

System Bath Correlations and the Nonlinear Response of Qubits

Arend G. DIJKSTRA* and Yoshitaka TANIMURA†

Department of Chemistry, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan

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In this letter we study qubits coupled to the bath formed by their environment. Although entanglement of the qubits is a well-known topic, much less effort has gone into the description of the correlations between the qubits and the bath. Here, we investigate these correlations, and study their effect on the qubits in equilibrium and their dynamics following the interaction with one or several external pulses. We find that a correct description of the correlations at the moment of these interactions is essential for a correct understanding of the dynamics.

KEYWORDS: qubit, system-bath, nonlinear response, initial correlations, non-Markovian

The entanglement of two qubits (two-level systems) measures the extent to which they are not separable, and is a purely quantum effect. It is formed and destroyed by the interaction of the qubit system with its environment. Formation of entanglement is often the result of a deliberate preparation procedure, while the destruction occurs because of uncontrollable interactions. After the preparation of an entangled state, the entanglement typically decays with time to an equilibrium value. In the case of Markovian dynamics, the entanglement smoothly decays with time, while temporary increases are possible if the evolution is non-Markovian. The dynamics of entanglement shows some surprising characteristics. Even if the single qubit coherence decays smoothly, the entanglement can go to zero in a finite time, a phenomenon called “sudden death” of entanglement.¹⁾ Furthermore, the entanglement can be revived from death, i.e., the system can become entangled after a period of zero entanglement.

All these time dependent phenomena are caused by the interaction of the qubits with a bath. Entanglement which is lost from the system during the time evolution is stored in the bath.²⁾ Crucially, correlations are created between the qubit system and the bath. Their presence influences the dynamics in the system. In particular, correlations present at the moment of the interaction with an external force are not well studied. Their detailed understanding is the topic of this letter. Of course, correlations are formed continuously when a system is placed in contact with a heat bath. This is closely related to the reorganization energy known in chemical physics.^{3,4)} In the case of non-Markovian dynamics, the correlations acts as a memory for the system, and influences the system entanglement at a later point in time.⁵⁾ Besides this, there is another phenomenon which is important for a complete description of the dynamics: the entanglement of system and bath states at time zero (where zero can be chosen arbitrarily, but in this paper will represent the moment of interaction with an external pulse). In general, this quantity, which reports on initial correlations,⁶⁻⁹⁾ does not vanish and can influence the system during its time evolution.¹⁰⁾

Theoretically, the dynamics of a system in contact with its environment is often studied using master equations, which have been extended to include initial correlations.¹¹⁾ It is

often assumed that the characteristic time scale of the environment is much faster than the dynamics in the system. A Lindblad form master equation can then be derived.¹²⁻¹⁴⁾ Because of the Markovian approximation of a fast bath, such approaches cannot be used to study the memory effect of the bath. The key effect is not the strength of the system bath coupling, which can be included in master equations,^{15,16)} but the time scale on which the system bath correlations are dissipated.

The dynamics of entanglement in the system can be studied experimentally by applying one or more external forces. These forces bring the system out of equilibrium, and can be used to study its subsequent time evolution. While a single external force probes the system’s linear response, multiple forces can be applied to measure the nonlinear response, which contains more detailed information about the system, and, of particular interest here, of the system bath correlations.

As a simple example, we start by studying a single qubit in the pure dephasing case, where the system Hamiltonian H_S commutes with the system bath interaction H_{SB} . This case is exactly solvable when the system linearly couples to a bath of harmonic oscillators.^{17,18)}

Because the system Hamiltonian commutes with the system bath interaction, the dynamics due to the system Hamiltonian can be separated from the interaction. In the interaction picture with respect to the bath Hamiltonian, denoted I , the time evolution is given by

$$\rho(t) = e^{-iH_S^\times t/\hbar} \langle e_+^{-i/\hbar \int_0^t d\tau H_{SB}^\times(\tau)} \rangle \rho(0). \quad (1)$$

In this equation, e_+ denotes the time-ordered exponential, $A^\times B = [A, B]$ denotes the commutator, and $\langle \dots \rangle = \text{Tr}_B \dots \exp(-\beta H_B) / \text{Tr}_B \exp(-\beta H_B)$ is the thermal average over the bath at a temperature $T = 1/k_B \beta$. In writing this equation, we have made the crucial assumption that the system and the bath are independent at time zero, which is not in general valid.

The propagator in this equation can be expanded exactly using the cumulant expansion¹⁷⁾ or path integral methods.¹⁹⁾ By writing the components of the density matrix as an excited state population $P = \langle 1|\rho(t)|1 \rangle$ and a coherence $C(t) = \langle 1|\rho(t)|2 \rangle$, $\rho(t) = (P, C(t); C^*(t), 1 - P)$, the population P is time-independent and the time evolution of the coherence can be derived as

*E-mail: dijkstra@kuchem.kyoto-u.ac.jp

†E-mail: tanimura@kuchem.kyoto-u.ac.jp

$$C(t) = e^{-i\epsilon t/\hbar} e^{-g(t)} C(0), \quad (2)$$

with the dephasing function $g(t) = 1/\hbar^2 \int_0^t dt' \int_0^{t'} dt'' L(t'')$. Here $L(t) = \sum_{\alpha} g_{\alpha}^2 \langle x_{\alpha}(t)x_{\alpha}(0) \rangle$ is the correlation function. It can be written in terms of the spectral density

$$\mathcal{J}(\omega) = \frac{\pi}{2} \sum_{\alpha} \frac{g_{\alpha}^2}{m_{\alpha}\omega_{\alpha}} \delta(\omega - \omega_{\alpha})$$

as²⁰⁾

$$L(t) = \frac{1}{\pi} \int_0^{\infty} d\omega \mathcal{J}(\omega) \left(\coth \frac{\beta\hbar\omega}{2} \cos \omega t - i \sin \omega t \right).$$

Note that most master equations are derived at zero temperature in the approximation $\coth \beta\hbar\omega/2 = 1$ and that recently the hierarchy method has also been extended to that case,²¹⁾ although for a limited form of the spectral density. When a model is chosen for the spectral density, the dynamics of the coherence can be evaluated from these formulas. Because the expressions are exact, the formation of system bath correlations during the time evolution and its effect on the dynamics of the system are fully included. What is missing, however, are the correlations in the initial state.

To include these initial correlations between system and bath states, we extend the model by allowing for a preparation time t_1 . During the preparation time, correlations build up. At time zero (which occurs after the preparation time, that is, after t_1 but before t_2), when some interaction with the outside world happens, they are present. They influence the dynamics during the subsequent evolution time t_2 . The system evolves freely before the pulse (t_1) and after the pulse (t_2). We will assume that the external pulse takes the form of a π -pulse, which flips the coherence (interchanges C and C^*). The expression for the coherence $C(t_2)$ can be evaluated in terms of the dephasing function to give¹⁸⁾

$$C(t_2) = e^{i\epsilon t_1/\hbar - i\epsilon t_2/\hbar} e^{-2g(t_1) - 2g(t_2) + g(t_1+t_2)} C^*(-t_1).$$

Correlations at time zero can now be formed by choosing a positive value for t_1 . Its effect can be seen from the presence of the $g(t_1 + t_2)$ term. If t_1 is set to zero, it vanishes.

As a simple model for the system-environment coupling, we choose the Drude spectral density $\mathcal{J}(\omega) = 2\lambda\omega\gamma/(\omega^2 + \gamma^2)$. The dephasing function at high temperature is found to be²²⁾

$$g(t) = \left(\frac{2\lambda}{\beta\hbar\gamma^2} - i \frac{\lambda}{\gamma} \right) (e^{-\gamma t} + \gamma t - 1). \quad (3)$$

The time dependence with and without correlations in the initial state for this model is shown in Fig. 1.

We next study a system of two qubits each coupled to a harmonic heat bath. Extensions to more than two qubits are straightforward. The Hamiltonian for the qubit system is now given by

$$H_S = \sum_{n=1}^2 \epsilon_n c_n^{\dagger} c_n + J(c_1^{\dagger} + c_1)(c_2^{\dagger} + c_2), \quad (4)$$

where ϵ_n is the excitation energy of qubit n and J is a coherent coupling between the two qubits. This coupling promotes entanglement in the system, i.e., if the qubits are initially not entangled, they can become entangled due to the

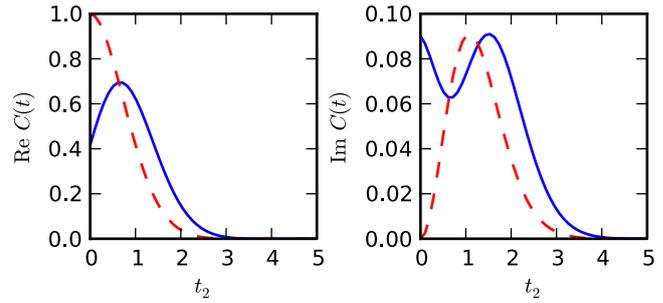


Fig. 1. (Color online) Time dependence of the coherence $C(t)$ with (solid line) and without (dashed line) system bath correlations at time $t_2 = 0$. The solid line was calculated for $t_1 = 1.0$. Parameters are $\beta\hbar\gamma = 0.5$, $\beta\hbar\lambda = 1.0$ and $\epsilon = 0$.

action of J . The excitation energies ϵ_n for each qubit can be different in general, but we take them equal and choose $\epsilon_1 = \epsilon_2 = 1.5J$ in the numerical results in this work. The system bath interaction becomes

$$H_{SB} = \sum_{n=1}^2 V_n X_n = - \sum_{n=1}^2 \sum_{\alpha} g_{n,\alpha} V_n x_{\alpha}. \quad (5)$$

X_n is an effective bath coordinate. The operators V_n , which we choose to be $V_n = c_n^{\dagger} + c_n$, denote the effect of the bath on the qubit n , with coupling strengths $g_{n,\alpha}$. While we take a heat bath coupled to each qubit separately here, more general forms of the interaction, including correlated fluctuations, can be included by a straightforward generalization of the operators V . In our current model, the baths lead to a flip in the state of the qubit.

Because the bath modes are harmonic oscillators which couple linearly to the system, all necessary information on the coupling of the system to the bath is contained in the spectral densities. To proceed, we need to choose a model for the spectral densities. We will take the fluctuations in each qubit to be independent and choose the spectral density of the system bath interaction of each qubit to be of the Drude form,

$$\mathcal{J}_n(\omega) = 2\lambda_n \gamma_n \frac{\omega \gamma_n}{\gamma_n^2 + \omega^2}. \quad (6)$$

While this choice of the spectral density, which corresponds to a single time scale of the system bath interaction (see below), leads to the simplest numerical propagation scheme, an extension to more general spectral densities is possible.^{23,24)}

To understand the effect of this spectral density on the qubit system, we consider the correlation function $L_n(t) = \langle X_n(t)X_n(0) \rangle$, where the average $\langle \dots \rangle$ is taken over the harmonic bath. The correlation functions are found to be exponential,^{25,26)}

$$L_n(t) = \sum_{k=0}^{\infty} c_{nk} e^{-v_{nk}|t|}, \quad (7)$$

where we have defined (for $k \geq 1$) $v_{nk} = 2\pi k/\hbar\beta_n$, $v_{n0} = \gamma_n$, $c_{n0} = \lambda_n \gamma_n (-i + \cot \hbar\beta_n \gamma_n/2)$, $c_{nk} = (4\lambda_n \gamma_n/\beta_n) v_{nk}/(v_{nk}^2 - \gamma_n^2)$. In what follows, we will take equal parameters for each qubit and drop the index n in our notation. The real part of these correlation functions describes the fluctuations induced by the bath at a temperature $T = 1/k_B\beta$, while the

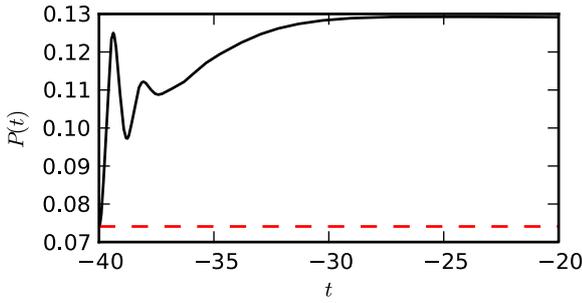


Fig. 2. (Color online) Approach to equilibrium. The initial state $\exp -\beta H_S / \text{Tr} \exp -\beta H_S$ is propagated in time. It is clearly not the equilibrium state, which is found at the end of the propagation. Parameters are $\lambda = J$, $\gamma = 0.5J$, $\beta = 2.5/J$, $\epsilon_1 = \epsilon_2 = 1.5J$. Plotted is the population of the state where both qubits are excited. The dotted line shows the Boltzmann population calculated from the system Hamiltonian only.

imaginary part is responsible for the dissipation of energy. In equilibrium, fluctuations and dissipation are balanced, and the correlation functions obey the fluctuation dissipation theorem.

The quantity of interest in our calculations is the reduced density matrix for the qubits $\rho(t)$, which is found by tracing the complete density matrix $R(t)$ over the bath degrees of freedom: $\rho(t) = \text{Tr}_B R(t)$. In equilibrium at inverse temperature β , the complete density matrix is given by the Boltzmann distribution $R^{\text{eq}} = \exp[-\beta(H_S + H_B + H_{SB})] / \text{Tr} \exp[-\beta(H_S + H_B + H_{SB})]$. Because of the presence of the system bath interaction, no simple analytical expression for the reduced density matrix $\rho^{\text{eq}} = \text{Tr}_B R^{\text{eq}}$ exists. In particular, it is in general not equal to the Boltzmann distribution for the system Hamiltonian, $\rho_0^{\text{eq}} \neq \exp -\beta H_S / \text{Tr} \exp -\beta H_S$ (see Fig. 2). This is a signature of the presence of system bath correlations in equilibrium, and leads to a shift in the linear absorption spectrum.²⁷⁾ If the time dependence of $\rho(t)$ can be obtained from a numerical simulation, the equilibrium distribution can be found after a long time propagation, $\rho^{\text{eq}} = \lim_{t \rightarrow \infty} \rho(t)$, provided that the system is ergodic.

Such numerical propagation schemes exist. The dynamics can be found from numerical path integration schemes,^{28–30)} or from the hierarchy of equations of motion.^{4,24–26,31–34)} In this paper, we will use the hierarchy of equations of motion, where the reduced density matrix is propagated alongside a set of auxiliary density matrices, which allows for the correct inclusion of the system bath correlations. The largest Matsubara frequency taken into account is set to $M = 2$ in this paper. Propagation of the hierarchy of equations of motion starting from a given initial condition produces the reduced density matrix as a function of time. The situation relevant to experiment can be simulated by applying external pulses to the system.

Because the system states can be detected in experiments, we are looking for quantities that depend on the reduced density matrix only. One quantity that reports on the system bath entanglement is the difference between a population in equilibrium and the population given by the Boltzmann distribution with respect to the system Hamiltonian only (P_0), plotted in Fig. 2. The shift from P_0 increases with increasing strength of the system bath coupling (results not shown).

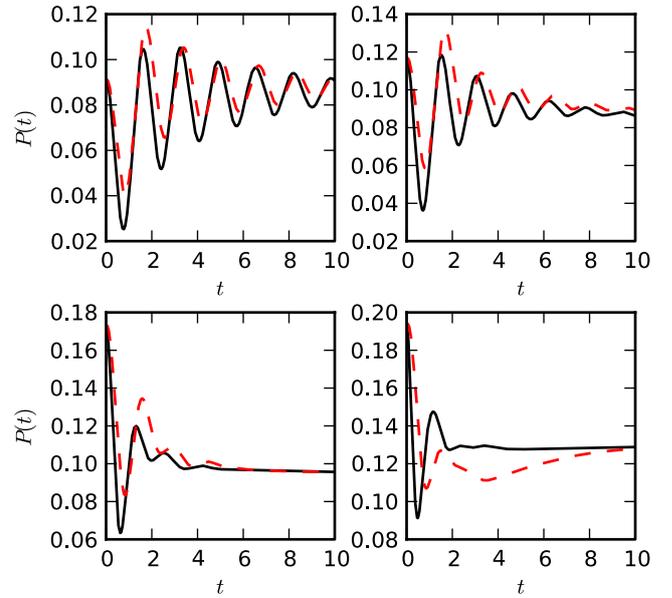


Fig. 3. (Color online) Response after a π -pulse (population of the state where both qubits are in the excited state) with (solid line) and without (dashed line) system bath correlations at time $t = 0$. Parameters are $\lambda = 0.1, 0.2, 0.5, 1.0J$, $\gamma = 0.5J$, $\beta = 2.5/J$, and $\epsilon_1 = \epsilon_2 = 1.5J$.

The shift of the equilibrium population, thus, reports on the system bath correlations. Note that this shift is known as the solvent shift in chemical physics. It is, however, not a directly measurable quantity. We therefore consider the dynamics of the qubit system after interaction with an external pulse. The pulse acts on the total equilibrium of system and bath, which includes the system bath correlations. It brings the system out of equilibrium. Following the pulse, the system will relax, until the equilibrium value is reached again. We will consider a pulse that operates on the first qubit only, and rotates its state over an angle π . The result of such a calculation is shown in Fig. 3. The correlations at $t = 0$ are seen to influence the dynamics for a time up to approximately $5/\gamma$. As expected, the effect is stronger for larger λ . A direct measure of the system bath correlations is the difference between the two lines in Fig. 3.

In order to study this shift in more detail, we consider the response as a function of two times, shown in Fig. 4. Two π pulses separated by a time τ interact with qubit one. Figure 4 shows the response with and without correlations at the moments of interaction with the external forces. Neglecting the correlations corresponds to the result obtained with conventional master equations, for which the system and bath are factorized at the moment of interaction with the external force. This clearly does not capture the complete quantum dynamics, as shown in Fig. 4. We expect this result to hold as well at lower temperature, which is relevant to many quantum information measurements.

In this letter, we studied the role of system bath correlations in the dynamics of qubits interacting with one or several external forces. These correlations is not only formed continuously during the time evolution, but, crucially, is also present at the moment of interaction of the system with external forces. This leads to memory in the system that extends from before to after the interaction. We demonstrated that the resulting response of the qubit system

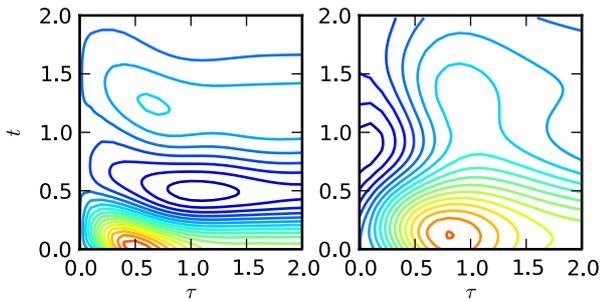


Fig. 4. (Color online) Nonlinear response (population of the state where both qubits are excited) with (left) and without (right) system bath correlations at the moment of interaction with the external pulses. Parameters are $\gamma = 0.5$, $\lambda = 1.0$, and $\beta = 2.5$. Contours are drawn at intervals of 0.01 starting from 0.08.

strongly depends on a correct inclusion of this initial correlations. A challenging question for future work is the representation of the amount of correlation, present by a single number, preferably in a way that is directly observable in nonlinear measurements. It will also be interesting to investigate to what extent the system and the bath are entangled and not only classically correlated, leading to the new concept of “bathentanglement”. Furthermore, it will be of interest to see whether master equations can be developed that capture the system bath correlations and hence can reproduce our results.

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