System–bath entanglement theorem with Gaussian environment

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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 system only. We validate the theorem with
the 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 demonstra-
tions are carried out on the Fano interference spectroscopies of spin–boson systems.

I. INTRODUCTION

System–bath entanglement plays a crucial role in dy-
namic and thermal properties of complex systems. How-
ever, most quantum dissipation theories (QDTs) focus
explicitly only on reduced system density operators. This
compromises the capabilities of conventional QDTs in
evaluation on such as the Fano resonances1–5 and the
correlated dynamics between chromophores and surface
plasmons.6–8 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,9
and its derivative equivalence, the hierarchical–
equations–of–motion (HEOM) implementation.10–14
Approximate methods often refer to quantum master
equations.15–25 These include the Redfield theory and
its modifications,24 polaron–transformed versions,25 and
self-consistent Born approximation improvements.26–30
The simplicity of Gaussian environment is rooted at the
underlying Gauss–Wick’s thermodynamics theorem.31–33
The influence of environment can be completely de-
scribed 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 arbi-
trary system coupled with Gaussian environment. As
usual the total system–plus–bath composite Hamiltonian
reads

$$\mathcal{H}_T = \mathcal{H}_S + \sum_a \dot{Q}_a \dot{F}_a + \mathcal{H}_B. \quad (1)$$

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

$$h_B = \frac{1}{2} \sum_j \omega_j (p_j^2 + x_j^2) \quad \text{and} \quad \dot{F}_a = \sum_j c_{aj} x_j. \quad (2)$$

Throughout the paper we set $\hbar = 1$ and $\beta = 1/(k_B T)$,
with $k_B$ the Boltzmann constant and $T$ the temperature.

The influence of Gaussian environment is com-
pletely characterized with

$$\phi_{ab}(t - \tau) \equiv \langle [\dot{F}_a(t), \dot{F}_b(\tau)] \rangle_n. \quad (3)$$

This represents the linear response functions of hybrid
bath modes in isolated bare–bath subspace, where $\dot{F}_a(t) \equiv e^{i\hbar \mathcal{H}_B t} F_a e^{-i\hbar \mathcal{H}_B t}$ and $\langle (\cdot) \rangle_n \equiv \text{tr}_n[(\cdot) \rho_n(T)]$ with
$\rho_n(T) \equiv e^{-\beta \mathcal{H}_B}/\text{tr}_n(e^{-\beta \mathcal{H}_B})$.

On the other hand, the system–bath entanglements
would require the response/correlation functions between
system operators \{\(\dot{O}_a\)\} and the hybrid bath modes \{\(\dot{F}_a\)\}
in the total system–and–bath composite space. The
system–bath entanglement theorem to be established re-
lates this type of response/correlation functions between
local system and non-local environment to local ones
only. The bare–bath response functions, \{\(\phi_{ab}(t)\)\} of
Eq. (3), serve as the bridge in this connection. Therefore
this work would enable the conventional QDTs for
their evaluations on the aforementioned entanglement re-
sponse/correlation functions. Moreover, those between
hybrid bath modes, $\langle [\dot{F}_a(t), \dot{F}_b(\tau)] \rangle$ in the total system–
and–bath composite space, can also be evaluated in terms
of the local system response functions. These are all the
ingredients in Fano interference spectroscopies.1–3,4,34,35

It is worth noting that the system–bath entanglement
theorem will be established in non-equilibrium steady-
state scenario. Therefore it is anticipated that this
work would be closely related to plasmon spectroscopies
dressed with strong plasmonic fields.6–8 Moreover, other
methods such as non-equilibrium Green’s function tech-
nique would also be readily exploited for system–bath
entanglement properties.

This paper is organized as follows. We establish the
system–bath entanglement theorem in Sec. II and nu-
merically demonstrate it in Sec. III. Validations are car-
ried out with respect to direct evaluation via the exact
dissipaton–equation–of–motion (DEOM) approach.36,37
Fano interference spectroscopies are evaluated on spin–
boson systems. We conclude this work in Sec. IV.
II. SYSTEM–ENVIRONMENT ENTANGLEMENT THEOREM

A. Langevin equation for solvation dynamics

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

$$\hat{x}_j(t) = -\omega_j^2 x_j(t) - \omega_j \sum_a c_{aj} \hat{Q}_a(t). \quad (4)$$

Its formal solution is

$$x_j(t) = x_j(0) \cos(\omega_j t) + p_j(0) \sin(\omega_j t)$$

$$- \sum_a c_{aj} \int_0^t d\tau \sin[\omega_j(t - \tau)] \hat{Q}_a(\tau). \quad (5)$$

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

$$\hat{F}_a(t) = \hat{F}_a^b(t) = - \sum_b \int_0^t d\tau \phi_{ab}(t - \tau) \hat{Q}_b(\tau), \quad (6)$$

with the bare–bath random force,

$$\hat{F}_a^b(t) = \sum_j c_{aj} [x_j(0) \cos(\omega_j t) + p_j(0) \sin(\omega_j t)]. \quad (7)$$

It is easy to obtain

$$i[\hat{F}_a(t), \hat{Q}_b(0)] = i[\hat{F}_a^b(t), \hat{Q}_b(0)] = \phi_{ab}(t). \quad (8)$$

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

Equation (6) 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. (7), $[\hat{F}_a^b(t), \hat{O}_s] = 0$ for an arbitrary system operator $\hat{O}_s$. Consequently, Eq. (6) results in

$$\langle [\hat{F}_a(t), \hat{O}_s(0)] \rangle = - \sum_b \int_0^t d\tau \phi_{ab}(t - \tau) \langle [\hat{Q}_b(\tau), \hat{O}_s(0)] \rangle. \quad (9)$$

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

B. System–environment entanglement theorem

In relation to the system–and–bath entanglement dynamics underlying the hybridization system and bath modes, $\{\hat{Q}_a\}$ and $\{\hat{F}_a\}$, denote in the following

$$\chi_{ab}^S(t) \equiv \chi_{Q_a Q_b}(t), \quad \chi_{ab}^B(t) \equiv \chi_{F_a F_b}(t), \quad \chi_{ab}^{SB}(t) \equiv \chi_{Q_a F_b}(t), \quad \chi_{ab}^{BS}(t) \equiv \chi_{F_a Q_b}(t), \quad (10)$$

for highlighting the system–and–bath entanglement nature. Equation (6) gives rise to

$$\chi_{ab}^{BS}(t) = - \sum_{b'} \int_0^t d\tau \phi_{ab'}(t - \tau) \chi_{b'b}^{SB}(\tau). \quad (11)$$

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

$$\chi_{ab}^{BB}(t) = \sum_{b'} \int_0^t d\tau \phi_{b'a}(t - \tau) \chi_{b'b}^{BB}(t), \quad (12)$$

obtained via changing the integral variable with $\tau' = -\tau$, followed by using the antisymmetric relation for both response functions in the integrand. Moreover, Eq. (6) also gives rise to

$$\chi_{ab}^{BB}(t) = \phi_{ab}(t) - \sum_{b'} \int_0^t d\tau \phi_{ab'}(t - \tau) \chi_{b'b}^{BB}(t) \quad (13)$$

In terms of the susceptibility or frequency resolution, $\tilde{f}(\omega) = \int_0^\infty dt e^{i\omega t} f(t)$, Eqs. (11)–(13) read

$$\tilde{\chi}_{ab}^{BS}(\omega) = - \sum_{b'} \tilde{\phi}_{ab'}(\omega) \tilde{\chi}_{b'b}^{SS}(\omega) \quad (14)$$

and

$$\tilde{\chi}_{ab}^{BB}(\omega) = \tilde{\phi}_{ab}(\omega) + \sum_{a'b'} \tilde{\phi}_{aa'}(\omega) \tilde{\chi}_{b'b}^{SS}(\omega) \tilde{\phi}_{b'b}(\omega) \quad (15)$$

Let $\tilde{\chi}_{ab}^{BS}(\omega) \equiv \{\tilde{\chi}_{ab}^{BS}(\omega)\}$ be a matrix, and similar for others, so that one can recast Eqs. (14) and (15) as

$$\tilde{\chi}_B^S(\omega) = - \tilde{\phi}(\omega) \tilde{\chi}_S^S(\omega), \quad \tilde{\chi}_B^B(\omega) = - \tilde{\chi}_S^S(\omega) \tilde{\phi}(\omega), \quad (16)$$

and

$$\tilde{\chi}_B^B(\omega) = \tilde{\phi}(\omega) + \tilde{\phi}(\omega) \tilde{\chi}_S^S(\omega) \tilde{\phi}(\omega). \quad (17)$$

We refer these identities the system–bath entanglement theorem that goes with the Gaussian bath model. They relate the nonlocal properties, $\tilde{\chi}_B^S(\omega)$, $\tilde{\chi}_B^B(\omega)$ and $\tilde{\chi}_B^B(\omega)$, with the local system $\tilde{\chi}_S^S(\omega)$ and the bare bath $\tilde{\phi}(\omega)$. Define the overall system–bath entanglement susceptibility,

$$\chi_{SB}(\omega) \equiv \sum_a \tilde{\chi}_{aa}^{SB}(\omega) = \text{tr} \tilde{\chi}_{SB}^S(\omega), \quad \chi_{BS}(\omega) \equiv \sum_a \tilde{\chi}_{aa}^{BS}(\omega) = \text{tr} \tilde{\chi}_{BS}^B(\omega). \quad (18)$$

From Eq. (16) we have immediately

$$\chi_{SB}(\omega) = \chi_{BS}(\omega). \quad (19)$$

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 steady–state scenario. In other words, the
established theorem, from Eq. (9) to Eq. (19), is for arbitrary systems coupled with Gaussian steady-state environments.

It is also noticed that in general, the frequency resolution can be expressed as \( \chi_{AB}^{\pm}(\omega) = \chi_{AB}^{(-)}(\omega) + i\chi_{AB}^{(+)}(\omega) \) with \( \chi_{AB}^{(\pm)}(\omega) = [\chi_{BA}^{(\pm)}(\omega)]^* \) being Hermite/anti-Hermite matrix component, respectively. Here \( A \) and \( B \) are both Hermitian operators. The anti-Hermite component, \( \chi_{AB}^{(-)}(\omega) \), refers also the spectral density. In the thermal equilibrium scenario, it is related to the correlation function via the fluctuation–dissipation theorem.\(^{31–33}\)

### III. NUMERICAL DEMONSTRATIONS

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

Consider a spin–boson model in the presence of an external field \( E(t) \), with the total composite Hamiltonian, 

\[
H_r(t) = \frac{\Omega}{2} \sigma_x + \sigma_x \hat{F} + h_B - \hat{\mu}_t E(t),
\]

where 

\[
\hat{\mu}_t = \mu_0 \sigma_x + \nu_0 \hat{F}.
\]

The total composite dipole susceptibility is given by 

\[
\chi(\omega) \equiv i \int_0^\infty dt e^{i\omega t} \langle [\hat{\sigma}_x(t), \sigma_x(0)] \rangle = \mu_0^2 \chi_{SS}(\omega) + 2\mu_0 \nu_0 \chi_{SB}(\omega) + \nu_0^2 \chi_{BB}(\omega). 
\]

The involving resolutions are concerned with [cf. Eq. (10)]

\[
\begin{align*}
\chi_{SS}(t) & \equiv i \langle [\hat{\sigma}_x(t), \sigma_x(0)] \rangle, \\
\chi_{SB}(t) & \equiv i \langle [\hat{\sigma}_x(t), \hat{F}(0)] \rangle, \\
\chi_{BB}(t) & \equiv i \langle [\hat{F}(t), \hat{F}(0)] \rangle.
\end{align*}
\]

Exploited is also \( \chi_{BB}(t) \equiv i [\hat{F}(t), \sigma_x(0)] = \chi_{SS}(t) \) via Eq. (19). The system–bath entanglement theorem, Eqs. (16) and (17), leads to Eq. (22) further the expression,

\[
\chi(\omega) = \nu_0^2 \tilde{\phi}(\omega) + [\mu_0 - \nu_0 \tilde{\phi}(\omega)]^2 \tilde{\chi}_{SS}(\omega).
\]

To proceed, we introduce 

\[
\xi(\omega) \equiv \frac{\tilde{\chi}_{SS}(\omega)}{[\tilde{\chi}_{SS}(\omega)]^2} = \xi_r(\omega) + i\xi_i(\omega).
\]

Here, \( \xi_r(\omega) \equiv \text{Re}[\xi(\omega)] \) and \( \xi_i(\omega) \equiv \text{Im}[\xi(\omega)] \). This notation is adopted throughout this paper. Some simple algebra will lead to \( \chi(\omega) \equiv \chi_r(\omega) + i\chi_i(\omega) \) of Eq. (24) the expressions [denoting \( \tilde{\phi}(\omega) \equiv \phi(\omega) + i\phi_i(\omega) \),

\[
\begin{align*}
\chi_r(\omega) & = \frac{\nu_0^2 \tilde{\phi}_i(\omega)}{[\chi_{SS}(\omega)]^2} + 2\nu_0 \tilde{\phi}_i(\omega)q(\omega)\xi_i(\omega) \\
& + [q^2(\omega) - \nu_0^2 \tilde{\phi}_i^2(\omega)]\xi_r(\omega),
\end{align*}
\]

and 

\[
\frac{\chi_i(\omega)}{[\chi_{SS}(\omega)]^2} = \xi_i(\omega) - \tilde{\phi}_i(\omega)[q^2(\omega) + \nu_0^2 \tilde{\phi}_i(\omega)\xi_i(\omega)]
\]

\[
+ \tilde{\phi}_i(\omega)[q(\omega) - \nu_0 \xi_r(\omega)]^2,
\]

where 

\[
q(\omega) \equiv \mu_0 - \nu_0 \tilde{\phi}_r(\omega).
\]

In fact, the \( \xi(\omega) \) of Eq. (25) is related to the self–energy \( \Sigma(\omega) \) in 

\[
\tilde{\chi}_{SS}(\omega) = \frac{\Omega}{\Omega^2 - \omega^2 - \Omega\Sigma(\omega)}.
\]

\[
\Sigma_r(\omega) \equiv \frac{[\Omega^2 - \omega^2 - \Omega\xi_r(\omega)]/\Omega,}{\Sigma_i(\omega) \equiv \xi_i(\omega)}
\]

Note that the boson–boson model goes with \( \Sigma(\omega) = \tilde{\phi}(\omega) \). However, for spin–boson model, the self–energy needs to be evaluated via \( \tilde{\chi}_{SS}(\omega) \) from certain QDT methods.

Figure 1 depicts \( \tilde{\chi}_{SS}(\omega) \) (upper–panel) and the associated self–energy \( \Sigma(\omega) \) (lower–panel), at a low temperature (\( \beta\Omega = 10 \); black) and a high temperature
Fano interference in linear response spectra $\chi_i(\omega) \equiv \text{Im} \chi(\omega)$, cf. Eq. (24). The inverse temperature $\beta \Omega = 2$. Other parameters are same as Fig. 1. The bath dipole $\mu_B \equiv 2 \lambda \nu_0$ 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.

As mentioned earlier, the boson–boson model goes with the absence of bath polarization ($\nu_0 = 0$). As anticipated, the peak is relatively sharp and strong in the low–temperature regime. In the lower panel, the asymptotic behavior of $\Sigma(\omega)$ is found to be quadratic in the large $\omega$ regime. This is also the behavior of $\xi(\omega)$, cf. Eq. (30). As mentioned earlier, the boson–boson model goes with $\Sigma(\omega) = \tilde{\phi}(\omega)$. We do observe that $\Sigma(\omega) \approx \tilde{\phi}(\omega)$, as anticipated for the low temperature case studied here, cf. the black versus thin-blue curves in the lower panel.

Figure 2 exhibits the Fano interference spectral line-shape, $\chi_i(\omega)$, with different values of relative bath dipole strength $\mu_B/\mu_S$, where $\mu_B \equiv 2 \lambda \nu_0$. 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 $\Sigma(\omega)$ [equal to $\xi(\omega)$] and $\tilde{\phi}(\omega)$ become smaller as the temperature decreases. This would lead to more similarities of Fano interference patterns between the spin–boson and boson–boson cases, cf. Eq. (27). All results of $\tilde{\chi}(\omega)$ here are evaluated via Eq. (24) from $\chi_{SS}(\omega)$ and $\tilde{\phi}(\omega)$, which have been confirmed to be consistent with those from the direct DEOM evaluations on Eq. (22). Thus the system–bath entanglement theorem [Eqs. (16) and (17)] is also numerically verified.

IV. SUMMARY

In this work, we propose the system–bath entanglement theorem, Eqs. (16) and (17), 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 only. It is established on basis of the convolution relation, Eq. (9), 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. (6). 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 direct DEOM approach on Eq. (22) and indirect entanglement–theorem approach on Eq. (24). We obtain full consistency between the results from these two approaches. The Fano analysis made here, Eqs. (21)–(30), could be readily extended to more complex systems. Noticed that the system–bath entanglement theorem here is established in 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 non-equilibrium Green’s function technique would also be readily exploited for system–bath entanglement properties.

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