\mathcal{Y}_\mathrm{c}^{<}(t_1)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\rho_\mathrm{c}(t_i)\right\} & t_2 > t_1 \end{cases} \\
&= \mathrm{tr_c}\left\{\mathrm{e}^{\mathrm{i}\nu H_\mathrm{c}}Y_\mathrm{c}(t_2)\mathrm{e}^{-\mathrm{i}\nu H_\mathrm{c}}\,\rho_\mathrm{c}(t_i)\,Y_\mathrm{c}(t_1)\right\} \\
&= \langle Y_\mathrm{c}(t_1)Y_\mathrm{c}(t_2+\nu)\rangle = C(t_1,t_2+\nu)
\end{aligned} \tag{B.5}
$$

where time on the chronological branch shifts by ν.

Appendix C. Time-ordered cumulant expansion

The starting point is an expansion analogous to (29) but using the cumulant expansion (33) instead of Wick's theorem. Both even and odd terms may appear. We

can consider interchanges within one group, say $G_3^{d_1,d_2,d_3}(t_1,t_2,t_3)G_3^{d_4,d_5,d_6}(t_4,t_5,t_6)$ with $G_3^{d_1,d_2,d_4}(t_1,t_2,t_4)G_3^{d_3,d_5,d_6}(t_3,t_5,t_6)$, which will contribute the same. This means that the quantum map can now be simplified to

$$\begin{aligned}\mathcal{M}(t_f;t_0) &= \sum_N \frac{(-i)^N}{N!} \sum_{n_1+2n_2+3n_3+\cdots=N} \frac{N!}{n_1!n_2!(2!)^{n_2}n_3!(3!)^{n_3}\cdots} \int_{t_i}^{t_f} \mathrm{d}t_1 \cdots \int_{t_i}^{t_f} \mathrm{d}t_N \\ &\quad \times \sum_{d_1} \cdots \sum_{d_N} \overleftarrow{\mathcal{T}} \left\{ \mathcal{X}_\mathrm{s}^{d_1}(t_1) \ \cdots \ \mathcal{X}_\mathrm{s}^{d_N}(t_N) \right\} \prod_i G_1^{d_i}(t_i) \prod_{ij} G_2^{d_i,d_j}(t_i,t_j) \cdots \\ &= \overleftarrow{\mathcal{T}} \exp\Bigg(\frac{(-i)^2}{2!} \int_{t_0}^{t_f} \mathrm{d}t_1 \int_{t_0}^{t} \mathrm{d}t_2 \sum_{d_1,d_2} G_2^{d_1,d_2}(t_1,t_2)\mathcal{X}_\mathrm{s}^{d_1}(t_1)\mathcal{X}_\mathrm{s}^{d_2}(t_2) \\ &\quad + \frac{(-i)^3}{3!} \int_{t_i}^{t_f} \mathrm{d}t_1 \int_{t_i}^{t_f} \mathrm{d}t_2 \int_{t_i}^{t_f} \mathrm{d}t_3 \sum_{d_1,d_2,d_3} G_3^{d_1,d_2,d_3} \\ &\quad \times (t_1,t_2,t_3)\mathcal{X}_\mathrm{s}^{d_1}(t_1)\mathcal{X}_\mathrm{s}^{d_2}(t_2)\mathcal{X}_\mathrm{s}^{d_3}(t_3) + \cdots \Bigg) \end{aligned} \tag{C.1}$$

In the last equation we have used that the operators have zero mean, $G_1 = 0$. The second order term is the standard Feynman–Vernon expansion as evaluated above.

The third term can be evaluated as follows. The cumulants with the same indexes G_3^{+++} and G_3^{---} remain the same when the times are permuted. This can be done in 3! different ways. If only two of the indexes d_i and d_j are the same, e.x., G_3^{++-}, give a factor two if permuted, and this can be done in three different ways. Furthermore $G_3^{++-}(t_1,t_2,t_3)$ equals $C(t_3,t_1,t_2)$ if $t_1 > t_2$ and $C(t_3,t_2,t_1)$ if $t_1 < t_2$ where C is the operator correlation function, and similarly for the other cases. Introducing for convenience Q_t the coordinate representation of $\mathcal{X}_\mathrm{s}^+(t)$ and $-\tilde{Q}_t$ the coordinate representation of $\mathcal{X}_\mathrm{s}^-(t)$, and $S^{(n,m}$ for the terms with n number of Q and m number of $\tilde{Q}$, the sum of all terms to third order is thus

$$\begin{aligned}\sum_{n+m=3} S^{(n,m)} &= i\left(\int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}u \int_{t_i}^{u} \mathrm{d}v Q(s)Q(u)Q(v)C(s,u,v) - \tilde{Q}(s)\tilde{Q}(u)\tilde{Q}(v)C(v,u,s) \right) \\ &\quad + i\left(\int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}u \int_{t_i}^{u} \mathrm{d}v \left(\tilde{Q}(s)\tilde{Q}(u)Q(v)C(u,s,v) - Q(s)Q(u)\tilde{Q}(v)C(v,s,u) \right) \right) \end{aligned} \tag{C.2}$$

A similar argument can be made for a term of order N. There are $\frac{N!}{n!(N-n)!}$ ways to select n indexes to be $+$, and $N-n$ indexes to be $-$. By the time ordering the cumulant G_N give the same if the times in the two groups are permuted within themselves which can be done in $n! \times (N-n)!$ ways. The contribution from n forward paths (Q) and m backward paths ($\tilde{Q}$) is thus n time-ordered and m reverse time-ordered integrals multiplying the corresponding correlation function, which is (35) in main text.

Equation (C.2) can be analyzed further by considering in the mixed terms the three ranges of v: less than u; between u and s, and larger than s. Renaming the variables so that times are always ordered $s > u > v$ this gives

$$\sum_{n+m=3} S^{(n,m)} = i\int_{t_i}^{t_f} \mathrm{d}s \int_{t_i}^{s} \mathrm{d}u \int_{t_i}^{u} \mathrm{d}v \Big(Q(s)Q(u)Q(v)C(s,u,v) - Q(s)Q(u)\tilde{Q}(v)C(v,s,u)$$
$$- Q(s)\tilde{Q}(u)Q(v)C(u,s,v) - \tilde{Q}(s)Q(u)Q(v)C(s,u,v) + \tilde{Q}(s)\tilde{Q}(u)Q(v)C(u,s,v)$$
$$+ \tilde{Q}(s)Q(u)\tilde{Q}(v)C(v,s,u) + Q(S)\tilde{Q}(u)\tilde{Q}(v)C(v,u,s) - \tilde{Q}(s)\tilde{Q}(u)\tilde{Q}(v)C(v,u,s)\Big) \tag{C.3}$$

Collecting terms with the same last entries one sees that this is

$$\sum_{n+m=3} S^{(n,m)} = i\int_{t_i}^{t_f} \mathrm{d}s \left(Q(s) - \tilde{Q}(s)\right) \int_{t_i}^{s} \mathrm{d}u \int_{t_i}^{u} \mathrm{d}v \,(Q(u)Q(v)C(s,u,v)$$
$$- Q(u)\tilde{Q}(v)C(v,s,u) - \tilde{Q}(u)Q(v)C(u,s,v) + \tilde{Q}(u)\tilde{Q}(v)C(v,u,s)\Big) \tag{C.4}$$

The third-order terms hence satisfy the general property of the Feynman–Vernon action that if $Q(s) = \tilde{Q}(s)$ for all s greater than τ, then the action does not depend on $Q(s)$ or $\tilde{Q}(s)$ for $s > \tau$. This conclusion also holds more generally: starting from (35) in main text one can first insert u_1 in any of the intervals $[t_f; s_1]$, $[s_1; s_2]$, …, $[s_n; t_i]$, then u_2 in the same interval as u_1 or any one further down the list, and so on. Each such insertion can be identified by a sequence $Z_{\tau_1}, Z_{\tau_2}, \ldots,\ Z_{\tau_N}$ where each symbol is Q or $\tilde{Q}$, and the times are ordered $\tau_1 > \tau_2 > \cdots > \tau_N$. Consider now two cases that only differ by the symbol Z_{τ_1}. The first case has n symbols Q and $N-n$ symbols $\tilde{Q}$ $(0 < n \leqslant N)$, and arises from inserting $(u_1, u_2, \ldots, u_{N-n})$ in $(s_1, s_2, \ldots, s_n)$ such that u_1 falls in one of the intervals $[s_1; s_2]$, …, $[s_n; t_0]$. The second case has on the other hand $n-1$ symbols Q and $N-n+1$ symbols $\tilde{Q}$ $(0 \leqslant n-1 < N)$ and arises from inserting $(u', u_1, u_2, \ldots, u_{N-n})$ in $(s_2, \ldots, s_n)$ such that $u' = s_1$ and $u_1, u_2, \ldots.$ fall as in the first case. The corresponding cumulant is in both cases $G_N(u_{N-n}, \ldots, u_1, s_1, s_2, \ldots, s_n)$ which does not depend on the symbol of the largest time $(u' = s_1)$. Each such combination is therefore proportional to $Q(s_1) - \tilde{Q}(s_1)$.

If written in terms of the $\zeta_+(s) = Q(s) + \tilde{Q}(s)$ and $\zeta_-(s) = Q(s) - \tilde{Q}(s)$, equation (C.4) can further be expressed as

$$\sum_{n+m=3} S^{(n,m)} = \frac{i}{4}\int_{t_i}^{t_f} \mathrm{d}s\zeta_-(s) \int_{t_i}^{s} \mathrm{d}u \int_{t_i}^{u} \mathrm{d}v \,\big(\zeta_+(u)\zeta_+(v)A + \zeta_+(u)\zeta_-(v)B$$
$$+ \zeta_-(u)\zeta_+(v)C + \zeta_-(u)\zeta_-(v)D\big) \tag{C.5}$$

where the combined amplitudes can be written out as

$$A = \mathrm{tr_b}\,[Y_\mathrm{b}(s)\,(Y_\mathrm{b}(u)Y_\mathrm{b}(v)\rho - Y_\mathrm{b}(u)\rho Y_\mathrm{b}(v) - Y_\mathrm{b}(v)\rho Y_\mathrm{b}(u) + \rho Y_\mathrm{b}(u)Y_\mathrm{b}(v))] \tag{C.6}$$

$$B = \mathrm{tr_b}\,[Y_\mathrm{b}(s)\,(Y_\mathrm{b}(u)Y_\mathrm{b}(v)\rho + Y_\mathrm{b}(u)\rho Y_\mathrm{b}(v) - Y_\mathrm{b}(v)\rho Y_\mathrm{b}(u) - \rho Y_\mathrm{b}(u)Y_\mathrm{b}(v))] \tag{C.7}$$

$$C = \mathrm{tr_b}\,[Y_\mathrm{b}(s)\,(Y_\mathrm{b}(u)Y_\mathrm{b}(v)\rho - Y_\mathrm{b}(u)\rho Y_\mathrm{b}(v) + Y_\mathrm{b}(v)\rho Y_\mathrm{b}(u) - \rho Y_\mathrm{b}(u)Y_\mathrm{b}(v))] \tag{C.8}$$

$$D = \mathrm{tr_b}\,[Y_\mathrm{b}(s)\,(Y_\mathrm{b}(u)Y_\mathrm{b}(v)\rho + Y_\mathrm{b}(u)\rho Y_\mathrm{b}(v) + Y_\mathrm{b}(v)\rho Y_\mathrm{b}(u) + \rho Y_\mathrm{b}(u)Y_\mathrm{b}(v))] \tag{C.9}$$

By comparison, the standard Feynman–Vernon action can be written in a similar way as

$$\sum_{n+m=2} S^{(n,m)} = -\frac{1}{2}\int_{t_i}^{t_f} \mathrm{d}s\zeta_-(s) \int_{t_i}^{s} \mathrm{d}u \,\big(\zeta_+(u)A' + \zeta_-(u)B'\big) \tag{C.10}$$

where

$$A' = \mathrm{tr_b}\left[Y_\mathrm{b}(s)\left(Y_\mathrm{b}(u)\rho - \rho Y_\mathrm{b}(u)\right)\right] \tag{C.11}$$

$$B' = \mathrm{tr_b}\left[Y_\mathrm{b}(s)\left(Y_\mathrm{b}(u)\rho + \rho Y_\mathrm{b}(u)\right)\right] \tag{C.12}$$

The contributions from the fourth order cumulants are analogously found to be

$$\begin{aligned}\sum_{n+m=4} S^{(n,m)} = \int_{t_i}^{t_f} \mathrm{d}s \left(Q(s) - \tilde{Q}(s)\right) \int_{t_i}^{s} \mathrm{d}u \int_{t_i}^{u} \mathrm{d}v \int_{t_i}^{v} \mathrm{d}w \Big(& Q(u)Q(v)Q(w)G_4(s,u,v,w) \\ & - \tilde{Q}(u)\tilde{Q}(v)\tilde{Q}(w)G_4(w,v,u,s) - Q(u)Q(v)\tilde{Q}(w)G_4(w,s,u,v) \\ & - Q(u)\tilde{Q}(v)Q(w)G_4(v,s,u,w) - \tilde{Q}(u)Q(v)Q(w)G_4(u,s,v,w) \\ & + \tilde{Q}(u)\tilde{Q}(v)Q(w)G_4(v,u,s,w) + \tilde{Q}(u)Q(v)\tilde{Q}(w)G_4(w,u,s,v) \\ & + Q(u)\tilde{Q}(v)\tilde{Q}(w)G_4(w,v,s,u)\Big)\end{aligned} \tag{C.13}$$

which can be re-written

$$\sum_{n+m=4} S^{(n,m)} = \frac{1}{8}\int_{t_i}^{t_f} \mathrm{d}s\zeta_-(s) \int_{t_i}^{s} \mathrm{d}u \int_{t_0}^{u} \mathrm{d}v \int_{t_0}^{v} \mathrm{d}w \sum_{pqr=\pm} \zeta_p(u)\zeta_q(v)\zeta_r(w)A_{pqr}(s,u,v,w,) \tag{C.14}$$

and

$$\begin{aligned}A_{+++} &= G_4(s,u,v,w) - G_4(w,v,u,s) - G_4(w,s,u,v) - G_4(v,s,u,w) \\ &\quad - G_4(u,s,v,w) + G_4(v,u,s,w) + G_4(w,u,s,v) + G_4(w,v,s,u) \\ A_{++-} &= G_4(s,u,v,w) + G_4(w,v,u,s) + G_4(w,s,u,v) - G_4(v,s,u,w) \\ &\quad - G_4(u,s,v,w) + G_4(v,u,s,w) - G_4(w,u,s,v) - G_4(w,v,s,u) \\ A_{+-+} &= G_4(s,u,v,w) + G_4(w,v,u,s) - G_4(w,s,u,v) + G_4(v,s,u,w) \\ &\quad - G_4(u,s,v,w) - G_4(v,u,s,w) + G_4(w,u,s,v) - G_4(w,v,s,u) \\ A_{-++} &= G_4(s,u,v,w) + G_4(w,v,u,s) - G_4(w,s,u,v) - G_4(v,s,u,w) \\ &\quad + G_4(u,s,v,w) - G_4(v,u,s,w) - G_4(w,u,s,v) + G_4(w,v,s,u) \\ A_{+--} &= G_4(s,u,v,w) - G_4(w,v,u,s) + G_4(w,s,u,v) + G_4(v,s,u,w) \\ &\quad - G_4(u,s,v,w) - G_4(v,u,s,w) - G_4(w,u,s,v) + G_4(w,v,s,u) \\ A_{-+-} &= G_4(s,u,v,w) - G_4(w,v,u,s) + G_4(w,s,u,v) - G_4(v,s,u,w) \\ &\quad + G_4(u,s,v,w) - G_4(v,u,s,w) + G_4(w,u,s,v) - G_4(w,v,s,u) \\ A_{--+} &= G_4(s,u,v,w) - G_4(w,v,u,s) - G_4(w,s,u,v) + G_4(v,s,u,w) \\ &\quad + G_4(u,s,v,w) + G_4(v,u,s,w) - G_4(w,u,s,v) - G_4(w,v,s,u) \\ A_{---} &= G_4(s,u,v,w) + G_4(w,v,u,s) + G_4(w,s,u,v) + G_4(v,s,u,w) \\ &\quad + G_4(u,s,v,w) + G_4(v,u,s,w) + G_4(w,u,s,v) + G_4(w,v,s,u)\end{aligned}$$

Appendix D. The path integral calculation of the generating function of heat for harmonic baths

We restate from the main text of the paper that the generating function of the energy change in one bath is defined as

$$G_{if}(\nu) = \mathrm{Tr}_B \langle f| \mathrm{e}^{\mathrm{i}\nu H_B} U \mathrm{e}^{-\mathrm{i}\nu H_B} \left(|i\rangle \langle i| \otimes \rho_B(\beta)\right) U^\dagger |f\rangle \tag{D.1}$$

where i and f are the initial and final state of the system, $\rho_B(\beta)$ is the initial thermal state of the bath at inverse temperature β and ν is the generating function parameter. In the Feynman–Vernon approach the two unitary operators U and $U^\dagger$, the final operator $\mathrm{e}^{\mathrm{i}\nu H_B}$, and the shifted initial thermal state of the $\mathrm{e}^{-\mathrm{i}\nu H_B}\rho_B(\beta)$ are all expressed as path integrals, and then the history of the bath is then integrated out.

For baths that are harmonic oscillators and for $\nu = 0$ this was done exactly by Feynman and Vernon, giving

$$P_{if} = G_{if}(\nu = 0) = \int_{if} \mathcal{D}X\mathcal{D}Y \mathrm{e}^{\frac{\mathrm{i}}{\hbar}S_S[X] - \frac{\mathrm{i}}{\hbar}S_S[Y] + \frac{\mathrm{i}}{\hbar}S_i^{\nu=0} - \frac{1}{\hbar}S_r^{\nu=0}} \tag{D.2}$$

where $\mathcal{D}X$ and $\mathcal{D}Y$ are integrals over the forward and backward system paths, S_i and S_r are the two terms in the Feynman–Vernon action from integrating out the bath, and $\int_{if}(\cdots)$ is a short-hand for projections on initial and final states. The Feynman–Vernon action is most commonly written as products of the sums and differences of the forward and backward paths, $X + Y$ and $X - Y$. When ν can also be different from zero it more convenient to instead write

$$\begin{aligned}\frac{i}{\hbar}S_i^{\nu=0}[X,Y] - \frac{1}{\hbar}S_r^{\nu=0}[X,Y] = {} & \frac{i}{\hbar}\int^t\int^s (XX' - YY')\kappa_i(s,s')\,\mathrm{d}s'\,\mathrm{d}s \\ & - \frac{1}{\hbar}\int^t\int^s (XX' + YY')\kappa_r(s,s')\,\mathrm{d}s'\,\mathrm{d}s \\ & + \frac{i}{\hbar}\int^t\int^s (XY' - X'Y)\kappa_i(s,s')\,\mathrm{d}s'\,\mathrm{d}s \\ & + \frac{1}{\hbar}\int^t\int^s (XY' + X'Y)\kappa_r(s,s')\,\mathrm{d}s'\,\mathrm{d}s \end{aligned} \tag{D.3}$$

where primed (unprimed) quantities refer to time s' (s) and the kernels κ_i and κ_r are given in (7) and (8) in main text.

Now consider the generating function $G_{if}(\nu)$ of (D.1). Since the path integrals for this quantity are also all Gaussian the path integrals pertaining to one harmonic oscillator reduce to a four-dimensional integral

$$\begin{aligned}\mathcal{F}_b = \int & \mathrm{d}x_i\mathrm{d}y_i\mathrm{d}x_f\mathrm{d}y_f K^f(x_f, y_f, \hbar\nu) K(x_f, x_i, t_f - t_i; X) \\ & \times K^*(y_f, y_i, t_f - t_i; Y) K^f(x_i, y_i, -\hbar\nu + i\beta)\end{aligned} \tag{D.4}$$

where K^f is the free propagator of the bath and $K(\cdot; X)$ is the propagator of the bath interacting linearly with an classical time-dependent field X, and similarly for $K^*(\cdot; Y)$. These propagators

contain terms constant, linear and quadratic in the initial and final point of each propagator. The quadratic terms are the same for the free and the interacting propagators, the linear terms are integrals in the driving fields (X and Y, respectively) and the constant term is one double integral in X minus one double integral in Y.

By algebraic manipulation given in [35] (appendix) one can reduce $\mathcal{F}_b$ to $\mathrm{e}^{\frac{i}{\hbar}S_i^{\nu=0}-\frac{1}{\hbar}S_r^{\nu=0}+\mathcal{J}^{(2)}+\mathcal{J}^{(3)}}$ where

$$\mathcal{J}^{(2)} = \frac{i}{2\,\mathrm{m}\omega\hbar}\int^t\int^t (XY'-X'Y)CC'\ \sin\ \omega(s-s')\left(\frac{yz'-y'z}{\Delta}-\frac{1}{2}\right) \tag{D.5}$$

$$\mathcal{J}^{(3)} = \frac{i}{2\,\mathrm{m}\omega\hbar}\int^t\int^t (XY'+X'Y)CC'\ \cos\ \omega(s-s')\left(\frac{z'-y'}{\Delta}+\frac{i}{2}\coth\frac{\omega\hbar\beta}{2}\right) \tag{D.6}$$

which depend on auxiliary parameters as follows: $x=\cot(\omega t)$, $x'=\sin^{-1}(\omega t)$, $y=\cot(\omega\hbar\nu)$, $y'=\sin^{-1}(\omega\hbar\nu)$, $z=\cot(\omega\hbar(\nu-i\beta))$ and $z'=\sin^{-1}(\omega\hbar(\nu-i\beta))$. Δ is the combination $2(z'y'-yz-1)$. Indices (2) and (3) in (D.5) and (D.6) are given for back-compatibility, and do not matter in the present discussion. In [35] the amplitude of $\mathcal{J}^{(2)}$ in (D.5) was incorrectly given as $\left(\frac{y'z'-yz}{\Delta}-\frac{1}{2}\right)$; the error was corrected in [36].

We can now simplify to

$$\Delta = 2\ \sin^{-1}(\omega\hbar(\nu-i\beta))\sin^{-1}(\omega\hbar\nu)\,(1-\cosh(\omega\hbar\beta)) \tag{D.7}$$

$$\frac{yz'-y'z}{\Delta} = \frac{1}{2}\ \cos(\omega\hbar\nu)+\frac{i}{2}\ \sin(\omega\hbar\nu)\coth\left(\frac{\omega\hbar\beta}{2}\right) \tag{D.8}$$

$$\frac{z'-y'}{\Delta} = -\frac{i}{2}\ \cos(\omega\hbar\nu)\coth\left(\frac{\omega\hbar\beta}{2}\right)+\frac{1}{2}\ \sin(\omega\hbar\nu) \tag{D.9}$$

and rewrite the integrands in (D.5) and (D.6). For terms proportional to $XY'CC'$ we have

$$\begin{aligned}\text{Expr.} = \sin\ \omega(s-s')&\left(\frac{1}{2}\ \cos(\omega\hbar\nu)+\frac{i}{2}\ \sin(\omega\hbar\nu)\coth\left(\frac{\omega\hbar\beta}{2}\right)\right)\\ &+\cos\ \omega(s-s')\left(-\frac{i}{2}\ \cos(\omega\hbar\nu)\coth\left(\frac{\omega\hbar\beta}{2}\right)+\frac{1}{2}\ \sin(\omega\hbar\nu)\right)\end{aligned}$$

By trigonometry this is $\frac{1}{2}\ \sin\ \omega(s-s'+\hbar\nu)-\frac{i}{2}\ \cos\ \omega(s-s'+\hbar\nu)\coth(\frac{\omega\hbar\beta}{2})$. The terms proportional to $X'YCC'$ are similarly $-\frac{1}{2}\ \sin\ \omega(s-s'-\hbar\nu)-\frac{i}{2}\ \cos\ \omega(s-s'-\hbar\nu)\coth(\frac{\omega\hbar\beta}{2})$. Exchanging labels and including the integrals and the prefactors in (D.5) and (D.6) the cross-terms between the forward and backward paths for the generating function are hence

$$\begin{aligned}\text{Cross}-\text{terms} = &\frac{i}{\hbar}\int^t\int^t XY'\sum_b C_bC_b'\frac{1}{2m_b\omega_b}\sin\ \omega_b(s-s'+\hbar\nu)\,\mathrm{d}s'\,\mathrm{d}s\\ &+\frac{1}{\hbar}\int^t\int^t XY'\sum_b C_bC_b'\frac{1}{2m_b\omega_b}\cos\ \omega_b(s-s'+\hbar\nu)\coth(\frac{\omega_b\hbar\beta}{2})\,\mathrm{d}s'\,\mathrm{d}s\end{aligned}$$

Comparing to the cross-terms in (D.3) this is but a simple time shift of the arguments of the sines and the cosines.

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