

# System–bath entanglement theorem with Gaussian environments

Cite as: J. Chem. Phys. **152**, 034102 (2020); <https://doi.org/10.1063/1.5134745>

Submitted: 01 November 2019 . Accepted: 05 January 2020 . Published Online: 16 January 2020

 Peng-Li Du,  Yao Wang,  Rui-Xue Xu,  Hou-Dao Zhang, and  YiJing Yan



View Online



Export Citation



CrossMark

## ARTICLES YOU MAY BE INTERESTED IN

[Entangled system-and-environment dynamics: Phase-space dissipation theory](#)

The Journal of Chemical Physics **152**, 041102 (2020); <https://doi.org/10.1063/1.5135776>

[Instanton formulation of Fermi's golden rule in the Marcus inverted regime](#)

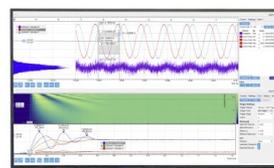
The Journal of Chemical Physics **152**, 034106 (2020); <https://doi.org/10.1063/1.5137823>

[Special topic on dynamics of open quantum systems](#)

The Journal of Chemical Physics **152**, 020401 (2020); <https://doi.org/10.1063/1.5142731>

Challenge us.

What are your needs for  
periodic signal detection?



Zurich  
Instruments



# System–bath entanglement theorem with Gaussian environments

Cite as: J. Chem. Phys. 152, 034102 (2020); doi: 10.1063/1.5134745

Submitted: 1 November 2019 • Accepted: 5 January 2020 •

Published Online: 16 January 2020 • Publisher error corrected 21 January 2020



View Online



Export Citation



CrossMark

Peng-Li Du, Yao Wang, Rui-Xue Xu,<sup>a)</sup> Hou-Dao Zhang, and YiJing Yan

## AFFILIATIONS

Hefei National Laboratory for Physical Sciences at the Microscale and Department of Chemical Physics and Synergetic Innovation Center of Quantum Information and Quantum Physics and Collaborative Innovation Center of Chemistry for Energy Materials (iChEM), University of Science and Technology of China, Hefei, Anhui 230026, China

<sup>a)</sup> Author to whom correspondence should be addressed: rxxu@ustc.edu.cn

## ABSTRACT

In this work, we establish a so-called “*system–bath entanglement theorem*,” for arbitrary systems coupled with Gaussian environments. This theorem connects the entangled system–bath response functions in the total composite space to those of local systems, as long as the interacting bath spectral densities are given. We validate the theorem with direct evaluation via the exact dissipaton-equation-of-motion approach. Therefore, this work enables various quantum dissipation theories, which originally describe only the reduced system dynamics, for their evaluations on the system–bath entanglement properties. Numerical demonstrations are carried out on the Fano interference spectroscopies of spin–boson systems.

Published under license by AIP Publishing. <https://doi.org/10.1063/1.5134745>

## I. INTRODUCTION

System–bath entanglement plays a crucial role in dynamic and thermal properties of complex systems. However, most quantum dissipation theories (QDTs) focus explicitly only on reduced system density operators. This compromises the capabilities of conventional QDTs in the evaluation of the Fano resonances<sup>1–5</sup> and the correlated dynamics between chromophores and surface plasmons.<sup>6–8</sup> System–bath entanglements also involve in thermodynamics functions and thermal transports.

Almost all existing QDTs are based on Gaussian bath statistics. Exact methods include the Feynman–Vernon influence functional path integral formalism,<sup>9</sup> and its derivative equivalence, the hierarchical-equations-of-motion (HEOM) implementation.<sup>10–13</sup> Approximate methods often refer to quantum master equations.<sup>14–24</sup> These include the Redfield theory and its modifications,<sup>23</sup> polaron-transformed versions,<sup>24</sup> and self-consistent Born approximation improvements.<sup>25–29</sup> The simplicity of the Gaussian environment is rooted at the underlying Gauss–Wick’s thermodynamics theorem.<sup>30–32</sup> The influence of the environment can be completely described within the linear response theory framework in the isolated bare-bath subspace. This feature has been exploited in various QDTs.

In this work, we address a missing ingredient, the so-called “*system–bath entanglement theorem*,” for an arbitrary system coupled with the Gaussian environment. As usual, the total system-plus-bath composite Hamiltonian reads

$$H_T = H_S + h_B + H_{SB} \equiv H_S + h_B + \sum_a \hat{Q}_a \hat{F}_a. \quad (1)$$

The system Hamiltonian  $H_S$  and dissipative modes  $\{\hat{Q}_a\}$  are arbitrary. In the above equation, we denote the bath Hamiltonian in the lower case for the Gaussian environment scenario. This requires not only  $h_B$  be harmonic but also the hybrid bath modes  $\{\hat{F}_a\}$  be linear. That is,

$$h_B = \frac{1}{2} \sum_j \omega_j (\hat{p}_j^2 + \hat{x}_j^2) \quad \text{and} \quad \hat{F}_a = \sum_j c_{aj} \hat{x}_j. \quad (2)$$

These microscopic expressions, with dimensionless coordinates  $\{\hat{x}_j\}$  and momentums  $\{\hat{p}_j\}$ , will be used explicitly later in Sec. II A. Throughout the paper, we set  $\hbar = 1$  and  $\beta = 1/(k_B T)$ , with  $k_B$  as the Boltzmann constant and  $T$  the temperature.

It is worth noting that an open quantum system is subject to dephasing, energy relaxation, and transport. These irreducible

processes are beyond the total composite Hamiltonian description. Additional information, such as temperature  $T$  and the interacting bath statistics, would be needed.<sup>30–32</sup> In fact, the total composite Hamiltonian,  $H_T$  of Eq. (1), constitutes a “closed system” in the thermodynamics nomenclature, which is in thermal contact with surroundings at a given temperature  $T$ . The resultant thermal equilibrium is given by the total system-and-bath composite density operator,  $\rho_T^{\text{eq}}(T) = e^{-\beta H_T}/Z_T$ , with  $Z_T \equiv \text{Tr} e^{-\beta H_T}$  being the thermodynamics partition functions. One typical example is a total composite solution system in chemistry, where  $H_S$  and  $h_B$  stand for the solute particle and solvent environment, respectively, and  $H_{SB}$  for the coupling between them. Apparently, physically relevant and directly measurable quantities such as correlation and response functions are all concerned with the total system-plus-bath composite space.

The simplicity of the Gaussian environment, Eq. (2), is that its influence on the reduced system can be completely described with the linear response theory in the bare-bath subspace.<sup>30–32</sup> The fundamental quantities here are the interacting bath response functions,

$$\phi_{ab}(t - \tau) \equiv i\langle[\hat{F}_a^{\text{B}}(t), \hat{F}_b^{\text{B}}(\tau)]\rangle_{\text{B}}, \quad (3)$$

where  $\hat{F}_a^{\text{B}}(t) \equiv e^{ih_B t} \hat{F}_a^{\text{B}} e^{-ih_B t}$  and  $\langle(\cdot)\rangle_{\text{B}} \equiv \text{tr}_{\text{B}}[(\cdot)\rho_{\text{B}}^0(T)]$  with  $\rho_{\text{B}}^0(T) \equiv e^{-\beta h_B}/\text{tr}_{\text{B}}(e^{-\beta h_B})$ . These are just uncorrelated bare-bath quantities, as if  $H_{SB} = 0$ . The interacting bath spectral density functions are<sup>30–32</sup>

$$J_{ab}(\omega) = \frac{1}{2i} \int_{-\infty}^{\infty} dt e^{i\omega t} \phi_{ab}(t), \quad (4)$$

which are often given through models in various QDTs.<sup>11,30–35</sup> However, most of these theories focus only on the reduced system dynamics and evaluate expectation values and correlation/response functions of various system operators  $\{\hat{O}_S\}$ .

As mentioned earlier, the total composite Hamiltonian  $H_T$  of Eq. (1) constitutes a closed system in thermodynamics. It is important to include the system–bath entanglement dynamics arising from  $H_{SB} = \sum_a \hat{Q}_a \hat{F}_a$ , the last term in Eq. (1), into explicit consideration. In fact,  $H_{SB}$  is related to the system–bath hybridization free-energy change.<sup>36</sup> It would require the response/correlation functions between system operators  $\{\hat{Q}_a\}$  and the hybrid bath modes  $\{\hat{F}_a\}$  in the total system-and-bath composite space.

The system–bath entanglement theorem to be established in this work relates the  $i\langle[\hat{Q}_a(t), \hat{F}_b(0)]\rangle$  and  $i\langle[\hat{F}_a(t), \hat{F}_b(0)]\rangle$  types of response functions to those of local system  $i\langle[\hat{Q}_a(t), \hat{Q}_b(0)]\rangle$ . Here,  $\hat{O}(t) \equiv e^{iH_T t} \hat{O} e^{-iH_T t}$  and  $\langle(\cdot)\rangle \equiv \text{Tr}[(\cdot)\rho_T^{\text{eq}}(T)]$  are defined in the total system-plus-bath composite space. Apparently,  $i\langle[\hat{F}_a(t), \hat{F}_b(0)]\rangle \neq \phi_{ab}(t)$  of Eq. (3). The latter is the fictitious uncorrelated bare-bath subspace property, which will serve as the bridge to the aforementioned relations. The conventional QDTs, such as the HEOM formalism,<sup>10–13</sup> are capable of evaluating the local system properties. This work would naturally enable their evaluations on those entanglement response/correlation functions between the local system and the non-local environment. These are all the ingredients in Fano interference spectroscopies.<sup>1–3,37,38</sup>

It is worth noting that we will establish the system–bath entanglement theorem in the non-equilibrium steady-state scenario.

Therefore, this work would be closely related to plasmon spectroscopies dressed with strong plasmonic fields.<sup>6–8</sup> Other methods such as the nonequilibrium Green’s function technique would also be enabled for the aforementioned system–bath entanglement properties. Moreover, one may exploit the system–bath entanglement theorem to bridge between all-atom simulations and implicit Gaussian solvent environment models.

This paper is organized as follows: We establish the system–bath entanglement theorem in Sec. II and numerically demonstrate it in Sec. III. Validations are carried out with respect to direct evaluation via the exact dissipaton-equation-of-motion (DEOM) approach.<sup>39,40</sup> Fano interference spectroscopies are evaluated on spin–boson systems. We conclude this work in Sec. IV.

## II. SYSTEM-BATH ENTANGLEMENT THEOREM

### A. Langevin equation for solvation dynamics

Consider the quantum Langevin equation for the hybrid bath dynamics, as implied in the total system-and-bath composite Hamiltonian,  $H_T$ , of Eq. (1) with Eq. (2). Let  $\hat{O}(t) \equiv e^{iH_T t} \hat{O} e^{-iH_T t}$ . We obtain

$$\ddot{\hat{x}}_j(t) = -\omega_j^2 \hat{x}_j(t) - \omega_j \sum_a c_{aj} \hat{Q}_a(t). \quad (5)$$

Its formal solution is

$$\begin{aligned} \hat{x}_j(t) = & \hat{x}_j(0) \cos(\omega_j t) + \hat{p}_j(0) \sin(\omega_j t) \\ & - \sum_a c_{aj} \int_0^t d\tau \sin[\omega_j(t - \tau)] \hat{Q}_a(\tau). \end{aligned} \quad (6)$$

It, together with the second identity of Eq. (2), leads to

$$\hat{F}_a(t) = \hat{F}_a^{\text{B}}(t) - \sum_b \int_0^t d\tau \phi_{ab}(t - \tau) \hat{Q}_b(\tau), \quad (7)$$

with the bare-bath random force operator,  $\hat{F}_a^{\text{B}}(t) \equiv e^{ih_B t} \hat{F}_a^{\text{B}} e^{-ih_B t}$ , that also involves in  $\phi_{ab}(t)$  of Eq. (3), the expression,

$$\hat{F}_a^{\text{B}}(t) = \sum_j c_{aj} [\hat{x}_j(0) \cos(\omega_j t) + \hat{p}_j(0) \sin(\omega_j t)]. \quad (8)$$

It is easy to obtain

$$i[\hat{F}_a^{\text{B}}(t), \hat{F}_b(0)] = i[\hat{F}_a^{\text{B}}(t), \hat{F}_b^{\text{B}}(0)] = \phi_{ab}(t). \quad (9)$$

This commutator itself is a  $c$ -number and equals to the bare-bath response function, Eq. (3).

Equation (7) describes the Langevin dynamics for the hybridizing bath modes. It differs from traditional Langevin equations which focus on reduced systems. However this serves as the starting point to the following establishment of system–bath entanglement theorem. Let  $\chi_{AB}(t - \tau) \equiv i\langle[\hat{A}(t), \hat{B}(\tau)]\rangle$  be the response function in the total composite space. As inferred from Eq. (8),  $[\hat{F}_a^{\text{B}}(t), \hat{O}_S] = 0$  for an arbitrary system operator  $\hat{O}_S$ . Consequently, Eq. (7) results in

$$\langle[\hat{F}_a(t), \hat{O}_S(0)]\rangle = - \sum_b \int_0^t d\tau \phi_{ab}(t - \tau) \langle[\hat{Q}_b(\tau), \hat{O}_S(0)]\rangle. \quad (10)$$

This expresses the non-local response as the convolution between the bare-bath and the local-system properties.

## B. System-bath entanglement theorem

In relation to the system-and-bath entanglement dynamics underlying the hybridization system and bath modes,  $\{\hat{Q}_a\}$  and  $\{\hat{F}_a\}$ , we denote the following response functions in the total composite space,

$$\begin{aligned}\chi_{ab}^{SS}(t) &\equiv i\langle[\hat{Q}_a(t), \hat{Q}_b(0)]\rangle, \\ \chi_{ab}^{SB}(t) &\equiv i\langle[\hat{Q}_a(t), \hat{F}_b(0)]\rangle, \\ \chi_{ab}^{BS}(t) &\equiv i\langle[\hat{F}_a(t), \hat{Q}_b(0)]\rangle, \\ \chi_{ab}^{BB}(t) &\equiv i\langle[\hat{F}_a(t), \hat{F}_b(0)]\rangle.\end{aligned}\quad (11)$$

The involving operators all arise from  $H_{SB} = \sum_a \hat{Q}_a \hat{F}_a$ , with specifying  $\{\hat{Q}_a\}$  and  $\{\hat{F}_a\}$  being the operators in the system and bath subspaces, respectively. Equation (7) gives rise to

$$\chi_{ab}^{BS}(t) = -\sum_{b'} \int_0^t d\tau \phi_{ab'}(t-\tau) \chi_{b'b}^{SS}(\tau). \quad (12)$$

By  $\chi_{ba}^{SB}(t) = -\chi_{ab}^{BS}(-t)$ , it leads to further

$$\chi_{ba}^{SB}(t) = -\sum_{b'} \int_0^t d\tau \phi_{b'a}(t-\tau) \chi_{bb'}^{SS}(\tau), \quad (13)$$

obtained via changing the integral variable with  $\tau' = -\tau$ , followed by using the antisymmetric relation for both response functions in the integrand. Moreover, Eq. (7) also gives rise to

$$\chi_{ab}^{BB}(t) = \phi_{ab}(t) - \sum_{b'} \int_0^t d\tau \phi_{ab'}(t-\tau) \chi_{b'b}^{SB}(\tau). \quad (14)$$

In terms of the susceptibility or frequency resolution,  $\tilde{f}(\omega) = \int_0^\infty dt e^{i\omega t} f(t)$ , Eqs. (12)–(14) read

$$\begin{aligned}\tilde{\chi}_{ab}^{BS}(\omega) &= -\sum_{b'} \tilde{\phi}_{ab'}(\omega) \tilde{\chi}_{b'b}^{SS}(\omega), \\ \tilde{\chi}_{ba}^{SB}(\omega) &= -\sum_{b'} \tilde{\chi}_{bb'}^{SS}(\omega) \tilde{\phi}_{b'a}(\omega),\end{aligned}\quad (15)$$

and

$$\tilde{\chi}_{ab}^{BB}(\omega) = \tilde{\phi}_{ab}(\omega) + \sum_{a'b'} \tilde{\phi}_{aa'}(\omega) \tilde{\chi}_{a'b'}^{SS}(\omega) \tilde{\phi}_{b'b}(\omega). \quad (16)$$

Let  $\tilde{\chi}^{BS}(\omega) \equiv \{\tilde{\chi}_{ab}^{BS}(\omega)\}$  be a matrix, and similar for others, so that one can recast Eqs. (15) and (16) as

$$\tilde{\chi}^{BS}(\omega) = -\tilde{\phi}(\omega) \tilde{\chi}^{SS}(\omega), \quad \tilde{\chi}^{SB}(\omega) = -\tilde{\chi}^{SS}(\omega) \tilde{\phi}(\omega) \quad (17)$$

and

$$\tilde{\chi}^{BB}(\omega) = \tilde{\phi}(\omega) + \tilde{\phi}(\omega) \tilde{\chi}^{SS}(\omega) \tilde{\phi}(\omega). \quad (18)$$

We refer these identities as the system-bath entanglement theorem that goes with the Gaussian bath model. They relate the nonlocal

properties,  $\tilde{\chi}^{BS}(\omega)$ ,  $\tilde{\chi}^{SB}(\omega)$ , and  $\tilde{\chi}^{BB}(\omega)$ , with the local system  $\tilde{\chi}^{SS}(\omega)$  and the bare bath  $\tilde{\phi}(\omega)$ . Define the overall system-bath entanglement susceptibility,

$$\begin{aligned}\chi_{SB}(\omega) &\equiv \sum_a \tilde{\chi}_{aa}^{SB}(\omega) = \text{tr} \tilde{\chi}^{SB}(\omega), \\ \chi_{BS}(\omega) &\equiv \sum_a \tilde{\chi}_{aa}^{BS}(\omega) = \text{tr} \tilde{\chi}^{BS}(\omega).\end{aligned}\quad (19)$$

From Eq. (17), we immediately have

$$\chi_{SB}(\omega) = \chi_{BS}(\omega). \quad (20)$$

This describes the reciprocal relation of the overall system-bath entanglement susceptibility.

It is worth emphasizing that the ensemble averages underlying all response functions in this work are concerned with the steady-state scenario. In other words, the established theorem, from Eqs. (10) to (20), is for arbitrary systems coupled with Gaussian steady-state environments.

It is also noticed that in general, the frequency resolution can be expressed as  $\tilde{\chi}_{AB}(\omega) = \tilde{\chi}_{AB}^{(+)}(\omega) + i\tilde{\chi}_{AB}^{(-)}(\omega)$ , with  $\tilde{\chi}_{AB}^{(\pm)}(\omega) = [\tilde{\chi}_{BA}^{(\pm)}(\omega)]^*$  being the Hermite/anti-Hermite matrix component, respectively. Here,  $\hat{A}$  and  $\hat{B}$  are both Hermitian operators. The anti-Hermite component,  $\tilde{\chi}_{AB}^{(-)}(\omega)$ , also refers to the spectral density. In the thermal equilibrium scenario, it is related to the correlation function via the fluctuation-dissipation theorem.<sup>30–32</sup>

## III. NUMERICAL DEMONSTRATIONS

### A. Fano profile in a spin-boson model

For numerical demonstrations, we focus on a single-dissipative mode spin-boson model and evaluate the Fano interference spectroscopy. We will see that the system-bath entanglement theorem-based indirect evaluations perfectly agree with the results of direct approach.<sup>37</sup>

The total system-and-bath composite model Hamiltonian, in the presence of an external field  $E(t)$ , assumes

$$H_T(t) = \frac{\Omega}{2} \hat{\sigma}_z + h_B + \hat{\sigma}_x \hat{F} - \hat{\mu}_T E(t), \quad (21)$$

where

$$\hat{\mu}_T = \mu_S \hat{\sigma}_x + \nu_B \hat{F}. \quad (22)$$

The first term,  $\hat{\mu}_S = \mu_S \hat{\sigma}_x$ , represents the transition dipole of the two-level system (or solute molecule), which itself has no permanent dipole. The second term in Eq. (22) describes the external light field  $E(t)$ -induced bath (solvent) environment polarization. Physically, this would correspond to the scenario where an individual solvent molecule has low polarity but with random orientations. Thus, the bulk of the solvent is isotropic around the nonpolar solute molecule. The external light field, assumed to be linear polarized, breaks the original isotropic symmetry and induces the solvent polarization. On the other hand, due to the form of system-bath coupling in Eq. (21), the transition dipole of the solute system could also induce the polarized solvation coordinate  $\hat{F}$ .

Nevertheless, for the main purpose of the present work, we set the bath polarization, the second term in Eq. (22), in the form of  $\hat{\mu}_B = v_B \hat{F}$ . The total composite dipole susceptibility is then given by

$$\begin{aligned}\Xi(\omega) &\equiv i \int_0^\infty dt e^{i\omega t} \langle [\hat{\mu}_T(t), \hat{\mu}_T(0)] \rangle \\ &= \mu_S^2 \chi_{SS}(\omega) + 2\mu_S v_B \chi_{SB}(\omega) + v_B^2 \chi_{BB}(\omega),\end{aligned}\quad (23)$$

with [cf. Eqs. (11) and (20)]

$$\begin{aligned}\chi_{SS}(\omega) &\equiv i \int_0^\infty dt e^{i\omega t} \langle [\hat{\sigma}_x(t), \hat{\sigma}_x(0)] \rangle, \\ \chi_{SB}(\omega) &\equiv i \int_0^\infty dt e^{i\omega t} \langle [\hat{\sigma}_x(t), \hat{F}(0)] \rangle, \\ \chi_{BB}(\omega) &\equiv i \int_0^\infty dt e^{i\omega t} \langle [\hat{F}(t), \hat{F}(0)] \rangle.\end{aligned}\quad (24)$$

The system–bath entanglement theorem, Eqs. (17) and (18), lead to Eq. (23) and further the expression,

$$\Xi(\omega) = v_B^2 \tilde{\phi}(\omega) + [\mu_S - v_B \tilde{\phi}(\omega)]^2 \chi_{SS}(\omega).\quad (25)$$

To proceed, we denote

$$\begin{aligned}\phi_r(\omega) &\equiv \text{Re} \tilde{\phi}(\omega), \quad \phi_i(\omega) \equiv \text{Im} \tilde{\phi}(\omega), \\ q(\omega) &\equiv \mu_S - v_B \phi_r(\omega).\end{aligned}\quad (26)$$

Further,

$$z(\omega) \equiv \frac{\chi_{SS}(\omega)}{|\chi_{SS}(\omega)|^2} \equiv z_r(\omega) + iz_i(\omega).\quad (27)$$

Here,  $z_r(\omega) \equiv \text{Re} z(\omega)$  and  $z_i(\omega) \equiv \text{Im} z(\omega)$ . Some simple algebra on Eq. (25) leads to the expressions,

$$\begin{aligned}\frac{\text{Re} \Xi(\omega)}{|\chi_{SS}(\omega)|^2} &= \frac{v_B^2 \phi_r(\omega)}{|\chi_{SS}(\omega)|^2} + 2v_B \phi_i(\omega) q(\omega) z_i(\omega) \\ &\quad + [q^2(\omega) - v_B^2 \phi_i^2(\omega)] z_r(\omega)\end{aligned}\quad (28)$$

and

$$\begin{aligned}\frac{\text{Im} \Xi(\omega)}{|\chi_{SS}(\omega)|^2} &= [z_i(\omega) - \phi_i(\omega)] [q^2(\omega) + v_B^2 \phi_i(\omega) z_i(\omega)] \\ &\quad + \phi_i(\omega) [q(\omega) - v_B z_r(\omega)]^2.\end{aligned}\quad (29)$$

In fact,  $z(\omega)$  of Eq. (27) is related to the self-energy  $\Sigma(\omega)$  in

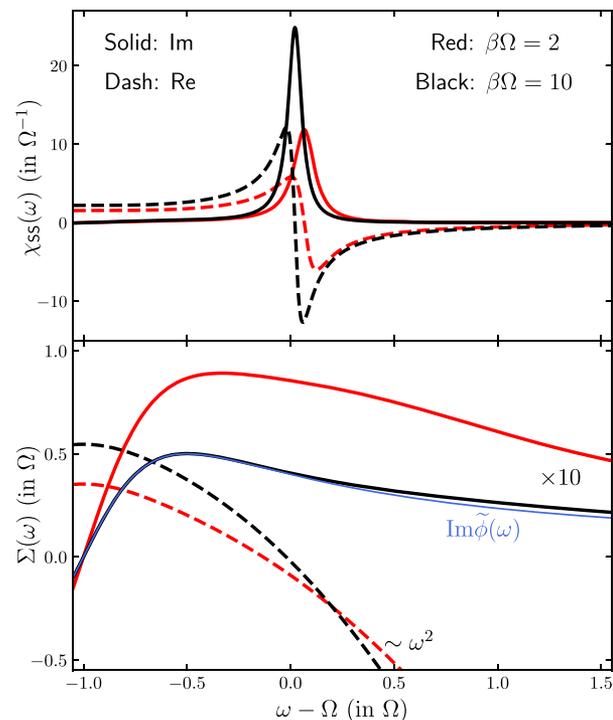
$$\chi_{SS}(\omega) = \frac{\Omega}{\Omega^2 - \omega^2 - \Omega \Sigma(\omega)},\quad (30)$$

via

$$\begin{aligned}\text{Re} \Sigma(\omega) &\equiv [\Omega^2 - \omega^2 - \Omega z_r(\omega)] / \Omega, \\ \text{Im} \Sigma(\omega) &\equiv z_i(\omega).\end{aligned}\quad (31)$$

Note that the boson–boson model goes with  $\Sigma(\omega) = \tilde{\phi}(\omega)$ .<sup>30–32</sup> However, for the spin–boson model, the self-energy needs to be evaluated via  $\chi_{SS}(\omega)$  from certain QDT methods.

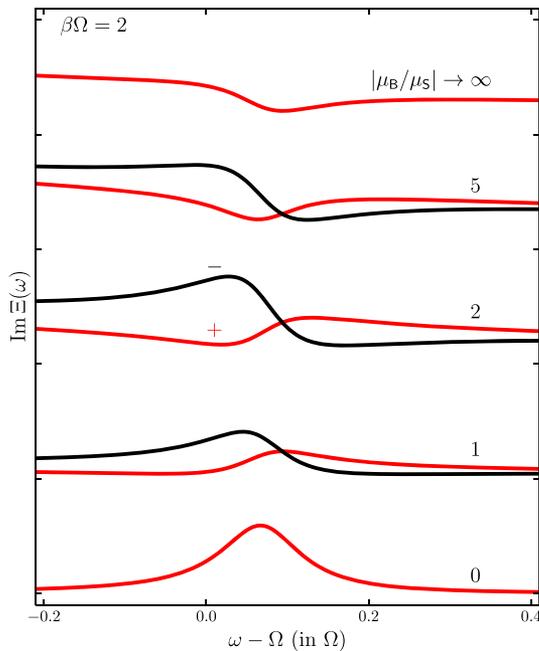
Figure 1 depicts  $\chi_{SS}(\omega)$  (upper panel) and the associated self-energy  $\Sigma(\omega)$  (lower panel), at a low temperature ( $\beta\Omega = 10$ ; black)



**FIG. 1.** Real (dash) and imaginary (solid) parts of  $\chi_{SS}(\omega)$  (upper panel) and  $\Sigma(\omega)$  (lower panel), at a low temperature ( $\beta\Omega = 10$ ; black) and a high temperature ( $\beta\Omega = 2$ ; red). Also depicted is  $\text{Im} \tilde{\phi}(\omega)$  (thin-solid) for comparison. The three solid curves in the lower panel are timed with 10. The asymptotic behavior of  $\text{Re} \Sigma(\omega)$  to infinity is found to be quadratic. Drude spectral bath model  $\tilde{\phi}(\omega) = 2\lambda\gamma/(\gamma - i\omega)$  is adopted with the parameters chosen as  $\lambda = 0.05 \Omega$  and  $\gamma = 0.5 \Omega$ .

and a high temperature ( $\beta\Omega = 2$ ; red). Here,  $\Sigma(\omega)$  is obtained via Eq. (30) from  $\chi_{SS}(\omega)$ . The latter is evaluated via the exact DEOM approach,<sup>39,40</sup> which is equivalent to the HEOM method,<sup>10–13</sup> in the absence of bath polarization. Physically,  $\text{Im} \chi_{SS}(\omega)$  and  $\text{Re} \chi_{SS}(\omega)$  are related to the reduced system linear spectrum and dispersion, respectively, in the absence of bath polarization ( $v_B = 0$ ). As anticipated, the peak is relatively sharp and strong in the low-temperature regime. In the lower panel, the asymptotic behavior of  $\text{Re} \Sigma(\omega)$  is found to be quadratic in the large  $\omega$  regime. This is also the behavior of  $z_r(\omega)$ , cf. Eq. (31). As mentioned earlier, the boson–boson model goes with  $\Sigma(\omega) = \tilde{\phi}(\omega)$ . We do observe that  $\text{Im} \Sigma(\omega) \approx \phi_i(\omega)$ , as anticipated for the low temperature case studied here; cf. the black vs thin-blue curves in the lower panel.

Figure 2 exhibits the Fano interference spectral lineshape,  $\text{Im} \Xi(\omega)$ , with different values of relative bath dipole strength  $\mu_B/\mu_S$ , where  $\mu_B \equiv 2\lambda v_B$ . It is noticed that the dipole ratio can be either positive or negative. The plus and minus signs in the figure represent the two special cases where the bath dipole is parallel and anti-parallel to the system one, respectively. As mentioned above, the differences between  $\text{Im} \Sigma(\omega)$  and  $\phi_i(\omega)$  become smaller as the temperature decreases. This would lead to more similarities of Fano interference patterns between the spin–boson and boson–boson cases,<sup>37,38</sup> cf. Eq. (29). All results of  $\Xi(\omega)$  here are evaluated via Eq. (25) from  $\chi_{SS}(\omega)$



**FIG. 2.** Response spectra,  $\text{Im } \Xi(\omega)$  [cf. Eq. (25)], for the high temperature case of Fig. 1. The bath dipole  $\mu_B \equiv 2\lambda_{\nu_B}$  is chosen as  $\mu_B/\mu_S = \infty, \pm 5, \pm 2, \pm 1, 0$ . The red and black curves correspond to the + and - signs, respectively.

and  $\tilde{\phi}(\omega) \equiv \phi_r(\omega) + i\phi_i(\omega)$ , which have been confirmed to be consistent with those from the direct DEOM evaluations on Eq. (23).<sup>37,38</sup> Thus, the system–bath entanglement theorem [Eqs. (17) and (18)] is also numerically verified.

#### IV. SUMMARY

In this work, we propose the system–bath entanglement theorem, Eqs. (17) and (18), for arbitrary systems coupled with Gaussian environments. This theorem expresses the entangled system–bath response functions in the total composite space with those of local system, as long as the interacting bath response functions  $\{\phi_{ab}(t)\}$  of Eq. (3) are given. This is the general case as the quantum dissipation formulations are concerned with. The captioned theorem is established on basis of the convolution relation, Eq. (10), between the bare-bath and the local-system responses for non-local system–bath properties, obtained by revisiting the Langevin dynamics for the hybridizing bath modes, Eq. (7). This theorem enables various quantum dissipation theories, which originally only deal with the reduced system dynamics, to evaluate system–bath entanglement properties.

To “visualize” the theorem, we evaluate the Fano interference spectra of spin–boson systems via both the direct DEOM approach on Eq. (23) and the indirect entanglement-theorem approach on Eq. (25). We obtain full consistency between the results from these two approaches. The Fano analysis made here, Eqs. (22)–(31), could be readily extended to more complex systems. Note that the system–bath entanglement theorem here is established in the non-equilibrium steady-state scenario. Therefore, it is anticipated to be closely related to plasmon spectroscopies dressed with strong

plasmonic fields. Moreover, other methods such as the non-equilibrium Green’s function technique would also be readily exploited for system–bath entanglement properties.

#### AUTHOR’S CONTRIBUTIONS

P.-L. Du and Y. Wang contributed equally to this work.

#### ACKNOWLEDGMENTS

Support from the Ministry of Science and Technology of China (Grant Nos. 2017YFA0204904 and 2016YFA0400904) and the Natural Science Foundation of China (Grant Nos. 21633006 and 21703225) is gratefully acknowledged.

#### REFERENCES

- U. Fano, *Phys. Rev.* **124**, 1866 (1961).
- A. E. Miroshnichenko, S. Flach, and Y. S. Kivshar, *Rev. Mod. Phys.* **82**, 2257 (2010).
- M. F. Limonov, M. V. Rybin, A. N. Poddubny, and Y. S. Kivshar, *Nat. Photonics* **11**, 543 (2017).
- Y. Zhang, T.-T. Tang, C. Girit, Z. Hao, M. C. Martin, A. Zettl, M. F. Crommie, Y. R. Shen, and F. Wang, *Nature* **459**, 820 (2009).
- T.-T. Tang, Y. Zhang, C.-H. Park, B. Geng, C. Girit, Z. Hao, M. C. Martin, A. Zettl, M. F. Crommie, S. G. Louie, Y. R. Shen, and F. Wang, *Nat. Nanotechnol.* **5**, 32 (2010).
- B. Luk’yanchuk, N. I. Zheludev, S. A. Maier, N. J. Halas, P. Nordlander, H. Giessen, and C. T. Chong, *Nat. Mater.* **9**, 707 (2010).
- R. R. Frontiera, N. L. Gruenke, and R. P. Van Duyne, *Nano Lett.* **12**, 5989 (2012).
- D. Rodrigo, O. Limaj, D. Janner, D. Etezadi, F. J. García de Abajo, V. Pruneri, and H. Altug, *Science* **349**, 165 (2015).
- R. P. Feynman and F. L. Vernon, Jr., *Ann. Phys.* **24**, 118 (1963).
- Y. Tanimura, *Phys. Rev. A* **41**, 6676 (1990).
- Y. Tanimura, *J. Phys. Soc. Jpn.* **75**, 082001 (2006).
- Y. A. Yan, F. Yang, Y. Liu, and J. S. Shao, *Chem. Phys. Lett.* **395**, 216 (2004).
- R. X. Xu, P. Cui, X. Q. Li, Y. Mo, and Y. J. Yan, *J. Chem. Phys.* **122**, 041103 (2005).
- A. G. Redfield, *Adv. Magn. Reson.* **1**, 1 (1965).
- B. J. Berne, in *Physical Chemistry: An Advanced Treatise*, edited by H. Eyring, D. Henderson, and W. Jost (Academic, New York, 1971), Vol. 8B, pp. 539–716.
- F. Haake, in *Quantum Statistics in Optics and Solid State Physics*, Springer Tracts in Modern Physics Vol. 66, edited by G. Höhler (Springer, Berlin, 1973), pp. 98–168.
- G. Lindblad, *Rep. Math. Phys.* **10**, 393 (1976).
- A. O. Caldeira and A. J. Leggett, *Physica A* **121**, 587 (1983).
- N. G. van Kampen, *Stochastic Processes in Physics and Chemistry* (North-Holland, Amsterdam, 1992).
- Y. J. Yan, *Phys. Rev. A* **58**, 2721 (1998).
- Y. J. Yan, F. Shuang, R. X. Xu, J. X. Cheng, X. Q. Li, C. Yang, and H. Y. Zhang, *J. Chem. Phys.* **113**, 2068 (2000).
- R. X. Xu and Y. J. Yan, *J. Chem. Phys.* **116**, 9196 (2002).
- W. M. Zhang, T. Meier, V. Chernyak, and S. Mukamel, *J. Chem. Phys.* **108**, 7763 (1998).
- S. Jang, Y.-C. Cheng, D. R. Reichman, and J. D. Eaves, *J. Chem. Phys.* **129**, 101104 (2008).
- B. B. Laird, J. Budimir, and J. L. Skinner, *J. Chem. Phys.* **94**, 4391 (1991).
- S. Jang, J. S. Cao, and R. J. Silbey, *J. Chem. Phys.* **116**, 2705 (2002).
- M. Esposito and M. Galperin, *Phys. Rev. B* **79**, 205303 (2009).
- M. G. Mavros and T. Van Voorhis, *J. Chem. Phys.* **141**, 054112 (2014).
- J. S. Jin, J. Li, Y. Liu, X.-Q. Li, and Y. J. Yan, *J. Chem. Phys.* **140**, 244111 (2014).
- U. Weiss, *Quantum Dissipative Systems*, Series in Modern Condensed Matter Physics Vol. 13, 3rd ed. (World Scientific, Singapore, 2008).

- <sup>31</sup>H. Kleinert, *Path Integrals in Quantum Mechanics, Statistics, Polymer Physics, and Financial Markets*, 5th ed. (World Scientific, Singapore, 2009).
- <sup>32</sup>Y. J. Yan and R. X. Xu, *Annu. Rev. Phys. Chem.* **56**, 187 (2005).
- <sup>33</sup>A. Leggett, S. Chakravarty, A. Dorsey, M. Fisher, A. Garg, and W. Zwerger, *Rev. Mod. Phys.* **59**, 1 (1987).
- <sup>34</sup>C. Meier and D. J. Tannor, *J. Chem. Phys.* **111**, 3365 (1999).
- <sup>35</sup>M. Thoss, H. B. Wang, and W. H. Miller, *J. Chem. Phys.* **115**, 2991 (2001).
- <sup>36</sup>J. G. Kirkwood, *J. Chem. Phys.* **3**, 300 (1935).
- <sup>37</sup>H. D. Zhang, R. X. Xu, X. Zheng, and Y. J. Yan, *J. Chem. Phys.* **142**, 024112 (2015).
- <sup>38</sup>R. X. Xu, H. D. Zhang, X. Zheng, and Y. J. Yan, *Sci. China Chem.* **58**, 1816 (2015), Special Issue: Lemin Li Festschrift.
- <sup>39</sup>Y. J. Yan, *J. Chem. Phys.* **140**, 054105 (2014).
- <sup>40</sup>Y. J. Yan, J. S. Jin, R. X. Xu, and X. Zheng, *Front. Phys.* **11**, 110306 (2016).