


Thermodynamic nonlinear response relation

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The fluctuation-dissipation theorem connects equilibrium to mildly (linearly) perturbed situations in a thermodynamic manner: It involves the observable of interest and the entropy production caused by the perturbation. We derive a relation which connects responses of arbitrary order in perturbation strength to correlations of entropy production of lower order, thereby extending the fluctuation-dissipation theorem to cases far from equilibrium in a thermodynamic way. The relation is validated and studied for a four-state model which is coarse-grained to a non-Markovian two-state model.

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I. INTRODUCTION

The theoretical footing of nonequilibrium states remains a fundamental challenge, despite important progress, e.g., given by fluctuation theorems and work relations [1–4]. When aiming at the nonequilibrium responses, the powerful *fluctuation-dissipation theorem* (FDT) provides the leading order. It states that measurements of thermodynamic fluctuations in the unperturbed equilibrium system predict behaviors of the perturbed one in linear order [5–9]. With stronger perturbations, it fails, making it necessary to consider *nonlinear responses*, which have been addressed by the response formula from Kawasaki [10], transient time correlation functions [11–13], equilibrium correlations deduced for specific systems [14–18], as well as by generic approaches using operator formalisms [8,19] and path integrals [20–23].

An important observation is that the nonlinear responses are of fundamentally different nature as compared to FDT: Already at second order, dynamical details of the system enter [21]. Determining and measuring these dynamical details in the considered system, such as (interaction) potentials, hinders application of the mentioned approaches to macroscopic systems with many degrees of freedom [24]. The thermodynamic nature is thus lost in higher orders, causing the notion of a theorem to stop at first order.

We derive a formula for nonlinear responses to arbitrary order, which is distinct from known relations by its thermodynamic appearance. It consists of correlations of the entropy and the observable of interest. These are taken out of equilibrium as well, but one order lower than the response. For the linear response, one order lower is equilibrium, so that the derived relation extends the FDT to far from equilibrium scenarios in a natural manner. We demonstrate this formula for a non-Markovian jump process by calculating the responses for time independent as well as time dependent perturbation

up to third order. We also investigate statistical convergence for the examples provided.

II. DERIVATION OF THE RESPONSE FORMULA

Consider a classical system in weak contact with a thermal bath, described by a phase space x_s at time s . It is driven out of equilibrium by an additional time dependent term H_1 in the Hamiltonian, of the form

$$H_1(x_s, s) = \varepsilon h(s)V(x_s). \quad (1)$$

$V(x_s)$ is a potential, ε is a dimensionless expansion parameter setting the strength of the perturbation, and $h(s)$ is the perturbation protocol of order unity, being finite for $0 \leq s \leq t$. Using perturbations of potential type eases the presentation, and we will comment on other types later in the paper.

We continue by reviewing the standard approach via path integrals [20,21,25,26] to arrive at Eq. (5) below. The path weight $P^\varepsilon(\omega)$ for a path $\omega = (x_s)$, which describes the phase space configurations x_s of the system on the time interval $[0, t]$, enables the computation of the expectation value $\langle O(x_t) \rangle^\varepsilon$ of a state observable $O(x_t)$ via the path integral

$$\langle O(x_t) \rangle^\varepsilon = \int \mathcal{D}\omega O(x_t) P^\varepsilon(\omega). \quad (2)$$

$P^\varepsilon(\omega) = e^{-A_\varepsilon(\omega)} P(\omega)$ is split into a part comprising the perturbation via the nonequilibrium action $A_\varepsilon(\omega)$ [20,21,25,26] and the equilibrium path weight $P(\omega)$. The latter satisfies time-reversibility $P(\omega) = P(\Theta\omega)$. The sequence $\Theta\omega = (\pi x_{t-s})$, $0 \leq s \leq t$, represents the time-reversed path where the operator π reverses the sign of kinematic components. Out of equilibrium, time-reversal symmetry is broken and $P^\varepsilon(\omega)$ does not equal its time-reversed counterpart. When speaking of time-reversal, also the protocol $h(s)$ is reversed [23,26], and the corresponding path weights are denoted by tilde superscripts, $\tilde{P}^\varepsilon(\Theta\omega) = e^{-\tilde{A}_\varepsilon(\Theta\omega)} P(\Theta\omega)$. The breaking of time-reversibility is quantified by the entropy flux $S_\varepsilon(\omega)$

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towards the environment [20,21,25–28]

$$\begin{aligned} S_\varepsilon(\omega) &= \tilde{A}_\varepsilon(\Theta\omega) - A_\varepsilon(\omega) \\ &= \varepsilon\beta \left[h(t)V(x_t) - h(0)V(x_0) - \int_0^t \dot{h}(s)V(x_s)ds \right], \end{aligned} \quad (3)$$

where $\beta = (k_B T)^{-1}$ with T the temperature and k_B the Boltzmann constant. The thermodynamic role of the entropy flux $S_\varepsilon(\omega)$ becomes clear by the explicit form given in the second line: It contains no system-specific information, and can be written down without specifying the system under consideration.

This time-antisymmetric part of $A_\varepsilon(\omega)$ is complemented by the time-symmetric part $D_\varepsilon(\omega) = \frac{1}{2}[\tilde{A}_\varepsilon(\Theta\omega) + A_\varepsilon(\omega)]$ [20,21,25–28]. In contrast to $S_\varepsilon(\omega)$, $D_\varepsilon(\omega)$ has no thermodynamic interpretation. A general form such as the lower line of Eq. (3) is not known, but has been given for specific systems [20,21,26]. To proceed, we assume that $A_\varepsilon(\omega)$ and $\tilde{A}_\varepsilon(\Theta\omega)$ can be expanded around $\varepsilon = 0$, i.e., $D_\varepsilon(\omega) = \varepsilon D'(\omega) + \varepsilon^2 D''(\omega)/2 + \dots$ and $S_\varepsilon(\omega) = \varepsilon S'(\omega)$, compared to Eq. (3) [21]. It has proven useful to define the expectation value of $O(x_t)$ under time-reversed dynamics [21,23]

$$\langle O(x_t) \Theta \rangle^\varepsilon = \int O(x_t) \tilde{P}^\varepsilon(\Theta\omega) \mathcal{D}\omega = \langle O(x_t) \rangle = \langle O(x) \rangle, \quad (4)$$

where $\langle O(x) \rangle = \int O(x) P(\omega) \mathcal{D}\omega$ is the equilibrium expectation value of the state observable. The equality in Eq. (4) can be derived via time-reversal relations [21,23]. Subtracting Eq. (4) from Eq. (2) and expanding in powers of ε gives

$$\begin{aligned} \langle O(x_t) \rangle^\varepsilon &= \langle O(x) \rangle + \sum_{n=0}^{\infty} \frac{\varepsilon^n}{n!} \left\langle \frac{d^n}{d\varepsilon^n} \left(e^{-D_\varepsilon(\omega) + \frac{\varepsilon}{2} S'(\omega)} \right. \right. \\ &\quad \left. \left. - e^{-D_\varepsilon(\omega) - \frac{\varepsilon}{2} S'(\omega)} \right) \right|_{\varepsilon=0} O(x_t) \right\rangle. \end{aligned} \quad (5)$$

Executing the derivatives and expanding Eq. (5) in ε results in the known expression of the responses via equilibrium correlations [20,21]. In linear order of ε , $D_\varepsilon(\omega)$ drops out, yielding the FDT. As mentioned, already the second order term involves $D_\varepsilon(\omega)$, as do higher orders, implying that nonlinear responses are not of thermodynamic nature; while this statement is of principal interest, it makes determination of the nonlinear responses challenging and laborious, particularly for complex systems [20,21,23,24,29]. To make progress, we use an identity based on the Leibniz rule [30],

$$\begin{aligned} \frac{d^n}{d\varepsilon^n} e^{-D_\varepsilon(\omega) \pm \frac{\varepsilon}{2} S'(\omega)} \Big|_{\varepsilon=0} \\ = \sum_{i=0}^n \binom{n}{i} \frac{d^{n-i}}{d\varepsilon^{n-i}} e^{-D_\varepsilon(\omega)} \Big|_{\varepsilon=0} \left(\pm \frac{S'(\omega)}{2} \right)^i \end{aligned} \quad (6)$$

with the binomial coefficient $\binom{n}{i}$. Plugging the identity of Eq. (6) into Eq. (5), substituting $e^{-D_\varepsilon(\omega)} = e^{-A_\varepsilon(\omega) - \frac{\varepsilon}{2} S'(\omega)}$ and using the identity once more [with $D_\varepsilon(\omega)$ replaced by $A_\varepsilon(\omega)$]

yields

$$\begin{aligned} \langle O(x_t) \rangle^\varepsilon &= \langle O(x) \rangle + 2 \sum_{n=1}^{\infty} \frac{1}{n!} \varepsilon^n \sum_{i=1, \text{ odd}}^n \binom{n}{i} \sum_{j=0}^{n-i} \binom{n-i}{j} \\ &\quad \times (-1)^j \frac{d^{n-i-j}}{d\varepsilon^{n-i-j}} \left\langle \left(\frac{S'(\omega)}{2} \right)^{i+j} O(x_t) \right\rangle^\varepsilon \Big|_{\varepsilon=0}. \end{aligned} \quad (7)$$

As required, the sum contains no term $n = 0$.

Equation (7), hereafter referred to as *thermodynamic response relation* (TRR), is the main result of this paper. It relates the mean of the observable far from equilibrium on the left hand side to correlation functions involving entropy on the right hand side. Its thermodynamic meaning is thus displayed with Eq. (3). It is insightful to regard the first orders in ε from Eq. (7) explicitly, mirroring the responses χ_n of order n ($\langle O(x_t) \rangle^\varepsilon = \langle O(x) \rangle + \chi_1 \varepsilon + \chi_2 \varepsilon^2 \dots$),

$$\begin{aligned} \chi_1^{\text{TRR}}(t) &= \langle S'(\omega) O(x_t) \rangle, \\ \chi_2^{\text{TRR}}(t) &= \frac{d}{d\varepsilon} \langle S'(\omega) O(x_t) \rangle^\varepsilon \Big|_{\varepsilon=0} - \frac{1}{2} \langle S'(\omega)^2 O(x_t) \rangle, \\ \chi_3^{\text{TRR}}(t) &= \frac{1}{2} \frac{d^2}{d\varepsilon^2} \langle S'(\omega) O(x_t) \rangle^\varepsilon \Big|_{\varepsilon=0} \\ &\quad - \frac{1}{2} \frac{d}{d\varepsilon} \langle S'(\omega)^2 O(x_t) \rangle^\varepsilon \Big|_{\varepsilon=0} + \frac{1}{6} \langle S'(\omega)^3 O(x_t) \rangle. \end{aligned} \quad (8)$$

The linear response, i.e., the first line in Eq. (8), resembles FDT. Higher order responses contain higher order correlations of $S'(\omega)$, and also derivatives of these correlations with respect to ε , so that the right hand side of Eq. (7) requires measurements under applied perturbation. Notably, the n th order response is related to correlation functions up to order $n - 1$. We thus interpret the TRR in Eq. (7) as follows: Even far from equilibrium, thermodynamics allows to predict one order in perturbation strength—this new insight contains FDT as a special case, connecting equilibrium to linear order.

III. DISCUSSION IN A COARSE-GRAINED FOUR-STATE MODEL

We illustrate and examine the TRR of Eq. (7) in a simple model, where four states A, B, C, D are connected via dimensionless transition rates as depicted in Fig. 1. The system's dynamics follows a simple master equation. For illustration

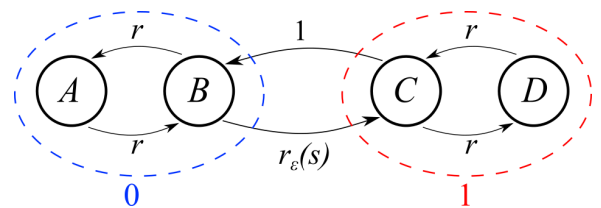


FIG. 1. Sketch of the four-state model. The rates r within the macrostates 0 and 1 are chosen to be small ($r = 0.1$), so that the resulting two state model is strongly non-Markovian [22]. The transition rate $r_\varepsilon(s)$ equals unity for $s < 0$ and follows a perturbation protocol for $s \geq 0$.

purposes, we pretend to be blind to microscopic details by coarse graining: Let states A and B form macrostate $X = 0$, and C and D macrostate $X = 1$ [22]. The resulting two state system is Markovian for large values of r and non-Markovian if r is small. We choose $r = 0.1$ to achieve the latter. This model thus allows to discuss the case of hidden degrees of freedom [31]. As it can also be solved analytically, it makes a good test case for our purposes [22,23].

Treating the so obtained setup via response theory is challenging, due to the presence of the mentioned hidden degrees of freedom: Eq. (5) is not applicable, as evaluation of $D_\varepsilon(\omega)$ requires microscopic resolution [26]. This example allows to illustrate the thermodynamic type of Eq. (7): It is applicable despite presence of hidden degrees of freedom.

The system is in equilibrium for $s < 0$ and perturbed for $s \geq 0$ by making the transition rate which connects macrostate 0 to macrostate 1 a function of the dimensionless time s via the protocol $h(s)$, i.e., $r_\varepsilon(s) = e^{\varepsilon h(s)}$. This perturbation corresponds to Eq. (1) with a dimensionless potential $V(X_s)$ with $V(0) = 0$ and $V(1) = 1$ [22,23]. The entropy production $S_\varepsilon(\omega)$ in Eq. (3) depends on X_s , so that Eq. (7) acts in the space of macrostates, and its thermodynamic nature is apparent. Once on that level, the ‘internal’ nature of the macrostates is not relevant concerning validity of Eq. (7), as required from a thermodynamic relation.

We start with a time independent perturbation (TIP), i.e., $h(s) = 1, s \geq 0$, so that Eq. (3) simplifies to $S'(\omega) = V(X_t) - V(X_0)$ ($\beta = 1$ here and in the following). Furthermore, we choose $O(X_t) = X_t$. We evaluate the responses up to third order, using computer simulations of the master equation, and applying Eq. (8). The derivatives with respect to ε appearing in Eq. (8) are calculated via central differencing, i.e., $\frac{d}{d\varepsilon} \langle \dots \rangle |_{\varepsilon=0} = \lim_{\varepsilon \rightarrow 0} \frac{\langle \dots \rangle^\varepsilon - \langle \dots \rangle^{-\varepsilon}}{2\varepsilon}$ for first order, $\frac{d^2}{d\varepsilon^2} \langle \dots \rangle |_{\varepsilon=0} = \lim_{\varepsilon \rightarrow 0} \frac{\langle \dots \rangle^\varepsilon - 2\langle \dots \rangle + \langle \dots \rangle^{-\varepsilon}}{\varepsilon^2}$ for second order and so on.¹ This requires a choice of ε . The results for χ_2^{TRR} and χ_3^{TRR} are shown in Fig. 2 as red data points using $\varepsilon = 0.1$ and a simulation time step of $\Delta t = 0.001$. In that curve, we also show the exact results χ_2^a, χ_3^a , which can be found analytically.² χ_2 raises to a maximum at around $t \approx 0.5$, and then goes to zero for $t \rightarrow \infty$, for reasons of symmetry. This final decay is slow due to the internal rate r being small. χ_3 exhibits a similar behavior, but levels off to a finite static response at $t \rightarrow \infty$. Figure 2 demonstrates the validity of Eq. (7) for the given model, and that model also allows to test practical aspects, such as the convergence of Eq. (7) in the statistical sense. For χ_2 and χ_3 , we provide two panels each with different number N of independent computer ‘measurements’. While the main graphs supply a qualitative impression, insets of the left panels give the *averaged relative deviation*, i.e., the time average of the ratio $|\langle \chi_n(t) - \chi_n^a(t) \rangle| / \chi_n^a(t)$. The panels show the decay of this relative error with $1/\sqrt{N}$, as expected. The insets in the right panels display this relative error for a fixed $N = 10^{10}$, as a function of ε used in the mentioned

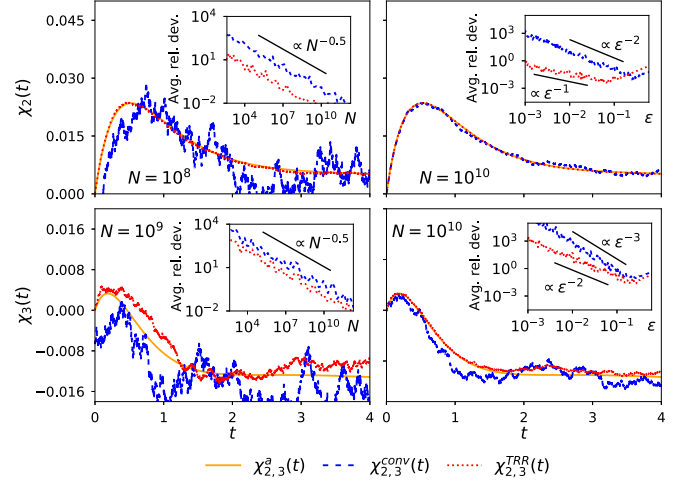


FIG. 2. Second (top) and third (bottom) order responses of the four-state model for the TIP, as functions of time t , found analytically or via simulations using Eq. (7) (TRR), or the conventional method. The number of independent measurements is denoted N . Insets depict the relative statistical error as a function of N (left) for $\varepsilon = 0.1$ and as a function of ε (right) for $N = 10^{10}$.

central differencing. The curves reveal a minimum: For large values of ε , Eq. (7) acquires a systematic error, while, for $\varepsilon \rightarrow 0$ and a fixed N , the statistical error diverges.

The statistical quality of results obtained via Eq. (7) can be compared to the outcome of *conventional response* measurements, for which the responses χ_n^{conv} of n th order are obtained from perturbed data as [22,29]

$$\chi_n^{\text{conv}}(t) = \frac{1}{n!} \frac{d^n}{d\varepsilon^n} \langle X_t \rangle^\varepsilon \Big|_{\varepsilon=0}. \quad (9)$$

One difference between evaluating Eqs. (7) and (9) is already evident from the method of central differencing: The higher the order n , the more different experimental or simulation setups have to be used (e.g., two, i.e., $+\varepsilon$ and $-\varepsilon$ for χ_1^{conv}). The TRR, Eq. (7) thus, for odd orders, requires a smaller number of different setups [one versus two for χ_1 (FDT), two versus two for χ_2 , three versus four for χ_3].

The results of the conventional method are displayed in Fig. 2 as blue data points. When comparing the two formalisms, we note similar behavior of the relative error (insets), but the scaling for $\varepsilon \rightarrow 0$ is different, with power laws that are advantageous for the TRR. The smaller the value of ε , i.e., the smaller a systematic error is sought, the better is the TRR in comparison to the conventional method.

We continue by investigation of a time dependent perturbation (TDP) of the form $h(s) = \sin(\omega_0 s)$, where $\omega_0 = \pi$ is chosen. Analogously to the above, second and third order responses are calculated, analytically², via the TRR in Eq. (8) and the conventional method in Eq. (9), here using $\Delta t = 0.005$ and $\varepsilon = 0.2$ for simulations. Figure 3 presents the resulting curves, again demonstrating the validity of Eq. (7). Furthermore, as for TIP, we show the relative error as a function of N for the two methods. Both scale as $1/\sqrt{N}$ as before, with the error of TRR being again smaller for the given value of ε . We also show a close up view of the short time

¹The third order is given by $\frac{d^3}{d\varepsilon^3} \langle \dots \rangle |_{\varepsilon=0} = \lim_{\varepsilon \rightarrow 0} \frac{\langle \dots \rangle^{2\varepsilon} - 2\langle \dots \rangle^\varepsilon + 2\langle \dots \rangle^{-\varepsilon} - \langle \dots \rangle^{-2\varepsilon}}{2\varepsilon^3}$.

²See Appendix for more details regarding the analytical expressions in the coarse-grained four-state model.

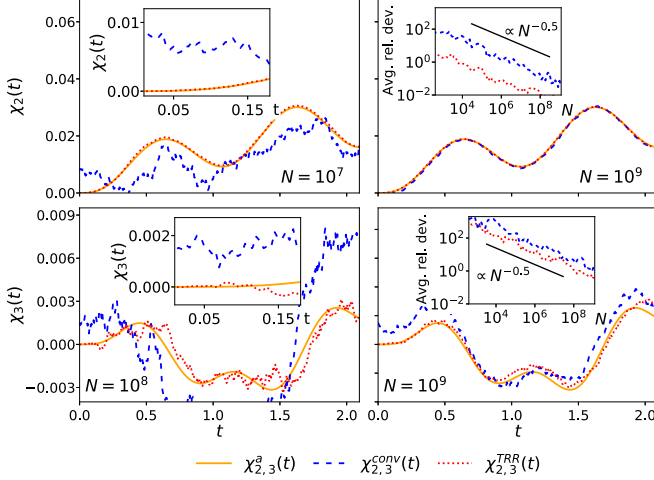


FIG. 3. Second (top) and third (bottom) order responses for a sinusoidal perturbation as functions of time t . Responses are computed analytically, or found by simulations via Eq. (7) (TRR) or via the conventional method. N gives the number of independent measurements. Left insets present a closer view of the short time behavior. Right insets show the relative error as a function of N for $\varepsilon = 0.2$.

behavior, which discloses a fundamental difference. For $t \rightarrow 0$, the responses vanish. While the error of the conventional method stays finite for any t , the relative error diverges for $t \rightarrow 0$. For this reason, we excluded times $t \leq 0.1$ when computing the time averaged relative deviation depicted in the other insets. In contrast, the TRR appears to be much more precise at short times, because $S'(\omega)$ naturally approaches zero leading to vanishing χ_n^{TRR} . While qualitatively advantageous for short times, one may expect the TRR to deviate stronger for very large t (larger than shown here), as the error may add up in the time integrals in $S'(\omega)$. This will be investigated in future work.

IV. SUMMARY

A thermodynamic nonlinear response relation connects responses to time correlation functions at arbitrary order of perturbation strength. With it, thermodynamics allows to predict one order in perturbation strength, a statement, which encloses the FDT at lowest order. Although we restricted the derivation of Eq. (8) to perturbations by a potential, Eq. (7) is also applicable for nonconservative perturbation forces, as long as S_ε is linear in ε , which is a typical case. Analyzing this relation for a coarse-grained four-state model displays similarities but also fundamental differences in the scaling of the statistical error compared to the conventional method. Especially for small value of perturbation strength, the new formula converges faster compared to the conventional method. Future work will address possible extension to perturbations around nonequilibrium steady states.

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APPENDIX: ANALYTICAL CALCULATION OF THE RESPONSES IN THE COARSE-GRAINED FOUR-STATE MODEL

Here, we detail, how the responses of the coarse-grained four-state model to the time independent perturbation and to the sinusoidal perturbation are analytically calculated and give the exact expressions for the second and third order response for these perturbations. The time evolution of the probability distributions of the four states $\rho_i(t)$, $i \in \{A, B, C, D\}$ is given by the master equation involving the generator matrix [22,23]

$$M = \begin{pmatrix} -r & r & 0 & 0 \\ r & -r - r_\varepsilon(s) & r_\varepsilon(s) & 0 \\ 0 & 1 & -r - 1 & r \\ 0 & 0 & r & -r \end{pmatrix}. \quad (\text{A1})$$

The initial condition is that the system is in equilibrium at $t = 0$ which corresponds to the distribution $\rho_i(0) = 1/4$, $i \in \{A, B, C, D\}$. The expectation value of the observable $O(X_t) = X_t$ on the coarse-grained level is computed by $\langle X_t \rangle^\varepsilon = \rho_C(t) + \rho_D(t)$ [22].

The master equation can be explicitly solved for the time independent perturbation. The second and third order responses can be calculated by the derivatives in Eq. (8) in the main text (for brevity, we provide all explicit forms for $r = 0.1$):

$$\chi_2^a(t) = \frac{1}{81608} e^{-1.1t} [\sqrt{101}(100 - 909t) \sinh(\sqrt{1.01}t) + 9191t \cosh(\sqrt{1.01}t)] \quad (\text{A2})$$

and

$$\begin{aligned} \chi_3^a(t) = & \frac{1}{197817792} e^{-(\sqrt{1.01}+1.1)t} (-303t[(1819\sqrt{101} \\ & + 18281)t - 4(507\sqrt{101} + 5093)] - 4121204 \\ & \times e^{(\sqrt{1.01}+1.1)t} + e^{2\sqrt{1.01}t} \{303t[(1819\sqrt{101} \\ & - 18281)t - 2028\sqrt{101} + 20372] + 21302\sqrt{101} \\ & + 2060602\} - 21302\sqrt{101} + 2060602). \end{aligned} \quad (\text{A3})$$

For the sinusoidal perturbation, the master equation cannot be explicitly solved. Therefore, the responses are determined by expanding the formal solution, which is a time-ordered exponential, in terms of ε resulting in a Dyson series [23]. When executing the obtained integrals, the following expressions for the second and third order are obtained:

$$\begin{aligned} \chi_2^a(t) = & -[1616(101 + 25\pi^2)(1 + 111\pi^2 + 25\pi^4)(1 + 444\pi^2 + 400\pi^4)]^{-1} e^{-(3\sqrt{1.01}+1.1)t} (-101\pi(101 + 25\pi^2) \\ & \times e^{(3\sqrt{1.01}+1.1)t} [(1 + 178\pi^2 + 8500\pi^4 - 5000\pi^6) \sin(2\pi t) - 3\pi(6 + 675\pi^2 + 5000\pi^4) \cos(2\pi t) + \pi(6 + 25\pi^2) \end{aligned}$$

$$\begin{aligned} & \times (1 + 444\pi^2 + 400\pi^4)] + 2\sqrt{101}e^{3\sqrt{1.01}t} \sinh(\sqrt{1.01}t)\{(1 + 444\pi^2 + 400\pi^4)[275\pi(1 + 5\pi^2) \sin(\pi t) + 2(202 \\ & + 23975\pi^2 + 6250\pi^4) \cos(\pi t)] + 2(1 + 111\pi^2 + 25\pi^4)(-202 - 94194\pi^2 - 52625\pi^4 + 2500\pi^6)\} + 202e^{3\sqrt{1.01}t} \\ & \times \cosh(\sqrt{1.01}t)\{2(1 + 111\pi^2 + 25\pi^4)(23 + 9756\pi^2 + 8625\pi^4 + 2500\pi^6) - (1 + 444\pi^2 + 400\pi^4)[25\pi(5\pi^2 - 1) \\ & \times \sin(\pi t) + 2(23 + 2400\pi^2 + 625\pi^4) \cos(\pi t)]\} \end{aligned} \quad (\text{A4})$$

and

$$\begin{aligned} \chi_3^a(t) = & [1958592\pi(101 + 25\pi^2)^2(101 + 100\pi^2)(1 + 111\pi^2 + 25\pi^4)^2(1 + 444\pi^2 + 400\pi^4)(1 + 999\pi^2 + 2025\pi^4)]^{-1} \\ & \times e^{-(3\sqrt{1.01}+2.2)t}(-303e^{(3\sqrt{1.01}+1.1)t}(1 + 444\pi^2 + 400\pi^4) \cosh(\sqrt{1.01}t)\{-101[25\pi^8(625\pi^4\{50\pi^2[75\pi^2 \\ & \times (26407 + 2400\pi^2) + 12370777] + 2900081621\} + 4621856501725\pi^2 + 4476537956287) + 33729449681951\pi^6 \\ & + 619396582987\pi^4 + 3215953221\pi^2 + 2631858] + 2\pi^2(101 + 25\pi^2)(101 + 100\pi^2)(1 + 111\pi^2 + 25\pi^4) \\ & \times (1 + 999\pi^2 + 2025\pi^4)(4366 + 484550\pi^2 + 119375\pi^4)t - 101(101 + 25\pi^2)(1 + 111\pi^2 + 25\pi^4)(1 + 999\pi^2 \\ & + 2025\pi^4)\{[25\pi^2(38929 + 48940\pi^2 + 9750\pi^4) + 8686] \cos(2\pi t) + \pi[25\pi^2(21576 + 25575\pi^2 + 5000\pi^4) \\ & + 4816] \sin(2\pi t)\}) + 101(101 + 100\pi^2)[909e^{(3\sqrt{1.01}+2.2)t}\pi^2(1 + 111\pi^2 + 25\pi^4)[25\pi^6(-185551 - 497900\pi^2 \\ & + 60000\pi^4) - 150379\pi^4 - 1209\pi^2 - 2] \cos(3\pi t)(101 + 25\pi^2)^2 - 101e^{(3\sqrt{1.01}+2.2)t}\pi(1 + 111\pi^2 + 25\pi^4)[50\pi^6 \\ & \times (-453291 - 761675\pi^2 + 1395000\pi^4) - 301283\pi^4 - 1011\pi^2 - 1] \sin(3\pi t)(101 + 25\pi^2)^2 - 3(1 + 999\pi^2 \\ & + 2025\pi^4)\{-101e^{(3\sqrt{1.01}+2.2)t}(6 + 3403\pi^2 + 320165\pi^4 + 1919125\pi^6 + 3312500\pi^8)(101\pi + 25\pi^3)^2 \\ & - 2e^{(4\sqrt{1.01}+1.1)t}(1 + 111\pi^2 + 25\pi^4)^2[125000\pi^8(9\sqrt{101} - 101) + 202(432\sqrt{101} - 4343) + 2500\pi^6(4718\sqrt{101} \\ & - 48177) + 707\pi^2(54819\sqrt{101} - 550981) + 100\pi^4(381602\sqrt{101} - 3840323)] + 2e^{(2\sqrt{1.01}+1.1)t}(1 + 111\pi^2 \\ & + 25\pi^4)^2[125000\pi^8(9\sqrt{101} + 101) + 202(432\sqrt{101} + 4343) + 2500\pi^6(4718\sqrt{101} + 48177) + 707\pi^2 \\ & \times (54819\sqrt{101} + 550981) + 100\pi^4(381602\sqrt{101} + 3840323)]\} \cos(\pi t) - 3(\pi + 999\pi^3 + 2025\pi^5) \\ & \times (-101e^{(3\sqrt{1.01}+2.2)t}\{50\pi^6[625\pi^2(-107 + 24\pi^2) - 51011] - 77699\pi^4 - 607\pi^2 - 1\}(101 + 25\pi^2)^2 \\ & - 50e^{(4\sqrt{1.01}+1.1)t}(1 + 111\pi^2 + 25\pi^4)^2[101(\sqrt{101} - 9) + 10000\pi^6\sqrt{101} + 50\pi^4(739\sqrt{101} - 4949) + 18\pi^2 \\ & \times (2399\sqrt{101} - 23331)] + 50e^{(2\sqrt{1.01}+1.1)t}(1 + 111\pi^2 + 25\pi^4)^2[10000\pi^6\sqrt{101} + 101(\sqrt{101} + 9) + 50\pi^4 \\ & \times (739\sqrt{101} + 4949) + 18\pi^2(2399\sqrt{101} + 23331)]\} \sin(\pi t)] + 3e^{(3\sqrt{1.01}+1.1)t}(1 + 444\pi^2 + 400\pi^4)\{\pi^2 \\ & \times [-25\pi^6(625\pi^4\{50\pi^2[25\pi^2(84528979 + 7435800\pi^2) + 13601411543] + 3173994538649) \\ & + 4973852941113775\pi^2 + 4751067447679093] - 35480872475621649\pi^4 - 639468614663713\pi^2 + 202(101 \\ & + 25\pi^2)(101 + 100\pi^2)(1 + 111\pi^2 + 25\pi^4)(1 + 999\pi^2 + 2025\pi^4)(434 + 48200\pi^2 + 11875\pi^4)t \\ & - 3263596534879] - 101(101 + 25\pi^2)(1 + 111\pi^2 + 25\pi^4)(1 + 999\pi^2 + 2025\pi^4)\{[25\pi^2(391061 + 488540\pi^2 \\ & + 97250\pi^4) + 87264] \cos(2\pi t) + \pi[25\pi^2(218854 + 256375\pi^2 + 50000\pi^4) + 48884] \\ & \times \sin(2\pi t)\} - 2670540192\}\sqrt{101} \sinh(\sqrt{1.01}t)). \end{aligned} \quad (\text{A5})$$

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