Quantum coherent dynamics in single molecule systems: generalized stochastic Liouville equation

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We propose the generalized stochastic Liouville equation to investigate the coherent dynamics in single molecule systems coupled to environments which exhibit both nonstationary and non-Markovian features. The generalized stochastic Liouville equation contains a generalized memory kernel associated with both the intrinsic system Hamiltonian and the non-Markovian features of the environmental noise, which returns to the well-known framework established by Kubo in the limit case that the environmental noise is stationary and memoryless. The coherence of the quantum system can be derived by means of the generalized stochastic Liouville equation in two separated averages with no need to consider the statistical characteristics of the environmental noise. We express the exact analytical expressions of the coherence of the single molecule systems induced by the nonstationary and non-Markovian RTN, and the analytical results of the system coherence are in well consistent with that derived by means of some other theoretical approaches.

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I. INTRODUCTION

Coherence as an important nature of quantum world has been demonstrated to be a useful resource which plays a crucial role in physical, chemical and biological communities \cite{1, 2, 3, 4, 5, 6, 7}. Investigations on quantum coherent dynamics in single molecule have draw much extensive attention due to our increasing capability to observe and control quantum systems at the single-molecule level. At such a scale, there are constantly revealing many new physical effects and mechanisms where the decoherence caused by the environments strongly influences the dynamical evolution of the single molecule \cite{8, 9, 10, 11, 12, 13, 14}. Environmental effects on quantum systems can be modeled by random telegraph noise (RTN) which plays an important role in some fundamental physical, chemical and biological processes, such as, rate process in chemical reactions \cite{15}, optical trapping in biological cells \cite{16, 17}, stochastic resonance in excitable systems \cite{18, 19}, and frequency modulation in quantum information science \cite{20}. In addition, RTN as an important non-Gaussian noise has also been used to generate the low-frequency $1/f^\alpha$ noise both theoretically and experimentally \cite{21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32}, which are responsible for coherent dynamics in quantum solid-state nanodevices.

For a long time, the coherent dynamics of an open quantum system such as a single molecule coupled to its environments is usually governed by a Lindblad type master equation within the Markov approximation, which assumes that the information flows unidirectionally from the system into the environments \cite{33, 34}. With the development of experimental technique, it has been observed accurately that the dynamical evolution of open quantum systems is closely associated with a backflow of information from the environments into the system. For instance, the processes of electronic energy transfer in photosynthesis and decoherence in quantum bits display strong non-Markovian behavior \cite{35, 36, 37, 38}. In the recent two decades, the non-Markovian effect of the coherent dynamics in open quantum systems has drawn increasing attention in a wide variety of fields due to its key role in the community of physics, chemistry and biology systems \cite{39, 40, 41, 42, 43, 44, 45, 46, 47}. Many excellent theoretical approaches have been proposed to investigate the non-Markovian dynamics of open quantum systems, such as, quantum state diffusion \cite{48, 49, 50}, projection operator \cite{51, 52, 53, 54, 55}, quantum jumps \cite{56, 57}, nonequilibrium Green-function \cite{58, 59} and dynamical maps \cite{60, 61}.

There have been well-established theoretical investigations on the coherent dynamics of open quantum systems coupled to environments which recover equilibrium instantly from the interaction between the system and environments, generally assuming that the environmental noise yields both stationary and Markovian statistical properties \cite{22, 24, 30, 62, 63, 64}. However, there are many situations where the nonequilibrium feature of the environments plays an essential role in the coherent dynamics of an open quantum system. The environment is no longer at thermal equilibrium which corresponds to physically that the excited phonons of the environments are in certain nonstationary states initially. For example, in some transient and ultrafast dynamical processes in physical or biological systems which usually take place on a very short time scale, the initial nonstationary states of the environmental excited phonons induced by the coupling to the quantum system can not have the chance to recover equilibrium instantly \cite{65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76}. On the other hand, a quantum system may not interact with a single environment where the coherent dynamics of the system also depends strongly on the interaction between the sub-environments \cite{77, 78, 79}. From this point of view,

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it is necessary to take the environmental nonequilibrium feature into extensive consideration to study the coherent dynamics of an open quantum system with both nonstationary and non-Markovian statistical properties of the environmental noise.

In this paper, we derive the generalized stochastic Liouville equation to investigate the coherent dynamics of single molecule systems induced by environmental noise exhibiting both nonstationary and non-Markovian features. The generalized stochastic Liouville equation contains a generalized memory kernel associated with both the intrinsic system Hamiltonian and the non-Markovian features of the environmental noise. In the limit case that the environment is in equilibrium and with no memory, the generalized stochastic Liouville equation returns to the well-known framework established by Kubo. In the presence of the nonstationary and non-Markovian RTN, we express the exact analytical results of the coherence of the single molecule systems. The analytical expressions of the system coherence are in good agreement with that obtained by some other theoretical approaches.

II. GENERALIZED STOCHASTIC LIOUVILLE EQUATION

We consider a single molecule system coupled to a fluctuating environment which displays both nonstationary and non-Markovian statistical properties. Based on the spectral diffusion framework initiated by Kubo and Anderson, the uncontrolled environmental degrees of freedom give rise to the stochastic fluctuations in the energy eigenvalues of the system during the dynamical evolution as follows [20, 80, 81]

\[ H(t) = H_0 + \delta H(t) = \hbar \sum_n [\omega_n + \epsilon_n(t)]|n\rangle\langle n|, \]

where \( H_0 \) and \( \delta H(t) \) are the intrinsic system Hamiltonian and its fluctuation term, \(|n\rangle\) is a set of orthonormal eigenstates, \( \omega_n \) denotes the intrinsic frequency of the state \(|n\rangle\) and \( \epsilon_n(t) \) is the environmental noise which is subject to a stochastic process.

The dynamical evolution of the stochastic density matrix is governed by the Liouville equation

\[ \frac{\partial}{\partial t} \rho(t; \delta H(t)) = -\frac{i}{\hbar}[H(t), \rho(t; \delta H(t))], \]

where \( \rho(t; \delta H(t)) \) is adopted to indicate that the system dynamics depends on the stochastic fluctuation term \( \delta H(t) \). The dynamical evolution of the stochastic density matrix elements satisfies the differential equations

\[ \frac{\partial}{\partial t} \rho_{nn}(t; \epsilon_n^m(t)) = 0, \]

\[ \frac{\partial}{\partial t} \rho_{nm}(t; \epsilon_n^m(t)) = -i[\omega_n^m - \omega_m^m] \rho_{nm}(t; \epsilon_n^m(t)), \]

where \( \omega_n^m = \omega_n - \omega_m \) is the intrinsic frequency difference between states \(|n\rangle\) and \(|m\rangle\) and the off-diagonal element \( \rho_{nm}(t; \epsilon_n^m(t)) \) depends on the composite environmental noise \( \epsilon_n^m(t) = \epsilon_n(t) - \epsilon_m(t) \). Due to the fact that \([H_0, \delta H(t)] = 0\), the quantum system undergoes pure decoherence and the off-diagonal elements of the reduced density matrix change with time while its populations are time independent and the diagonal elements of the reduced density matrix are constant. The reduced density matrix can be, by averaging over different realizations of the environmental noise, expressed as

\[ \rho(t) = \langle \rho(t; \delta H(t)) \rangle = \sum_n \rho_{nn}(0)|n\rangle\langle n| + \sum_{n,m} \rho_{nm}(0)e^{-i\omega_n^m t}F_{nm}^m(t)|n\rangle\langle m|, \]

where \( F_{nm}^m(t) = \langle \exp[-i \int_0^t \epsilon_n^m(t')dt'] \rangle \) is the decoherence function describing the loss of superposition between states \(|n\rangle\) and \(|m\rangle\). The coherence of the quantum system can be quantified in terms of the \( l_1 \) norm of coherence as [82]

\[ C_{l_1}(t) = \sum_{n,m \neq m} |\rho_{nm}(t)| = \sum_{n,m \neq m} |\rho_{nm}(t; \epsilon_n^m(t))|. \]

In the following, we mainly focus on the derivation of the off-diagonal elements of the reduced density matrix by taking the average of the environmental noise. We consider that the composite environmental noise \( \epsilon_n^m(t) \) is subject to a stochastic process which exhibits both nonstationary and non-Markovian features. The non-Markovian feature of the noise process is characterized by a generalized master equation for the conditional probability which depends on its previous history

\[ \frac{\partial}{\partial t} P(\epsilon_n^m, t|\epsilon_n^m, t') = \int_0^t K_n^m(t - \tau)\mathcal{M}_n^m P(\epsilon_n^m, \tau|\epsilon_n^m, t')d\tau, \]

where the initial condition is given by \( P(\epsilon_n^m, t'|\epsilon_n^m, t') = \delta(\epsilon_n^m - \epsilon_n^m), K_n^m(t - \tau) \) denotes the memory kernel composite environmental noise \( \epsilon_n^m(t) \) and \( \mathcal{M}_n^m \) is a differential operator only involving derivatives with respect to \( \epsilon_n^m \). The composite environmental noise \( \epsilon_n^m(t) \) is non-Markovian because the Chapman-Kolmogorov equation is no longer valid unless it is memoryless, for instance, the memory kernel is proportional to a \( \delta \) function [83]. The nonstationary feature of the noise process \( \epsilon_n^m(t) \) is characterized by the time dependent single-time probability distribution [83]

\[ P(\epsilon_n^m, t) = \int P(\epsilon_n^m, t|\epsilon_n^m, 0)P(\epsilon_n^m, 0)d\epsilon_n^m, \]

where \( P(\epsilon_n^m, 0) \) is a nonstationary probability which represents that the environmental states are nonstationary initially. In contrast to the usual treatment, the statistical properties of the environmental noise are nonstationary, corresponding physically to impulsively excited phonons of the environment with sharply defined
initial values at \( t = 0 \). Under this assumptions, the environment is not at thermal equilibrium. For the case \( P(\varepsilon_{m0}^n, 0) = \lim_{t \to \infty} P(\varepsilon_{m}^n, t)|\varepsilon_{m0}^n = 0) \), the environmental noise \( \varepsilon_{m}^n(t) \) is stationary and the environment is at thermal equilibrium \([83, 84]\).

To generalize the stochastic Liouville equation to the coherent dynamics in the presence of environments exhibiting both nonstationary and non-Markovian features, we first introduce the bivariate process \( \{\varrho_{nm}, \varepsilon_{m}^n\} \) and define the joint probability \([83, 85]\)

\[
P(\varrho_{nm}, \varepsilon_{m}^n) = P(\varrho_{nm}, t|\varepsilon_{m}^n, t)P(\varepsilon_{m}^n, t) = \langle \delta(\varrho_{nm}(t) - \varrho_{nm})\delta(\varepsilon_{m}^n(t) - \varepsilon_{m}^n) \rangle. \tag{8}
\]

Consequently, the time evolution of the joint probability \( P(\varrho_{nm}, \varepsilon_{m}^n, t) \) describes a flow in \((\varrho_{nm}, \varepsilon_{m}^n)\) space

\[
\frac{\partial}{\partial t} P(\varrho_{nm}, \varepsilon_{m}^n, t) = i(\omega_{m}^n + \varepsilon_{m}^n) \frac{\partial}{\partial \varrho_{nm}} \varrho_{nm} P(\varrho_{nm}, \varepsilon_{m}^n, t) + \int_0^t K_{nm}^{m}(t-t')\mathcal{M}_{\varepsilon_{m}}^n P(\varrho_{nm}, \varepsilon_{m}^n, t') dt', \tag{9}
\]

where the initial condition satisfies \( P(\varrho_{nm}, \varepsilon_{m}^n, 0) = \delta(\varrho_{nm}(0) - \varrho_{nm0})P(\varepsilon_{m}^n, 0) \) and it has been assumed that the environmental noise \( \varepsilon_{m}^n(t) \) is not influenced by the system and that there is no initial correlation between the system and its environment.

The off-diagonal element of the reduced density matrix of the quantum system can be obtained in \((\varrho_{nm}, \varepsilon_{m}^n)\) space by means of two separated averages. We first define the partial average over \( \varrho_{nm} \) for fixed \( \varepsilon_{m}^n \) as

\[
\rho_{nm}(\varepsilon_{m}^n, t) = \int \varrho_{nm} P(\varrho_{nm}, \varepsilon_{m}^n, t) d\varrho_{nm}. \tag{10}
\]

By multiplying Eq. (9) with \( \varrho_{nm} \) and integrating, we obtain the generalized stochastic Liouville equation

\[
\frac{\partial}{\partial t} \rho_{nm}(\varepsilon_{m}^n, t) = -i(\omega_{m}^n + \varepsilon_{m}^n)\rho_{nm}(\varepsilon_{m}^n, t) + \int_0^t K_{nm}^{m}(t-t')\mathcal{M}_{\varepsilon_{m}}^n \rho_{nm}(\varepsilon_{m}^n, t') dt', \tag{11}
\]

where \( K_{nm}^{m}(t-t') = K_{nm}^{m}(t-t')e^{-i\omega_{m}^n(t-t')} \) denotes the generalized memory kernel which is associated with both the intrinsic frequency difference \( \omega_{m}^n \) between states \( |n\rangle \) and \( |m\rangle \) and the memory kernel of the composite environmental noise \( \varepsilon_{m}^n(t) \). Here the generalized stochastic Liouville equation (11) contains both the unitary and nonunitary parts of the evolution arising from the environmental nonstationary and non-Markovian features, respectively. Consequently, the off-diagonal element of the reduced density matrix \( \rho_{nm}(t) \) can be obtained by the complete average

\[
\rho_{nm}(t) = \int \rho_{nm}(\varepsilon_{m}^n, t) d\varepsilon_{m}^n. \tag{12}
\]

It is worth mentioning that when the composite environmental noise is memoryless, i.e. \( K_{nm}^{m}(t-t') = \delta(t-t') \), the generalized stochastic Liouville equation returns to the well-known framework established by Kubo \([86]\). It is convenient to derive the reduced density matrix of the quantum system by means of the generalized stochastic Liouville equation since we just need to know the statistical properties of the environmental noise, e.g., stationary or nonstationary, and Markovian or non-Markovian, instead of its statistical characteristics, such as the correlation function of each order. However, the differential operator \( \mathcal{M}_{\varepsilon_{m}}^n \) describing the statistical properties of the environmental noise in Eq. (6) is of arbitrary form or even nonlinear. Therefore, the reduced density matrix can be analytically solved only for a few special cases but we can obtain its numerical solution by means of the generalized stochastic Liouville equation.

### III. THEORETICAL RESULTS AND DISCUSSION

In this section, we study a special and important case of the coherent dynamics in the presence of the environments exhibiting nonstationary and non-Markovian RTN features. We will derive analytically the off-diagonal element of the reduced density matrix of the quantum system by means of the generalized stochastic Liouville equation. We assume that the composite environmental noise \( \varepsilon_{m}^n(t) \) obeys a nonstationary non-Markovian RTN process associated with an initially nonstationary distribution and an environmental memory kernel, which jumps randomly between the values \( \pm \nu_{m}^n \) with the switching rate \( \lambda_{m}^n \) \([71-73]\). The time evolution of the conditional probability of the composite environmental noise \( \varepsilon_{m}^n(t) \) is governed by the generalized master equation \([87]\)

\[
\frac{\partial}{\partial t} P(\pm \nu_{m}^n, t|\varepsilon_{m}^n(t), t') = \int_{t'}^{t} \lambda_{m} K_{m}^{n}(t-t') \left[ P(-\nu_{m}^n, \tau|\varepsilon_{m}^n(t'), t') - P(\pm \nu_{m}^n, \tau|\varepsilon_{m}^n(t'), t') \right] d\tau,
\]

\[
\frac{\partial}{\partial t} P(-\nu_{m}^n, t|\varepsilon_{m}^n(t), t') = \int_{t'}^{t} \lambda_{m} K_{m}^{n}(t-t') \left[ P(+\nu_{m}^n, \tau|\varepsilon_{m}^n(t'), t') - P(-\nu_{m}^n, \tau|\varepsilon_{m}^n(t'), t') \right] d\tau. \tag{13}
\]

The environment is not at thermal equilibrium, corresponding to the initially nonstationary distribution for environmental noise as

\[
P(\varepsilon_{m0}^n, 0) = \frac{1}{2}(1 - a_{m}^n)\delta_{\varepsilon_{m0}^n, -\nu_{m}^n} + \frac{1}{2}(1 + a_{m}^n)\delta_{\varepsilon_{m0}^n, +\nu_{m}^n}, \tag{14}
\]

where \( \varepsilon_{m0}^n = \pm \nu_{m}^n \) and \( a_{m}^n \) is the nonequilibrium parameter and \(-1 \leq a_{m}^n \leq 1 \). For the case \( a_{m}^n = 0 \), the environmental states are stationary initially corresponding to that the environment is in equilibrium.

Based on Eq. (11), the generalized stochastic Liouville equation for the coherent dynamics induced by nonsta-
tionary and non-Markovian RTN can be expressed as
\[
\frac{\partial}{\partial t} \rho_{nm}(+\nu_m^nt) = -i(\omega_m^nt + \nu_m^nt)\rho_{nm}(+\nu_m^nt, t) + \int_0^t \lambda_m^nt \\
\times K_m^n(t-t')e^{-i\nu_m^nt(-t-t')}[\rho_{nm}(-\nu_m^nt, t') - \rho_{nm}(+\nu_m^nt, t')]dt',
\]
where the initial conditions are given by \(\rho_{nm}(\pm\nu_m^nt, 0) = \frac{1}{2}(1 \pm a_m^nt)\rho_{nm}(0)\). By means of Laplace transform, the off-diagonal element of the reduced density matrix \(\rho_{nm}(t)\) can be written in a sum as
\[
\rho_{nm}(t) = \rho_{nm}(+\nu_m^nt, t) + \rho_{nm}(-\nu_m^nt, t) \\
e^{-i\nu_m^nt}F^n_m(t)\rho_{nm}(0),
\]
where the decoherence function \(F^n_m(t)\) can be analytically solved as
\[
F^n_m(t) = \mathcal{L}^{-1}[\tilde{F}^n_m(p)],
\]
\[
\tilde{F}^n_m(p) = \frac{p + 2\lambda_m^ntK_m^n(p) + ia_m^nt\nu_m^n}{p^2 + 2\lambda_m^ntK_m^n(p) + (\nu_m^n)^2}.
\]
Here \(\mathcal{L}^{-1}\) indicates the inverse Laplace transform, \(K_m^n(p)\) is the Laplace transform of the memory kernel and the initial condition is given by \(F^n_m(0) = 1\). Clearly, it is more convenient to calculate the decoherence function induced by a nonstationary and non-Markovian RTN process based on the expression derived in Eq. (17) rather than to derive a closed equation for the characteristic function based on the statistical characteristics of the environmental noise as we did in Refs. [71, 72].

1. Composite memory kernel

We first consider the case that the composite environmental noise \(\epsilon_m^n(t)\) is with a composite form of memory kernel
\[
K_m^n(t-t') = w_m^n\delta(t-t') + (1 - w_m^n)\kappa_m^ne^{-\kappa_m^nt(t-t')},
\]
where \(0 \leq w_m^n \leq 1\) denotes a weight factor for Markovian and non-Markovian features of the environmental noise and \(\kappa_m^n\) is the memory decay rate of the exponential kernel. In Laplace domain, the composite memory kernel yields \(\tilde{K}^n_m(p) = w_m^n + (1 - w_m^n)\frac{\kappa_m^n}{p + \kappa_m^n}\) and the decoherence function in Eq. (17) can be expressed as
\[
F^n_m(t) = \mathcal{L}^{-1}[\tilde{F}^n_m(p)],
\]
\[
\tilde{F}^n_m(p) = \frac{p + 2\lambda_m^nt[w_m^n + (1 - w_m^n)\frac{\kappa_m^n}{p + \kappa_m^n}] + ia_m^nt\nu_m^n}{p^2 + 2\lambda_m^nt[w_m^n + (1 - w_m^n)\frac{\kappa_m^n}{p + \kappa_m^n}] + (\nu_m^n)^2}.
\]
In the memoryless limit of the environmental noise, namely, the weight \(w_m^n = 1\) or the decay rate of the exponential kernel \(\kappa_m^n \rightarrow +\infty\). In this case, the Laplace domain decoherence function in Eq. (19) can be reduced to
\[
\tilde{F}^n_m(p) = \frac{p + 2\lambda_m^nt + ia_m^nt\nu_m^n}{p^2 + 2\lambda_m^nt + (\nu_m^n)^2}.
\]
By taking the inverse Laplace transform of Eq. (20), we can express the decoherence function in time domain as
\[
F^n_m(t) = e^{-\lambda_m^nt \pm \sqrt{(\lambda_m^n)^2 - (\nu_m^n)^2}}[\cosh(\beta_m^n t) + \frac{\lambda_m^n}{\beta_m^n}\sinh(\beta_m^n t)]
\]
\[
+ ia_m^nt\frac{\nu_m^n}{\beta_m^n}\sin(\beta_m^n t),
\]
where \(\beta_m^n = \sqrt{|(\lambda_m^n)^2 - (\nu_m^n)^2|}\) for \(\nu_m^n < \lambda_m^n, \nu_m^n = \lambda_m^n\) and \(\nu_m^n > \lambda_m^n\), respectively. This expression of the decoherence function in Eq. (21) is consistent with that obtained in Ref. [72] and it returns to the well-known results [88–90] when the environment is in equilibrium, namely, the statistical property of the environmental noise is stationary.

2. Modulatable memory kernel

We now consider the case that the composite environmental noise \(\epsilon_m^n(t)\) is subject to a modulatable non-Markovian process with the memory kernel
\[
K_m^n(t-t') = \kappa_m^ne^{-\kappa_m^nt(t-t')}\cos[\Omega_m^n(t-t')],
\]
where \(\Omega_m^n\) denotes the external modulation frequency of the environment [91, 92]. The modulatable memory kernel becomes an exponential form when there is no environmental frequency modulation \(\Omega_m^n = 0\). The memory kernel in Laplace domain yields \(\tilde{K}^n_m(p) = \frac{\kappa_m^n(p + \kappa_m^n)}{p^2 + (\Omega_m^n)^2}\) and the decoherence function in Eq. (17) can be written as
\[
F^n_m(t) = \mathcal{L}^{-1}[\tilde{F}^n_m(p)],
\]
\[
\tilde{F}^n_m(p) = \frac{p + 2\lambda_m^nt[\frac{\kappa_m^n}{p + \kappa_m^n}] + ia_m^nt\nu_m^n}{p^2 + 2\lambda_m^nt[\frac{\kappa_m^n}{p + \kappa_m^n}] + (\nu_m^n)^2}.
\]
The expression of the decoherence function in Eq. (23) returns to the result obtained in Ref. [73] in the case that the statistical property of the environmental noise is stationary, namely, when the environment is in equilibrium. For the case of no environmental frequency modulation \(\Omega_m^n = 0\), the memory kernel in Eq. (22) becomes an exponential form and the decoherence function in Eq. (23) is in consistent with that derived in Refs. [71, 72].
IV. CONCLUSIONS

We have proposed the generalized stochastic Liouville equations to investigate the coherent dynamics of single molecule systems coupled to the environment of which the statistical properties are both nonstationary and non-Markovian. The coherence of the quantum system can be obtained by means of the generalized stochastic Liouville equation in two separated averages. The generalized stochastic Liouville equation yields a generalized master equation containing a memory kernel related to the intrinsic system Hamiltonian and the non-Markovian features of the environmental noise and it returns to the well-known framework established by Kubo in the limit case that the environment is in equilibrium and memoryless. We express the exact analytical results of the coherent dynamics of the single molecule systems induced by the nonstationary and non-Markovian RTN. The analytical expressions of the system coherence are in well consistent with that obtained by means of some other theoretical approaches.

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