

Quantum Dynamics of System Strongly Coupled to Low-Temperature Colored Noise Bath: Reduced Hierarchy Equations Approach

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Reduced equations of motion for a two-level system strongly coupled to a harmonic oscillators bath are constructed by extending the hierarchy of equations introduced by Tanimura and Kubo [J. Phys. Soc. Jpn. **58** (1989) 101]. The set of equations treats the bath in a nonperturbative manner and is applicable to a low-temperature system with taking into account the correlation time of noise. By numerically calculating linear absorption spectra for different temperatures, we demonstrate that the present theory is not afflicted with the dynamical positivity problem that occurs at low temperatures without the rotating wave approximation. Remarkable changes are found in the spectra when the temperature is lower than the resonant energy of the two-level system.

KEYWORDS: quantum dissipative system, Gaussian–Markovian noise, nonperturbative treatment, low temperature, hierarchy of equations

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Quantum systems in a dissipative environment have been a subject of great interest for many years.^{1,2)} There are several approaches to deal with such systems,^{3–7)} but the most commonly used approach is based on the reduced equations of motion, which are obtained by tracing over the heat-bath degrees of freedom under the rotating wave approximation (RWA) or high-temperature approximation. The perturbative approximation together with the factorization condition is also commonly employed. These approximations, however, limit the applicability of the equations.⁸⁾ Due to the advent of experimental technology, we can now test a system under ultimate conditions, i.e., at very low temperatures, in a very short time scale with extreme accuracy. It is therefore crucial to establish a reliable theory that can accurately treat the effects of dissipation under such extreme conditions. In this letter, we show that we can remove the limitations by extending the hierarchy treatments in the equations of motion developed by Tanimura and Kubo.⁹⁾

We consider a spin-Boson system modeled by a two-level system (TLS) coupled to a harmonic oscillators bath. The total Hamiltonian is expressed as¹⁾

$$\hat{H}_{\text{tot}} = \frac{\hbar\omega_0}{2} (\hat{\psi}^\dagger \hat{\psi} - \hat{\psi} \hat{\psi}^\dagger) + \sum_j \left[\frac{\hat{p}_j^2}{2m_j} + \frac{m_j\omega_j^2}{2} \left(\hat{x}_j - \frac{c_j \hat{V}}{m_j\omega_j^2} \right)^2 \right], \quad (1)$$

where $\hat{\psi}^\dagger$ and $\hat{\psi}$ are the Fermion creation and annihilation operators, which obey the anticommutator relations $\{\hat{\psi}, \hat{\psi}^\dagger\} = 1$ and $\{\hat{\psi}, \hat{\psi}\} = \{\hat{\psi}^\dagger, \hat{\psi}^\dagger\} = 0$. The parameters \hat{x}_j , \hat{p}_j , m_j and ω_j are the coordinate, momentum, mass, and frequency of the j th bath oscillator, respectively. In eq. (1), the system-bath interaction is expressed as $\hat{H}_I = -\hat{V} \sum_j c_j \hat{x}_j$, where c_j denotes the coupling constant to the j th bath mode, and \hat{V} is the dimensionless operator of the TLS given by

$$\hat{V} = V_1 \cdot (\hat{\psi}^\dagger + \hat{\psi}) + \frac{V_2}{2} \cdot (\hat{\psi}^\dagger \hat{\psi} - \hat{\psi} \hat{\psi}^\dagger), \quad (2)$$

with V_1 and V_2 being real C-numbers. Note that under the rotating wave approximation for the system-bath coupling, the inelastic part $\hat{\psi}^\dagger + \hat{\psi}$ is responsible for the longitudinal (T_1 -type) and transversal (T_2 -type) relaxation processes in the TLS, while the elastic part $\hat{\psi}^\dagger \hat{\psi} - \hat{\psi} \hat{\psi}^\dagger$ gives rise to the pure dephasing (T_2^* -type) process.¹²⁾ Although our treatment can handle both terms, here we focus on the energy relaxation process and set $V_1 = 1$ and $V_2 = 0$.

All information on the bath is contained in the spectral distribution function $J(\omega) = \sum_j [c_j^2 / (2m_j\omega_j)] \delta(\omega - \omega_j)$. Here we consider the nearly Gaussian–Markovian noise bath, whose distribution function is given by the Ohmic form with the Lorentzian cutoff:⁹⁾

$$J(\omega) = \frac{\hbar\zeta}{\pi\omega_0} \frac{\gamma^2\omega}{\omega^2 + \gamma^2}, \quad (3)$$

where γ represents the width of the spectral distribution of the bath modes and is related to the correlation time of the noise induced by the bath, $\tau_c = 1/\gamma$. The parameter ζ is related to the system-bath coupling strength.

To reduce the equation of motion, we set the temporary initial condition of the total system in the factorized form as $\hat{\rho}_{\text{tot}}(t_0 \rightarrow -\infty) = \hat{\rho}(-\infty) \otimes \hat{\rho}_{\text{B}}^{\text{eq}}$, where $\hat{\rho}(-\infty)$ is the initial state of the TLS and $\hat{\rho}_{\text{B}}^{\text{eq}}$ is the thermal equilibrium state of the bath. This is not the ‘true’ equilibrium state of the total system because it neglects the correlated effects of the system-bath interaction. The true equilibrium initial condition can be set by utilizing the hierarchy equations, as will be explained below.

Now let us introduce the Fermion coherent state $|\psi\rangle$, which satisfies $\hat{\psi}|\psi\rangle = \psi|\psi\rangle$ and $\langle\psi|\hat{\psi}^\dagger = \langle\psi|\psi\rangle$, where ψ and $\langle\psi|$ are Grassmann variables. Then, the reduced density matrix element for the TLS is expressed in the path integral form with the factorized initial condition as

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$$\rho(\bar{\psi}, \psi'; t) = \int \mathcal{D}\bar{\psi} \mathcal{D}\psi \int \mathcal{D}\bar{\psi}' \mathcal{D}\psi' \rho(\bar{\psi}_0, \psi'_0; t_0) \times e^{iS[\bar{\psi}, \psi]/\hbar} \mathcal{F}_{\text{FV}}[\bar{\psi}, \psi; \bar{\psi}', \psi'] e^{-iS[\bar{\psi}', \psi']/\hbar}. \quad (4)$$

Here, $S[\bar{\psi}, \psi]$ is the action of the TLS defined by $S[\bar{\psi}, \psi] = \int_{t_0}^t ds L(\bar{\psi}, \psi)$ for the Lagrangean,

$$L(\bar{\psi}, \psi) = \frac{i\hbar}{2} (\bar{\psi}\dot{\psi} - \dot{\bar{\psi}}\psi) - \frac{\hbar\omega_0}{2} (\bar{\psi}\psi - \psi\bar{\psi}), \quad (5)$$

and $\mathcal{F}_{\text{FV}}[\bar{\psi}, \psi; \bar{\psi}', \psi']$ is the Feynman–Vernon influence functional given by¹³⁾

$$\begin{aligned} \mathcal{F}_{\text{FV}}[\bar{\psi}, \psi; \bar{\psi}', \psi'] &= \exp\left(-\frac{1}{\hbar} \int_0^\infty d\omega J(\omega) \int_{t_0}^t ds \int_{t_0}^s ds' V^\times(s) \right. \\ &\times \left[V^\times(s') \coth\left(\frac{\beta\hbar\omega}{2}\right) \cos(\omega(s-s')) \right. \\ &\left. \left. - iV^\circ(s') \sin(\omega(s-s')) \right] \right). \end{aligned} \quad (6)$$

Here, we have introduced the abbreviations

$$V^\times(t) \equiv V(\bar{\psi}(t), \psi(t)) - V(\bar{\psi}'(t), \psi'(t)) \quad (7)$$

and

$$V^\circ(t) \equiv V(\bar{\psi}(t), \psi(t)) + V(\bar{\psi}'(t), \psi'(t)). \quad (8)$$

For the distribution eq. (3), we can rewrite eq. (6) as

$$\begin{aligned} \mathcal{F}_{\text{FV}}[\bar{\psi}, \psi; \bar{\psi}', \psi'] &= \exp\left(\int_{t_0}^t ds \int_{t_0}^s ds' \Phi(s) \Theta(s') \gamma e^{-\gamma(s-s')}\right) \\ &\times \prod_{k=1}^{\infty} \exp\left(\int_{t_0}^t ds \int_{t_0}^s ds' \Phi(s) \Psi_k(s') v_k e^{-v_k(s-s')}\right), \end{aligned} \quad (9)$$

with

$$\Phi(t) \equiv iV^\times(t), \quad (10)$$

$$\Theta(t) \equiv \frac{i\zeta}{\beta\hbar\omega_0} \left[-i \frac{\beta\hbar\gamma}{2} V^\circ(t) + \frac{\beta\hbar\gamma}{2} \cot\left(\frac{\beta\hbar\gamma}{2}\right) V^\times(t) \right], \quad (11)$$

$$\Psi_k(t) \equiv \frac{i\zeta}{\beta\hbar\omega_0} \frac{2\gamma^2}{v_k^2 - \gamma^2} V^\times(t), \quad (12)$$

where $v_k = 2\pi k/(\beta\hbar)$ is a Bosonic Matsubara frequency. It was shown that one can deduce the hierarchy of equations of motion for the above system by evaluating the time derivative of the reduced density matrix elements for the high-temperature case.⁹⁾ Inclusion of the low-temperature correction terms that appeared in eq. (9) was also discussed¹⁰⁾ and numerically examined.¹⁴⁾ The former formulation for the low-temperature correction was, however, somewhat cumbersome and impractical for systems with large degrees of freedom, since the members of the hierarchy elements relate in a rather complex manner and their number quickly increases as the temperature decreases. In this letter, we present a simpler formulation to deal with low-temperature correction terms.

First, we should notice that if we choose K so as to satisfy $v_K = 2\pi K/(\beta\hbar) \gg \omega_0$, where ω_0 is the characteristic frequency of the TLS, the factor $e^{-v_K(s-s')}$ in eq. (9) can be replaced by Dirac's delta function as

$$v_k e^{-v_k(s-s')} \simeq \delta(s-s') \quad (k \geq K+1). \quad (13)$$

Thus, by choosing the relevant K , eq. (9) can be reduced to

$$\begin{aligned} \mathcal{F}_{\text{FV}}[\bar{\psi}, \psi; \bar{\psi}', \psi'] &\simeq \exp\left(-\int_{t_0}^t ds \Phi(s) e^{-\gamma s} \left[-\int_{t_0}^s ds' \gamma \Theta(s') e^{\gamma s'}\right]\right) \\ &\times \prod_{k=1}^K \exp\left(-\int_{t_0}^t ds \Phi(s) e^{-v_k s} \left[-\int_{t_0}^s ds' v_k \Psi_k(s') e^{v_k s'}\right]\right) \\ &\times \prod_{k=K+1}^{\infty} \exp\left(\int_{t_0}^t ds \Phi(s) \Psi_k(s)\right). \end{aligned} \quad (14)$$

In order to derive the equation of motion, we introduce the auxiliary operator $\hat{\rho}_{j_1, \dots, j_K}^{(n)}(t)$ by its matrix element as^{9–11)}

$$\begin{aligned} \rho_{j_1, \dots, j_K}^{(n)}(\bar{\psi}, \psi'; t) &= \int \mathcal{D}\bar{\psi} \mathcal{D}\psi \int \mathcal{D}\bar{\psi}' \mathcal{D}\psi' \rho(\bar{\psi}_0, \psi'_0; t_0) \\ &\times \left\{ e^{-\gamma t} \left[-\int_{t_0}^t ds \gamma \Theta(s) e^{\gamma s} \right] \right\}^n \\ &\times \prod_{k=1}^K \left\{ e^{-v_k t} \left[-\int_{t_0}^t ds v_k \Psi_k(s) e^{v_k s} \right] \right\}^{j_k} \\ &\times e^{iS[\bar{\psi}, \psi]/\hbar} \mathcal{F}_{\text{FV}}[\bar{\psi}, \psi; \bar{\psi}', \psi'] e^{-iS[\bar{\psi}', \psi']/\hbar}, \end{aligned} \quad (15)$$

for nonnegative integers n, j_1, \dots, j_K . Note that only $\hat{\rho}_{0, \dots, 0}^{(0)}(t) = \hat{\rho}(t)$ has a physical meaning and the others are introduced for computational purposes only. Differentiating $\rho_{j_1, \dots, j_K}^{(n)}(\bar{\psi}, \psi'; t)$ with respect to t , we obtain the following hierarchy of equations in operator form:

$$\begin{aligned} \frac{\partial}{\partial t} \hat{\rho}_{j_1, \dots, j_K}^{(n)}(t) &= \\ &- \left[i\hat{\mathcal{L}} + n\gamma + \sum_{k=1}^K (j_k v_k + \hat{\Phi} \hat{\Psi}_k) + \hat{\Xi} \right] \hat{\rho}_{j_1, \dots, j_K}^{(n)}(t) \\ &- \hat{\Phi} \hat{\rho}_{j_1, \dots, j_K}^{(n+1)}(t) - n\gamma \hat{\Theta} \hat{\rho}_{j_1, \dots, j_K}^{(n-1)}(t) \\ &- \sum_{k=1}^K \hat{\Phi} \hat{\rho}_{j_1, \dots, j_{k+1}, \dots, j_K}^{(n)}(t) \\ &- \sum_{k=1}^K j_k v_k \hat{\Psi}_k \hat{\rho}_{j_1, \dots, j_{k-1}, \dots, j_K}^{(n)}(t), \end{aligned} \quad (16)$$

where $\hat{\mathcal{L}}$ is the Liouvillian of the TLS, the relaxation operators $\hat{\Phi}$, $\hat{\Theta}$, and $\hat{\Psi}_k$ are obtained by replacing $V^\times(t) \rightarrow \hat{V}^\times$ and $V^\circ(t) \rightarrow \hat{V}^\circ$ in eqs. (10)–(12), and

$$\hat{\Xi} \equiv \frac{\zeta}{\beta\hbar\omega_0} \left[1 - \frac{\beta\hbar\gamma}{2} \cot\left(\frac{\beta\hbar\gamma}{2}\right) \right] \hat{V}^\times \hat{V}^\times. \quad (17)$$

In the above equations, we introduced the following two superoperators for any physical operator $\hat{\mathcal{O}}$: $\hat{\mathcal{O}}^\times \hat{f} \equiv \hat{\mathcal{O}} \hat{f} - \hat{f} \hat{\mathcal{O}}^\dagger$ and $\hat{\mathcal{O}}^\dagger \hat{f} \equiv \hat{\mathcal{O}} \hat{f} + \hat{f} \hat{\mathcal{O}}^\dagger$, for any operand operator \hat{f} . The above expression is similar to the expression given in refs. 9–11, and the number of the hierarchy in the present formula is safely suppressed by K because of the operator $\hat{\Xi}$ in eq. (17). What makes the present formulation unique is the way in which the hierarchy is terminated, as will be explained below.

The hierarchy equation, eq. (16), continues to infinity, which is not easy to solve numerically. To terminate eq. (16), let us solve eq. (16) formally as

$$\begin{aligned} \hat{\rho}_{j_1, \dots, j_K}^{(n)}(t) = & \int_{t_0}^t ds e^{-[i\hat{\mathcal{L}} + n\gamma + \sum_{k=1}^K (j_k \nu_k + \hat{\Phi}\hat{\Psi}_k) + \hat{\Xi}](t-s)} \\ & \times \left[-\hat{\Phi}\hat{\rho}_{j_1, \dots, j_K}^{(n+1)}(s) - n\gamma\hat{\Theta}\hat{\rho}_{j_1, \dots, j_K}^{(n-1)}(s) \right. \\ & - \sum_{k=1}^K \hat{\Phi}\hat{\rho}_{j_1, \dots, j_{k-1}, j_{k+1}, \dots, j_K}^{(n)}(s) \\ & \left. - \sum_{k=1}^K j_k \nu_k \hat{\Psi}_k \hat{\rho}_{j_1, \dots, j_{k-1}, \dots, j_K}^{(n)}(s) \right]. \end{aligned} \quad (18)$$

If $n\gamma + \sum_{k=1}^K j_k \nu_k$ is large enough compared with the characteristic frequency of the TLS, ω_0 , the time kernel of the integral can be replaced by Dirac's delta function as

$$\left(n\gamma + \sum_{k=1}^K j_k \nu_k \right) e^{-(n\gamma + \sum_{k=1}^K j_k \nu_k)(t-s)} \simeq \delta(t-s), \quad (19)$$

and then eq. (16) becomes

$$\frac{\partial}{\partial t} \hat{\rho}_{j_1, \dots, j_K}^{(n)}(t) \simeq - \left(i\hat{\mathcal{L}} + \sum_{k=1}^K \hat{\Phi}\hat{\Psi}_k + \hat{\Xi} \right) \hat{\rho}_{j_1, \dots, j_K}^{(n)}(t), \quad (20)$$

which works as the terminator for the hierarchy equation, eq. (16). This termination is valid for the integers n, j_1, \dots, j_K , satisfying

$$N \equiv n + \sum_{k=1}^K j_k \gg \frac{\omega_0}{\min(\gamma, \nu_1)} \quad (21)$$

because $n\gamma + \sum_{k=1}^K j_k \nu_k \geq (n + \sum_{k=1}^K j_k) \min(\gamma, \nu_1)$. As shown in Fig. 1, the hierarchical elements $\hat{\rho}_{j_1, \dots, j_K}^{(n)}$ can be represented by the lattice points (n, j_1, \dots, j_K) contained within the $(K+1)$ -simplex,¹⁵⁾ $\sigma^{K+1} = \{(x_0, x_1, \dots, x_K) \in \mathbb{R}^{K+1} \mid 0 \leq \sum_j x_j \leq N, 0 \leq x_j \leq N\}$. In particular, the lattice points corresponding to the terminators lie on a K -face¹⁵⁾ of σ^{K+1} , $\{(x_0, x_1, \dots, x_K) \in \mathbb{R}^{K+1} \mid \sum_j x_j = N, 0 \leq x_j \leq N\}$. This termination is simple and easy to utilize in numerical calculations. In practice, we may use the lower values of N which do not satisfy eq. (21).

The factorized initial condition is, as mentioned above, not the true equilibrium state because the correlation effects of

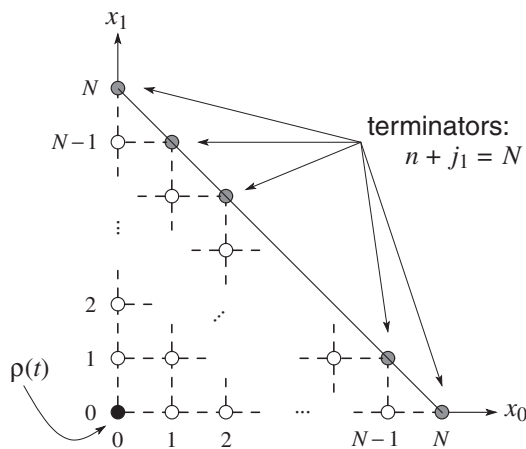


Fig. 1. The tree-like structure of the hierarchical elements in the case of $K=1$. On the x_0x_1 -plane (\mathbb{R}^2), a lattice point (n, j_1) in the right triangle (2-simplex σ^2) denotes a hierarchical element $\hat{\rho}_{j_1}^{(n)}$. The lattice points corresponding to the terminators lie on the hypotenuse (a 1-face of σ^2). The lattice points connecting with each other via the broken line interact by means of the operators $\hat{\Phi}$, $\hat{\Theta}$, and $\hat{\Psi}_1$.

the system-bath coupling are neglected. The present formalism can take into account such effects with the nonzero hierarchical elements $\hat{\rho}_{j_1, \dots, j_K}^{(n)} \neq 0$ for $(n, j_1, \dots, j_K) \neq (0, 0, \dots, 0)$. Note that all hierarchical elements reach steady-state values after a sufficiently long time evolution, even though we started from the factorized initial condition. We regard these steady-state elements as the correlated initial equilibrium condition,¹¹⁾ $\rho_{j_1, \dots, j_K}^{(n)}(t=0)$.

What has to be noticed is that the present formulation does not employ the Markov approximation, the high-temperature assumption, the perturbative approximation, or the rotating wave approximation (RWA). The conventional quantum master equations or the Bloch equation cannot be applied to low-temperature systems, where quantum effects play a major role, without RWA. Generalized quantum master equations can handle a colored noise bath, but can treat only weak system-bath couplings. In the present approach, the nonperturbative manner allows us to treat the strong system-bath interaction. Best of all, our formulation is rather compact and fits numerical calculations.

To demonstrate the advantage of the formalism, we numerically evaluated the time evolution of the density matrix element and linear absorption spectrum for the TLS. We chose the depth of the hierarchy $N=3-10$ for low-temperature cases ($\beta\hbar\omega_0 > 1$), and $N=20-50$ for high-temperature cases ($\beta\hbar\omega_0 < 1$). For all calculations, the accuracies were checked by changing the values of N .

We denote the density matrix elements of the ground state $|g\rangle$, excited state $|e\rangle$ and their coherence at time t , respectively, by $\rho_{gg}(t)$, $\rho_{ee}(t)$, and $\rho_{ge}(t)$. Figure 2 demonstrates how $\rho_{ee}(t)$ attains the steady-state values for various numbers of the Matsubara frequencies $\{\nu_1, \dots, \nu_K\}$, where $K=1, 2, 3$, and 4 from the top to the bottom. The characteristic frequency of the TLS is $\omega_0 = 1000 \text{ cm}^{-1}$ ($1/\omega_0 = 33.3 \text{ fs}$), which is the typical value for intramolecular vibrational motion. The system-bath parameters are $T=300 \text{ K}$ ($\beta\hbar\omega_0 = 4.79$), $\zeta = 0.5 \omega_0$ (strong coupling), and $\gamma = 0.05 \omega_0$. Here, we consider the nonequilibrium initial state $\rho_{ee}(0) = 1$ and $\rho_{gg}(0) = \rho_{ge}(0) = 0$. As is shown in Fig. 2 (note that the ordinate is a log scale), the steady-state value of $\rho_{ee}(t)$ converges in the vicinity of the canonical equilibrium value, $\rho_{ee}^{\text{can}} = e^{-\beta\hbar\omega_0} / (1 + e^{-\beta\hbar\omega_0})$, as the value

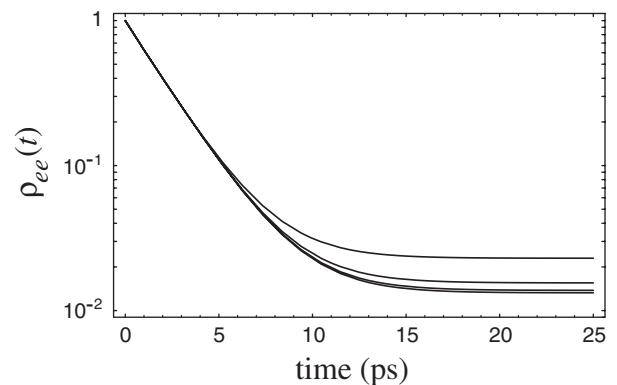


Fig. 2. The relaxation of the excited population $\rho_{ee}(t)$ for the different number of the Matsubara frequencies $\{\nu_1, \dots, \nu_K\}$. From the top to the bottom, $K=1, 2, 3$, and 4. The frequency of the TLS is $\omega_0 = 1000 \text{ cm}^{-1}$. The system-bath parameters are $T=300 \text{ K}$ ($\beta\hbar\omega_0 = 4.79$), $\zeta = 0.5 \omega_0$ (strong coupling), and $\gamma = 0.05 \omega_0$.

of K increases. Our formalism is not afflicted with the positivity problem, which is a notorious and grave issue in conventional quantum master equation theories in which the populations of the excited states become negative at low temperatures.¹⁶⁾ This point deserves explicit emphasis. In addition, we note that the relaxation rates of $\rho_{ee}(t)$ are similar regardless of the values of K . We may, therefore, extract the information on the relaxation process, *e.g.*, relaxation time T_1 and so forth, by means of calculations with small values of K .

In many measurements, including optical and NMR spectroscopies, the dynamics of the system are probed by a correlation function of the physical observables. Let us consider the two-point correlation function defined by

$$R^{(1)}(t) = \frac{i}{\hbar} \text{Tr} \{ [\hat{Q}_F(t), \hat{Q}_F(0)] \hat{\rho}_{\text{tot}}^{\text{eq}} \}. \quad (22)$$

Here, $\hat{\rho}_{\text{tot}}^{\text{eq}}$ is the thermal equilibrium density operator of the total system $\hat{\rho}_{\text{tot}}^{\text{eq}} = \exp(-\beta\hat{H}_{\text{tot}}) / \text{Tr} \exp(-\beta\hat{H}_{\text{tot}})$. $\hat{Q}_F(t)$ is the Heisenberg representation of the operator \hat{Q}_F defined by $\hat{Q}_F \equiv \hat{\psi}^\dagger + \hat{\psi}$, in which the subscript F shows that Q_F is the coordinate of a *Fermionic* oscillator. The Fourier transform of eq. (22), $R^{(1)}(\omega) = \int_0^\infty dt R^{(1)}(t) \exp(i\omega t)$, is equivalent to the linear absorption spectrum of the TLS in optical measurement. In Fig. 3, we present the imaginary part of $R^{(1)}(\omega)$ for $\omega_0/2\pi = 1.0$ THz in the cases of $T = 5$ K ($\beta\hbar\omega_0 = 9.59$) and $T = 300$ K ($\beta\hbar\omega_0 = 0.15$). For both the cases, the system-bath parameters are set to be $\zeta = 0.5\omega_0$ and $\gamma = 0.2\omega_0$. In addition, we chose the number of the Matsubara frequencies $K = 1-4$ for the calculations, as we have verified that further increases in K do not change the linear absorption spectra. Figure 3 shows that the line shape in the low-temperature case exhibits a sharp Lorentzian

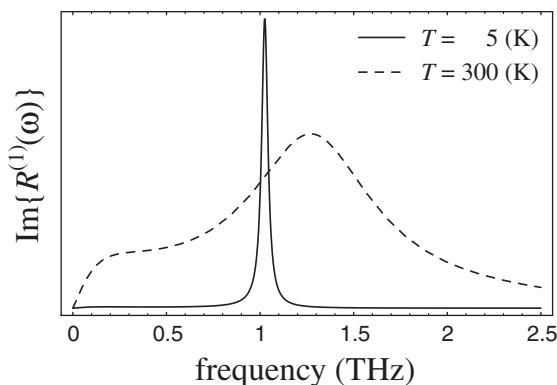


Fig. 3. The linear absorption spectra, $\text{Im}\{R^{(1)}(\omega)\}$, of the TLS ($\omega_0/2\pi = 1.0$ THz) at different temperatures: $T = 5$ K (solid line, $\beta\hbar\omega_0 = 9.59$) and $T = 300$ K (dashed line, $\beta\hbar\omega_0 = 0.15$). The other system-bath parameters are $\zeta = 0.5\omega_0$ and $\gamma = 0.2\omega_0$.

peak, whereas that in the high-temperature case exhibits a featureless broadened peak. This difference can be explained as follows. Since we can define the effective coupling strength¹⁷⁾ as $\zeta_{\text{eff}}(\beta) \equiv \zeta/(\beta\hbar\omega_0)$ from eqs. (11), (12), and (17), the coupling strength effectively decreases when the temperature decreases. Thus, the perturbation on the TLS dies down at the low temperature, and accordingly suppresses the destruction of the quantum coherence.

In this letter, we derived a novel quantum dissipative equation that has applicability to low-temperature systems ($\beta\hbar\omega_0 \gg 1$) strongly coupled to a harmonic bath without employing the rotating wave approximation for the system-bath coupling. Although we only described the spin-Boson system, the extension to a discrete multilevel system or to a potential system expressed in the phase space is straightforward. Applying our approach to the study of quantum dynamics in potential systems, one can explore the intramolecular vibrational energy relaxation, the frequency fluctuation, and their interplay in the condensed phases. In addition, the dissipative effects of mode couplings in multimodal anharmonic potential systems remain the key issue of investigations into molecular dynamics in condensed phases. Extensions of the present theory in these directions are left for future studies.

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- 1) A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg and W. Zwerger: *Rev. Mod. Phys.* **59** (1987) 1.
- 2) U. Weiss: *Quantum Dissipative Systems* (World Scientific, Singapore, 1999) 2nd ed.
- 3) R. Egger and C. H. Mak: *Phys. Rev. B* **50** (1994) 15210.
- 4) J. Cao, L. W. Ungar and G. A. Voth: *J. Chem. Phys.* **104** (1996) 4189.
- 5) N. Makri: *J. Chem. Phys.* **106** (1997) 2286.
- 6) J. Shao and P. Hänggi: *Phys. Rev. Lett.* **81** (1998) 5710.
- 7) J. T. Stockburger and C. H. Mak: *J. Chem. Phys.* **110** (1999) 4983.
- 8) P. Meystre and M. Sargent III: *Elements of Quantum Optics* (Springer-Verlag, Berlin, 1999) 3rd ed.
- 9) Y. Tanimura and R. Kubo: *J. Phys. Soc. Jpn.* **58** (1989) 101.
- 10) Y. Tanimura: *Phys. Rev. A* **41** (1990) 6676.
- 11) Y. Tanimura and P. G. Wolynes: *Phys. Rev. A* **43** (1991) 4131.
- 12) Y. Tanimura and T. Steffen: *J. Phys. Soc. Jpn.* **69** (2000) 4095.
- 13) R. P. Feynman and F. L. Vernon: *Ann. Phys. (N.Y.)* **24** (1963) 118.
- 14) R. X. Xu, P. Cui, X. Q. Li, Y. Mo and Y. J. Yan: *J. Chem. Phys.* **122** (2005) 041103.
- 15) I. M. Singer and J. A. Thorpe: *Lecture Notes on Elementary Topology and Geometry* (Springer-Verlag, New York, 1967).
- 16) E. B. Davies: *Quantum Theory of Open Systems* (Academic Press, London, 1976).
- 17) A. Ishizaki and Y. Tanimura: *J. Chem. Phys.* **123** (2005) 014503.