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Steady quantum coherence in non-equilibrium environment

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ABSTRACT

We study the steady state of a three-level system in contact with a non-equilibrium environment, which is composed of two independent heat baths at different temperatures. We derive a master equation to describe the non-equilibrium process of the system. For the three level systems with two dipole transitions, i.e., the Λ-type and V-type, we find that the interferences of two transitions in a non-equilibrium environment can give rise to non-vanishing steady quantum coherence, namely, there exist non-zero off-diagonal terms in the steady state density matrix (in the energy representation). Moreover, the non-vanishing off-diagonal terms increase with the temperature difference of the two heat baths. Such interferences of the transitions were usually omitted by secular approximation, for it was usually believed that they only take effect in short time behavior and do not affect the steady state. Here we show that, in non-equilibrium systems, such omission would lead to the neglect of the steady quantum coherence.

1. Introduction

Isolated quantum systems evolve unitarily according to the Schrödinger equation, while an open quantum system, which is inevitably coupled to a heat bath in practical, usually quickly lose all its quantum coherence. That is, all the off-diagonal terms of the density matrix of the system $\langle E_m|\rho|E_n\rangle$

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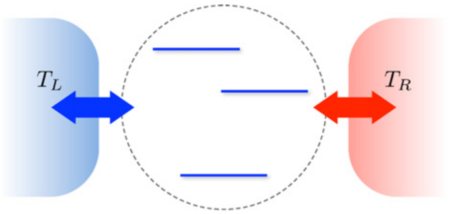

Fig. 1. (Color online) Demonstration for a non-equilibrium system: a multi-level system contacted with two heat baths at different temperatures $T_{L/R}$.

(in the energy representation) will decay to zero when the open system approaches the steady state [1–4]. This phenomenon is called decoherence, and it is also believed that this is why our world appears as a classical one and no macroscopic superposition can exist stably in usual cases [5]. It has been reported that if some non-vanishing steady quantum coherence exists in certain special environment, even with a quite small amount, it can result to some novel physics, such as lasing without inversion [6], or extracting work from a single heat bath [3,7,8].

Then an important question arises: how can quantum coherence survive stably in the steady state against decoherence [3]? In this paper, we find that the steady quantum coherence can indeed exist stably when the system contacts with a non-equilibrium environment, which is composed of multiple equilibrium heat baths at different temperatures. Here we study the steady state of a three-level system, which is coupled to two heat baths with temperatures $T_{L/R}$ respectively (Fig. 1). We find that, for the Λ-type and V-type systems, non-vanishing quantum coherence can exist in the steady state when the temperatures of the two heat baths are different. Moreover, the amount of the nonzero off-diagonal terms increase with the temperature difference ΔT of the two heat baths. While the quantum coherence always vanishes in a Ξ-type system. Here we must emphasize that, unlike previous studies [7,8], in our model there is no quantum coherence in the environment in priori, and the steady quantum coherence in the system is naturally brought in by the non-equilibrium environment.

Physically, this steady quantum coherence results from the interference of transitions in non-equilibrium systems. In the three kinds of three-level systems we study, there are two transition pathways, and there exist interferences between the transitions for the Λ-type and V-type systems [9–16]. We have to point out that such interferences were often omitted by secular approximations in many previous literatures [1,17–19]. Here we show that this omission is consistent for equilibrium environments, i.e., when all the temperatures of different baths equal to each other and thus they become a whole equilibrium heat bath. However, in non-equilibrium systems, such omission of interference between transitions would lead to the neglect of the steady quantum coherence, and that would also lead to some other unphysical results [20,21].

Moreover, in a simple example we will show that the quantum coherence exactly reflects the non-equilibrium flux inside a composite system, thus it has a clear physical meaning and should not be neglected in non-equilibrium systems [21].

The paper is organized as follows. In Section 2, we derive a master equation for a Λ-type system contacting with two heat baths, and discuss the effect of secular approximation. In Section 3, we show that non-vanishing quantum coherence can exist in the non-equilibrium steady state of the Λ-type system, if we take into account the interferences between transitions and do not apply the secular approximation. We also give the condition for the existence of steady quantum coherence. In Section 4, we show that steady quantum coherence can also appear in V-type systems, but cannot appear in Ξ-type systems. In Section 5, we show that the quantum coherence reflects the non-equilibrium flux inside a composite system. Finally we draw conclusions in Section 6.

2. Non-equilibrium Λ-type system

We first consider a Λ-type system contacting with two independent heat baths (bath-L/R) with different temperatures $T_{L/R}$. We derive a master equation via Born–Markovian approximation to describe the dynamics of the open quantum system. Especially, we consider what physical process has been ignored in the conventional secular approximation during the derivation of the master equation.

2.1. Model setup and master equation

We consider a Λ-type system [see Fig. 2(a)], which is described by the Hamiltonian

$$\hat{H}_S = \sum_{n=1}^{3} E_n |E_n\rangle\langle E_n|, \tag{1}$$

where we denote the three energy levels by $|E_n\rangle = |g_1\rangle,\ |g_2\rangle$ (the lower two states) and $|E_n\rangle = |e\rangle$ (the highest excited state) correspondingly, with eigenenergy $E_{g_1},\ E_{g_2}$ and E_e.

Due to the interaction with the environment, there are two transitions in the system, i.e., $|g_1\rangle \leftrightarrow |e\rangle$ and $|g_2\rangle \leftrightarrow |e\rangle$. Here we use the lowering and raising operators to represent these two transitions, denoted as $\hat{\varphi}_i^- := |g_i\rangle\langle e|$ and $\hat{\varphi}_i^+ := |e\rangle\langle g_i|$ respectively for $i = 1, 2$. And we denote the energy difference of each transition as $\varepsilon_i := E_e - E_{g_i}$.

The two heat baths are modeled as collections of boson modes, described by the Hamiltonian

$$\hat{H}_B = \sum_{k_L} \omega_{k_L} \hat{b}_{k_L}^\dagger \hat{b}_{k_L} + \sum_{k_R} \omega_{k_R} \hat{b}_{k_R}^\dagger \hat{b}_{k_R}. \tag{2}$$

The two transitions $\hat{\varphi}_i^\pm$ of the Λ-type system are coupled to both the two heat baths, which are in their own equilibrium thermal states $\rho_{B,\alpha}^{\text{th}} = \mathcal{Z}_\alpha^{-1} \exp[-\sum \omega_{k_\alpha} \hat{b}_{k_\alpha}^\dagger \hat{b}_{k_\alpha}/T_\alpha]$ with temperatures T_α for $\alpha = L,\ R$ respectively. The interaction Hamiltonian reads

$$\hat{H}_{SB} = \hat{\varphi}_1^+ \cdot (\hat{B}_{1,L} + \hat{B}_{1,R}) + \hat{\varphi}_1^- \cdot (\hat{B}_{1,L}^\dagger + \hat{B}_{1,R}^\dagger) + \hat{\varphi}_2^+ \cdot (\hat{B}_{2,L} + \hat{B}_{2,R}) + \hat{\varphi}_2^- \cdot (\hat{B}_{2,L}^\dagger + \hat{B}_{2,R}^\dagger), \tag{3}$$

where

$$\hat{B}_{i,\alpha} = \sum_{k_\alpha} g_{i,k_\alpha} \hat{b}_{k_\alpha}, \quad \alpha = L, R \text{ and } i = 1, 2$$

is the collective operator of bath-α coupled to transition-i.

To derive a master equation for this open quantum system, we apply the Born–Markovian approximation in the interaction picture (without the secular approximation) [1],

$$\dot{\rho} = -\int_0^\infty ds\, \mathrm{Tr}_B\big[\hat{H}_{SB}(t),\ [\hat{H}_{SB}(t-s),\ \rho(t) \otimes \rho_B]\big]. \tag{4}$$

And we obtain the following time-dependent master equation in the interaction picture (the detailed calculation is presented in Appendix A),

$$\begin{aligned}\dot{\rho} = \sum_{i,j=1}^{2} & e^{i\Delta_{ij}t}\Big\{\Gamma_{ji}^+(\varepsilon_j)\big[\hat{\varphi}_i^+,\ \rho\hat{\varphi}_j^-\big] + \Gamma_{ji}^+(\varepsilon_i)\big[\hat{\varphi}_i^+\rho,\ \hat{\varphi}_j^-\big]\Big\} \\ & + e^{-i\Delta_{ij}t}\Big\{\Gamma_{ij}^-(\varepsilon_j)\big[\hat{\varphi}_i^-,\ \rho\hat{\varphi}_j^+\big] + \Gamma_{ij}^-(\varepsilon_i)\big[\hat{\varphi}_i^-\rho,\ \hat{\varphi}_j^+\big]\Big\},\end{aligned} \tag{5}$$

where $\Delta_{ij} := \varepsilon_i - \varepsilon_j$, and

$$\begin{aligned}\Gamma_{ij}^-(\omega) &:= \frac{1}{2}\gamma_{ij}^{(L)}(\omega)[N_L(\omega)+1] + \frac{1}{2}\gamma_{ij}^{(R)}(\omega)[N_R(\omega)+1], \\ \Gamma_{ij}^+(\omega) &:= \frac{1}{2}\gamma_{ij}^{(L)}(\omega)N_L(\omega) + \frac{1}{2}\gamma_{ij}^{(R)}(\omega)N_R(\omega),\end{aligned} \tag{6}$$

are called the dissipation rates. Here $N_\alpha(\omega) := [\exp(\omega/T_\alpha) - 1]^{-1}$ is the Planck distribution. And $\gamma_{ij}^{(\alpha)}(\omega)$ is the coupling spectrum of bath-α, which is defined as

$$\gamma_{ij}^{(\alpha)}(\omega) := 2\pi \sum_{k_\alpha} g_{i,k_\alpha}^* g_{j,k_\alpha} \delta(\omega - \omega_{k_\alpha}) = [\gamma_{ji}^{(\alpha)}(\omega)]^*. \tag{7}$$

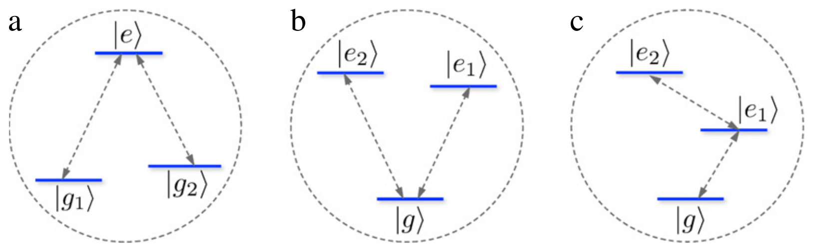

Fig. 2. (Color online) Three types of three-level systems: (a) Λ-type, (b) V-type and (c) Ξ-type.

We should notice that besides the *individual spectrum* $\gamma_{ii}(\omega)$ for each transition, we also have the *cross spectrums* [22], $\gamma_{12}(\omega) = [\gamma_{21}(\omega)]^*$, which describe the interference effect between the two transitions [9,12,14].

There is a relation between the cross spectrums $\gamma_{ij}^{(\alpha)}(\omega)$ and the individual spectrums $\gamma_{ii}^{(\alpha)}(\omega)$, shown as follows (see the proof in Appendix B),

$$|\gamma_{12}^{(\alpha)}(\omega)|^2 = f_\alpha(\omega) \cdot \gamma_{11}^{(\alpha)}(\omega)\gamma_{22}^{(\alpha)}(\omega), \tag{8}$$

where $f_\alpha(\omega)$ is a weight factor and $0 \le f_\alpha(\omega) \le 1$. If $f_\alpha(\omega) = 0$, it means there is no interference between transitions. In some special cases, we have $f_\alpha(\omega) = 1$, which means the interference effect achieves the maximum. In general cases, the concrete form of $f_\alpha(\omega)$ depends on the form of coupling strength g_{i,k_α} in specific physical systems [23,24] (see also the example in Appendix B). For example, in quantum optics, the weight factor $f_\alpha(\omega)$ is equivalent to the p parameter which measures the angle between the transition dipole moments [10,13–16].

2.2. Secular approximation

We can understand the physical meaning of each summation term in the master equation (5) as a second order process. In fact, due to the interaction with the environment, the system first absorbs an energy quanta from the environment through the transition $\hat{\varphi}_i^+$, and then immediately followed by emitting an energy quanta through transition $\hat{\varphi}_j^-$ back to the environment. And the reversed process also happens, namely, the system first emits an energy quanta through transition $\hat{\varphi}_i^-$, and then absorbs through transition $\hat{\varphi}_j^+$.

If these two successive energy exchange processes experience the same transition ($i = j$), the total process is stable. While if these two processes experience two different transitions ($i \neq j$), which usually have different energies, the total process, with an oscillating factor $\exp(i\Delta_{ij}t)$, is often considered to be not stable, which will averagely vanish to zero after oscillations for several periods $\Delta t \sim h/\Delta_{ij}$ [25]. We call them cross transitions, and these terms results from the interference of the two transitions. It was believed that such cross transition terms would have no effect after a long enough oscillating time $t \gg \hbar/\left|\Delta_{ij}\right|$ [1,25], and they do not contribute to the steady state ($t \to \infty$).

For this reason, the secular approximation is often applied to Eq. (5), and only the terms with $\Delta_{ij} = \varepsilon_i - \varepsilon_j = 0$ are remained. Then we obtain a time-independent Lindblad master equation [1,17–19],

$$\begin{aligned}\dot{\rho} = \sum_{i=1}^{2} &\Gamma_{ii}^+(\varepsilon_i)\big(2\hat{\varphi}_i^+\rho\hat{\varphi}_i^- - \hat{\varphi}_i^-\hat{\varphi}_i^+\rho - \rho\hat{\varphi}_i^-\hat{\varphi}_i^+\big)\\ &+ \Gamma_{ii}^-(\varepsilon_i)\big(2\hat{\varphi}_i^-\rho\hat{\varphi}_i^+ - \hat{\varphi}_i^+\hat{\varphi}_i^-\rho - \rho\hat{\varphi}_i^+\hat{\varphi}_i^-\big).\end{aligned} \tag{9}$$

We should notice that all the cross transition terms are omitted by the secular approximation in this master equation.

Here we make some clarification on our terminology in this paper. When we say "secular approximation", we mean the omission of the cross transition terms we mentioned above with energy difference $\Delta_{ij} = \varepsilon_i - \varepsilon_j \neq 0$. When we say "rotating-wave approximation (RWA)", we mean the omission of double creation or annihilation terms, for example, in the derivation of Jaynes–Cummings coupling from dipole interaction [26]. The interaction Hamiltonian we used in Eq. (3) is usually obtained via RWA in real physics system.

For the master equation (9) with secular approximation, the equations for the diagonal and off-diagonal terms of ρ are decoupled [1,17–19], and we can obtain a rate equation only involving the populations $\bar{n}_i := \langle g_i|\rho|g_i\rangle$ and $\bar{n}_e := \langle e|\rho|e\rangle$ of each energy level as follows,

$$\begin{aligned}\dot{\bar{n}}_1 &= 2\Gamma_{11}^{-}(\varepsilon_1)\bar{n}_e - 2\Gamma_{11}^{+}(\varepsilon_1)\bar{n}_1,\\ \dot{\bar{n}}_2 &= 2\Gamma_{22}^{-}(\varepsilon_2)\bar{n}_e - 2\Gamma_{22}^{+}(\varepsilon_2)\bar{n}_2.\end{aligned} \tag{10}$$

We notice that $\bar{n}_1 + \bar{n}_2 + \bar{n}_e = 1$.

Setting $\dot{\bar{n}}_i = 0$ in the rate equation (10), we obtain the steady population of the open quantum system, that is,

$$\frac{\bar{n}_1}{\bar{n}_e} = \frac{\Gamma_{11}^{-}(\varepsilon_1)}{\Gamma_{11}^{+}(\varepsilon_1)}, \qquad \frac{\bar{n}_2}{\bar{n}_e} = \frac{\Gamma_{22}^{-}(\varepsilon_2)}{\Gamma_{22}^{+}(\varepsilon_2)}. \tag{11}$$

We see that no matter whether the environment is in equilibrium, we always obtain a steady state of a diagonal form $\rho_s = \sum P_n|n\rangle\langle n|$, and all the off-diagonal terms of ρ_s vanish. Thus, using the master equation (9) with secular approximation, we always obtain a solution where no quantum coherence is left after a long-time evolution.

Here we make some clarification about the concept of "quantum coherence" we mention in this paper. Intuitively, people usually say there is quantum coherence when there are some nonzero off-diagonal terms in the density matrix, but for any density matrix, we can always make it diagonalized in a certain basis. Thus it seems that the concept of quantum coherence is not free of representation [27]. However, the energy representation is distinctive from others. The canonical thermal state is diagonal only in the energy representation. Therefore, in this paper, when we say there is quantum coherence, we mean that the density matrix ρ of the system has some non-zero off-diagonal terms in the energy representation [12].

3. Steady quantum coherence

In this section, we take into account the interference between the transitions, and thus we do not apply the secular approximation. We show that non-vanishing quantum coherence can exist in the non-equilibrium steady state of the Λ-type system when $T_L \neq T_R$. Then we study the condition for the existence of steady quantum coherence.

3.1. Steady state equation and numerical result

Notice that the master equation (5) without secular approximation is time-independent in Schrödinger picture [21], that is,

$$\begin{aligned}\dot{\rho} = i[\rho,\ \hat{H}_S + \hat{H}_c] &+ \sum_{i,j=1}^{2}[\Gamma_{ji}^{+}(\varepsilon_i) + \Gamma_{ji}^{+}(\varepsilon_j)]\left(\hat{\varphi}_i^{+}\rho\hat{\varphi}_j^{-} - \frac{1}{2}\{\rho,\ \hat{\varphi}_j^{-}\hat{\varphi}_i^{+}\}_{+}\right)\\ &+ [\Gamma_{ij}^{-}(\varepsilon_i) + \Gamma_{ij}^{-}(\varepsilon_j)]\left(\hat{\varphi}_i^{-}\rho\hat{\varphi}_j^{+} - \frac{1}{2}\{\rho,\ \hat{\varphi}_j^{+}\hat{\varphi}_i^{-}\}_{+}\right),\end{aligned} \tag{12}$$

where

$$\hat{H}_c = \frac{1}{2i}\sum_{i,j=1}^{2}[\Gamma_{ji}^{+}(\varepsilon_i) - \Gamma_{ji}^{+}(\varepsilon_j)]\cdot\hat{\varphi}_j^{-}\hat{\varphi}_i^{+} + [\Gamma_{ij}^{-}(\varepsilon_i) - \Gamma_{ij}^{-}(\varepsilon_j)]\cdot\hat{\varphi}_j^{+}\hat{\varphi}_i^{-}$$

can be regarded as the non-diagonal Lamb shift resulted from interference between transitions [14]. Notice that Eq. (12) has a modified Lindblad form [28]. From this master equation, we obtain the dynamics of the population expectations on the three levels of the Λ-type system, $\overline{n}_i := \langle g_i|\rho|g_i\rangle$ and $\overline{n}_e := \langle e|\rho|e\rangle$, as follows,

$$\begin{aligned}
\dot{\overline{n}}_1 &= 2\Gamma_{11}^-(\varepsilon_1)\overline{n}_e - 2\Gamma_{11}^+(\varepsilon_1)\overline{n}_1 - \Gamma_{12}^+(\varepsilon_2)\overline{\tau}_{12} - \Gamma_{21}^+(\varepsilon_2)\overline{\tau}_{21}, \\
\dot{\overline{n}}_2 &= 2\Gamma_{22}^-(\varepsilon_2)\overline{n}_e - 2\Gamma_{22}^+(\varepsilon_2)\overline{n}_2 - \Gamma_{12}^+(\varepsilon_1)\overline{\tau}_{12} - \Gamma_{21}^+(\varepsilon_1)\overline{\tau}_{21}, \\
\dot{\overline{\tau}}_{12} &= [\Gamma_{21}^-(\varepsilon_1)\overline{n}_e - \Gamma_{21}^+(\varepsilon_1)\overline{n}_1] + [\Gamma_{21}^-(\varepsilon_2)\overline{n}_e - \Gamma_{21}^+(\varepsilon_2)\overline{n}_2] \\
&\quad + i\Delta_{12}\overline{\tau}_{12} - [\Gamma_{11}^+(\varepsilon_1) + \Gamma_{22}^+(\varepsilon_2)]\overline{\tau}_{12}.
\end{aligned} \tag{13}$$

Here we denote $\hat{\tau}_{12} := |g_1\rangle\langle g_2|$, and we have $\overline{\tau}_{12} = \langle g_2|\rho|g_1\rangle = \rho_{21}$.

We see that the dynamics of the populations $\overline{n}_i$ and $\overline{n}_g$ are not decoupled from that of the off-diagonal terms $\rho_{12/21}$, which is different from the case with secular approximation Eq. (10) where we obtained a closed system of equations only about the populations. Thus $\rho_{12/21}$ may not be zero even in the steady state after long time evolution. And unlike previous study, this steady quantum coherence is not resulted from the state degeneracy [13].

Setting $\dot{\overline{n}}_i = \dot{\overline{\tau}}_{12} = 0$ in Eq. (13), we obtain the steady state of the open quantum system by solving the linear equations, i.e.,

$$\begin{aligned}
-2\Gamma_{11}^-(\varepsilon_1)\overline{n}_e &= -2\Gamma_{11}^+(\varepsilon_1)\overline{n}_1 - \Gamma_{12}^+(\varepsilon_2)\overline{\tau}_{12} - \Gamma_{21}^+(\varepsilon_2)\overline{\tau}_{21}, \\
-2\Gamma_{22}^-(\varepsilon_2)\overline{n}_e &= -2\Gamma_{22}^+(\varepsilon_2)\overline{n}_2 - \Gamma_{12}^+(\varepsilon_1)\overline{\tau}_{12} - \Gamma_{21}^+(\varepsilon_1)\overline{\tau}_{21}, \\
-[\Gamma_{21}^-(\varepsilon_1) + \Gamma_{21}^-(\varepsilon_2)]\overline{n}_e &= -\Gamma_{21}^+(\varepsilon_1)\overline{n}_1 - \Gamma_{21}^+(\varepsilon_2)\overline{n}_2 - [\Gamma_{11}^+(\varepsilon_1) + \Gamma_{22}^+(\varepsilon_2) - i\Delta_{12}]\overline{\tau}_{12}, \\
-[\Gamma_{12}^-(\varepsilon_1) + \Gamma_{12}^-(\varepsilon_2)]\overline{n}_e &= -\Gamma_{12}^+(\varepsilon_1)\overline{n}_1 - \Gamma_{12}^+(\varepsilon_2)\overline{n}_2 - [\Gamma_{11}^+(\varepsilon_1) + \Gamma_{22}^+(\varepsilon_2) + i\Delta_{12}]\overline{\tau}_{21}.
\end{aligned} \tag{14}$$

This is a set of algebraic linear equations for $\overline{n}_1,\ \overline{n}_2,\ \overline{\tau}_{12},\ \overline{\tau}_{21}$, and the determinant of the coefficient matrix is

$$\begin{aligned}
\det &= [\Gamma_{11}^+(\varepsilon_1) + \Gamma_{22}^+(\varepsilon_2)]^2 \Big[4\Gamma_{11}^+(\varepsilon_1)\Gamma_{22}^+(\varepsilon_2) - 4\Re\mathrm{e}[\Gamma_{12}^+(\varepsilon_2)\Gamma_{21}^+(\varepsilon_1)] + \Delta_{12}^2\Big] \\
&\quad - \Big[2\Im\mathrm{m}[\Gamma_{12}^+(\varepsilon_2)\Gamma_{21}^+(\varepsilon_1)] + \Delta_{12}[\Gamma_{11}^+(\varepsilon_1) - \Gamma_{22}^+(\varepsilon_2)]\Big]^2.
\end{aligned} \tag{15}$$

For the case when the weight factors $f_{L/R}(\omega)$ are real, we have that $\Gamma_{12}^+(\omega) = \Gamma_{21}^+(\omega)$ are also real, and

$$\det = 4[\Gamma_{11}^+(\varepsilon_1) + \Gamma_{22}^+(\varepsilon_2)]^2[\Gamma_{11}^+(\varepsilon_1)\Gamma_{22}^+(\varepsilon_2) - \Gamma_{12}^+(\varepsilon_2)\Gamma_{21}^+(\varepsilon_1)] + 4\Delta_{12}^2\Gamma_{11}^+(\varepsilon_1)\Gamma_{22}^+(\varepsilon_2). \tag{16}$$

If $\det = 0$, the system does not have a unique steady state, and the long time behavior depends on the initial state. For example, if we choose $\Delta_{12} = 0$, $\gamma_{ii}^{(L)}(\varepsilon_1) = \gamma_{ii}^{(L)}(\varepsilon_2) = \gamma_{ii}^{(R)}(\varepsilon_1) = \gamma_{ii}^{(R)}(\varepsilon_2)$ and $f_{L/R}(\varepsilon_i) = 1$ for $i = 1, 2$, we can check from Eq. (16) that we would always get $\det = 0$, no matter how much the temperatures $T_{L/R}$ take. Indeed this is just the case in the study of spontaneous decay-induced coherences [10,13,14]. But this condition is usually too strong for practical physical systems. Especially, here we care more about non-degenerated cases $\Delta_{12} \neq 0$. Indeed, in usual cases $\det = 0$ seldom happen, or it only has several discretely distributed solutions, which have measure 0 in the parameter space of ε_1, ε_2, T_L, T_R, $\gamma_{ij}^{(\alpha)}(\omega)$, unless the spectrums $\gamma_{ij}^{(\alpha)}(\omega)$ have some very novel structures. Therefore, for most practical cases, the steady state is unique and does not depend on the initial state.

We present a numerical result for the steady state in Fig. 3(a), where we set $T_L = T$ and $T_R = T + \Delta T$. The steady solution is unique and does not depend on the initial condition (We checked this numerically for parameters in Fig. 3). We see that $|\rho_{12}| = |\overline{\tau}_{21}|$ does not vanish in the steady state, that is, there exist steady quantum coherence in the non-equilibrium system. And the amplitude of $|\rho_{12}|$ is not a negligibly small value. Here we have set $f_L(\omega) = f_R(\omega) = 1$ to achieve the maximum effect of interference between transitions. If we have $f_\alpha(\omega) < 1$, the steady quantum coherence is suppressed, but still keeps nonzero. This is different from some previous studies about the noise-induced coherence in quantum optical setups, where the weight factor $f_\alpha(\omega)$ (or the p parameter measuring the angle between the transition dipole moments in quantum optics) must take its maximum 1 [10,13].

Moreover, we should notice that when the lower temperature $T_L = T$ is given, the nonzero off-diagonal term $|\rho_{12}|$ increases with the temperature difference ΔT. Besides, when the lower temperatures T_L is quite high or approaches the zero temperature, $|\rho_{12}|$ becomes small but keeps nonzero. We

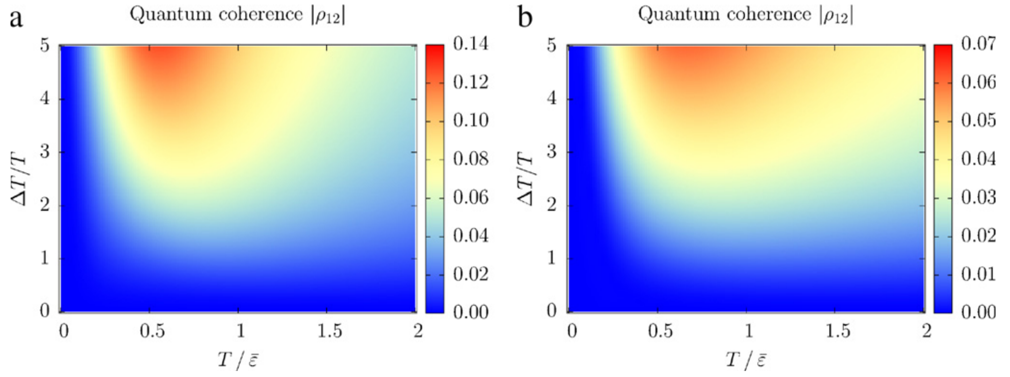

Fig. 3. (Color online) Numerical result for $|\rho_{12}|$ in the non-equilibrium steady state of (a) Λ-type and (b) V-type system. Here we set $T_L = T$ and $T_R = T + \Delta T$. And we set $\bar{\varepsilon} = \frac{1}{2}(\varepsilon_1 + \varepsilon_2) = 1$ as the energy unit. We set the coupling strengths to be $\gamma_{11}^{(L)}(\omega) = 0.01$, $\gamma_{11}^{(R)}(\omega) = 0.02$ and $\gamma_{22}^{(L)}(\omega) = 0.02$, $\gamma_{22}^{(R)}(\omega) = 0.01$. The cross spectrums are set to be real, i.e., $\gamma_{12}^{(\alpha)}(\omega) = \gamma_{21}^{(\alpha)}(\omega) = [f_\alpha(\omega)]^{\frac{1}{2}} \cdot [\gamma_{11}^{(\alpha)}(\omega)\gamma_{22}^{(\alpha)}(\omega)]^{\frac{1}{2}}$ for $\alpha = L, R$, and we set the weight factors as $f_L(\omega) = f_R(\omega) = 1$. For the Λ-type system (a), we set the energy difference as $\Delta_{12} = \varepsilon_1 - \varepsilon_2 = 0.01$, while for the V-type system (b) we set $\Delta_{12} = \varepsilon_1 - \varepsilon_2 = -0.01$.

also need to emphasize that the two states, $|g_1\rangle$ and $|g_2\rangle$, which have steady quantum coherence between them, do not need to be degenerated, which is different from what has been studied in previous literatures [29].

We need to emphasize that the steady quantum coherence we obtained above is not resulted from the "decoherence-free subspace" (DFS) [30–32], and there is no DFS in this model. For a system with DFS, the states in the DFS are decoupled from the environment, and they evolve unitarily. The quantum coherence protected in the DFS is determined by the initial state, and there does not exist a unique steady state in such systems. However, in our model, the steady state, which is obtained from Eq. (13), is unique and does not depend on the initial state, thus there is no DFS here and the steady coherence in this model is not resulted from the DFS.

Now we show that when the temperature difference ΔT decreases to zero exactly, i.e., when we return to case of an equilibrium heat bath with a single temperature, the steady quantum coherence also vanishes completely. In this case, we have $T_L = T_R := T$, and $N_L(\omega) = N_R(\omega) := N(\omega)$. From Eq. (6) we see that the dissipation rates $\Gamma_{ij}^{\pm}(\omega)$ satisfy the following relation of *micro-reversibility* [33,34],

$$\frac{\Gamma_{ij}^{+}(\omega)}{\Gamma_{ij}^{-}(\omega)} = e^{-\omega/T}, \tag{17}$$

which leads to the detailed balance and the equilibrium distribution. With the help of the above relation, we can verify that

$$\frac{\bar{n}_i}{\bar{n}_g} = \frac{\Gamma_{ii}^{+}(\varepsilon_i)}{\Gamma_{ii}^{-}(\varepsilon_i)} = e^{-\varepsilon_i/T}, \qquad \rho_{12} = \rho_{21}^{*} = 0, \tag{18}$$

is exactly the steady solution of Eq. (13). Thus, in the equilibrium case, all the off-diagonal terms vanish to zero after long-time evolution, and this steady solution is same with the result Eq. (11) obtained from the rate equation with secular approximation. That means, the secular approximation is consistent in the case of equilibrium environment.

3.2. Condition for steady quantum coherence

Now we give the condition for the existence of steady quantum coherence. As we will see below, non-equilibrium is a necessary but not sufficient condition for the existence of steady quantum coherence.

To guarantee there is no steady quantum coherence in the steady state $\rho_{12} = \rho_{21}^* = 0$, the necessary condition is

$$\varGamma_{21}^{+}(\varepsilon_1)\left[\frac{\varGamma_{21}^{-}(\varepsilon_1)}{\varGamma_{21}^{+}(\varepsilon_1)} - \frac{\varGamma_{11}^{-}(\varepsilon_1)}{\varGamma_{11}^{+}(\varepsilon_1)}\right] + \varGamma_{21}^{+}(\varepsilon_2)\left[\frac{\varGamma_{21}^{-}(\varepsilon_2)}{\varGamma_{21}^{+}(\varepsilon_2)} - \frac{\varGamma_{22}^{-}(\varepsilon_2)}{\varGamma_{22}^{+}(\varepsilon_2)}\right] = 0. \tag{19}$$

This can be derived directly by putting $\overline{\tau}_{12} = 0$ into Eq. (13). Thus it is the necessary condition to guarantee zero quantum coherence in the steady state. Moreover, if the steady solution is unique [see Eq. (15) and the discussion below], the above condition is also sufficient [we just need to verify that the trial solution $\overline{\tau}_{12} = 0$ and Eq. (11) are consistent with the above condition (19), and the uniqueness of the solution guarantees the sufficiency]. Notice that, as a special case, in the equilibrium case we mentioned above, the dissipation rates automatically satisfy that the two terms in condition (19) both equal to zero, which roots from the micro-reversibility Eq. (17).

Besides the equilibrium case, there is another case that the condition (19) still hold even for non-equilibrium environment. That is, when the coupling spectrums $\gamma_{ij}^{(\alpha)}$ satisfy the following relation

$$\frac{\gamma_{21}^{(L)}(\varepsilon_1)}{\gamma_{21}^{(R)}(\varepsilon_1)} = \frac{\gamma_{11}^{(L)}(\varepsilon_1)}{\gamma_{11}^{(R)}(\varepsilon_1)}, \qquad \frac{\gamma_{21}^{(L)}(\varepsilon_2)}{\gamma_{21}^{(R)}(\varepsilon_2)} = \frac{\gamma_{22}^{(L)}(\varepsilon_2)}{\gamma_{22}^{(R)}(\varepsilon_2)}. \tag{20}$$

It can be verified from Eq. (6) that the above relation also guarantees

$$\frac{\varGamma_{21}^{-}(\varepsilon_1)}{\varGamma_{21}^{+}(\varepsilon_1)} - \frac{\varGamma_{11}^{-}(\varepsilon_1)}{\varGamma_{11}^{+}(\varepsilon_1)} = 0, \quad \frac{\varGamma_{21}^{-}(\varepsilon_2)}{\varGamma_{21}^{+}(\varepsilon_2)} - \frac{\varGamma_{22}^{-}(\varepsilon_2)}{\varGamma_{22}^{+}(\varepsilon_2)} = 0. \tag{21}$$

Thus the condition (19) is satisfied to give rise to vanishing quantum coherence. If the relation (20) is not satisfied, usually we obtain nonzero $\rho_{12/21}$ in the steady state.

Now we discuss the physical meaning of the relation (20). We will show that the relation (20) implies that the two transitions couple to the two heat baths with the same strength proportion.

Remember that we have a relation about the cross spectrum, $|\gamma_{12}^{(\alpha)}(\omega)|^2 = f_\alpha(\omega)\cdot\gamma_{11}^{(\alpha)}(\omega)\gamma_{22}^{(\alpha)}(\omega)$ Eq. (8). Here we consider a simple case that $f_L(\omega) = f_R(\omega) > 0$. In this case, the module square of Eq. (20) gives the following relation,

$$\frac{\gamma_{11}^{(L)}(\varepsilon_1)}{\gamma_{11}^{(R)}(\varepsilon_1)} = \frac{\gamma_{22}^{(L)}(\varepsilon_1)}{\gamma_{22}^{(R)}(\varepsilon_1)}, \qquad \frac{\gamma_{11}^{(L)}(\varepsilon_2)}{\gamma_{11}^{(R)}(\varepsilon_2)} = \frac{\gamma_{22}^{(L)}(\varepsilon_2)}{\gamma_{22}^{(R)}(\varepsilon_2)}. \tag{22}$$

That means, for the two transitions, their coupling strengths with the two heat baths must be of the same proportion, so as to guarantee vanishing quantum coherence.

Here we give an example for the spectrums that the above relation (22) is broken. As demonstrated in Fig. 4(a), (b), transition-1 couples to bath-R more strongly than bath-L [$\gamma_{11}^{(L)}(\varepsilon_1) < \gamma_{11}^{(R)}(\varepsilon_1)$], but on contrary transition-2 couples to bath-L more strongly than bath-R [$\gamma_{22}^{(L)}(\varepsilon_2) > \gamma_{22}^{(R)}(\varepsilon_2)$]. We can check that the above strength proportion relation Eq. (22) is violated, and so is the condition (19). In this case, there exists non-vanishing steady quantum coherence between the two lower states $|g_{1/2}\rangle$. In the above numerical result [Fig. 3(a)], the coupling strengths $\gamma_{ij}^{(\alpha)}(\varepsilon_i)$ are chosen just in this way. Such configuration can be realized by imposing coupling spectrums of proper shapes as shown in Fig. 4(c).

4. V-type and $\varXi$-type systems

We have shown that for a $\varLambda$-type system in a non-equilibrium environment, the interference between transitions can give rise to non-vanishing steady quantum coherence. In this section, we consider the case of V-type and $\varXi$-type systems.

We should notice that the master equation (12) we derived for a $\varLambda$-type system is also valid for V-type and $\varXi$-type systems, as long as we properly redefine the raising and lowering operators $\hat{\varphi}_i^{\pm}$ for the two transitions.

For a V-type system, we define $\hat{\varphi}_i^{-} := |g\rangle\langle e_i|$ and $\hat{\varphi}_i^{+} := |e_i\rangle\langle g|$ [see the notations in Fig. 2(b)]. With these operators for the transitions, we can obtain a master equation having the same form with

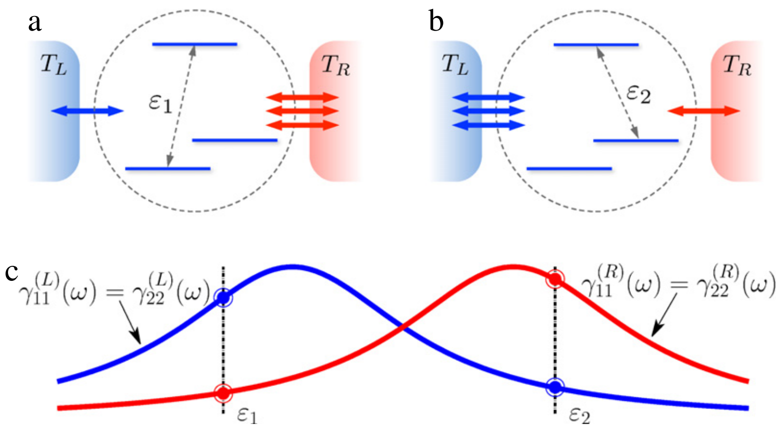

Fig. 4. (Color online) (a) Transition-1 couples more strongly to bath-R, $\gamma_{11}^{(L)}(\varepsilon_1) < \gamma_{11}^{(R)}(\varepsilon_1)$. (b) Transition-2 couples more strongly to bath-L, $\gamma_{22}^{(L)}(\varepsilon_2) > \gamma_{22}^{(R)}(\varepsilon_2)$. (c) An example of spectrums $\gamma_{ii}^{(\alpha)}(\omega)$ to support such configurations of (a) and (b).

Eq. (12). The steady state equation for the V-type system is

$$\begin{aligned}\dot{\bar{n}}_1 &= 2\Gamma_{11}^{+}(\varepsilon_1)\bar{n}_g - 2\Gamma_{11}^{-}(\varepsilon_1)\bar{n}_1 - \Gamma_{12}^{-}(\varepsilon_2)\bar{\tau}_{12} - \Gamma_{21}^{-}(\varepsilon_2)\bar{\tau}_{21},\\ \dot{\bar{n}}_2 &= 2\Gamma_{22}^{+}(\varepsilon_2)\bar{n}_g - 2\Gamma_{22}^{-}(\varepsilon_2)\bar{n}_2 - \Gamma_{12}^{-}(\varepsilon_1)\bar{\tau}_{12} - \Gamma_{21}^{-}(\varepsilon_1)\bar{\tau}_{21},\\ \dot{\bar{\tau}}_{12} &= [\Gamma_{21}^{+}(\varepsilon_1)\bar{n}_g - \Gamma_{21}^{-}(\varepsilon_1)\bar{n}_1] + [\Gamma_{21}^{+}(\varepsilon_2)\bar{n}_g - \Gamma_{21}^{-}(\varepsilon_2)\bar{n}_2]\\ &\quad + i\Delta_{12}\bar{\tau}_{12} - [\Gamma_{11}^{-}(\varepsilon_1) + \Gamma_{22}^{-}(\varepsilon_2)]\bar{\tau}_{12}.\end{aligned} \tag{23}$$

Here we denote $\bar{n}_i := \langle e_i|\rho|e_i\rangle$, $\bar{n}_g := \langle g|\rho|g\rangle$, $\hat{\tau}_{12} := |e_1\rangle\langle e_2|$ and $\bar{\tau}_{12} = \langle e_2|\rho|e_1\rangle$. We see again that the populations $\bar{n}_i$ and $\bar{n}_e$ also depend on the value of off-diagonal terms ρ_{12} and ρ_{21}. As we discussed above, in the steady state, we can also obtain nonzero quantum coherence in the steady state. We show the numerical result of the steady quantum coherence in the V-type system in Fig. 3(b). Notice that the maximum value of $|\rho_{12}|$ in the V-type system is much smaller than that in the Λ-type system [see Fig. 3(a)], because the steady quantum coherence in the V-type system exists between excited energy levels, which possess much less populations.

For the Ξ-type system, we define $\hat{\varphi}_1^{+} := |e_1\rangle\langle g|$, $\hat{\varphi}_2^{+} := |e_2\rangle\langle e_1|$, and $\hat{\varphi}_i^{-} = [\hat{\varphi}_i^{+}]^{\dagger}$ for the two transitions [see the notations in Fig. 2(c)]. The master equation of the Ξ-type system still has the form of Eq. (12), while the steady state equation is

$$\begin{aligned}\dot{\bar{n}}_1 &= 2[\Gamma_{11}^{+}(\varepsilon_1)\bar{n}_g - \Gamma_{11}^{-}(\varepsilon_1)\bar{n}_1],\\ \dot{\bar{n}}_2 &= 2[\Gamma_{22}^{+}(\varepsilon_2)\bar{n}_1 - \Gamma_{22}^{-}(\varepsilon_2)\bar{n}_2].\end{aligned} \tag{24}$$

Here we denote $\bar{n}_i := \langle e_i|\rho|e_i\rangle$ and $\bar{n}_g := \langle g|\rho|g\rangle$. Different from the case of V-type and Λ-type systems, the populations alone form a closed set of equations and do not depend on any off-diagonal terms. More precisely speaking, the interference between transitions do not affect the time evolution of the populations. Thus, for the Ξ-type system, there is always no quantum coherence left in the steady state, and this result is consistent with secular approximation [Eq. (11)].

5. Physical meaning of quantum coherence

In this section, we demonstrate an example to study the physical meaning of the quantum coherence. In this example, we will see that the quantum coherence between the energy eigenstates exactly reflects the non-equilibrium flux inside the composite system [21].

We consider a composite system of two coupled two-level-systems (TLSs), which is described by

$$\hat{H}_S = \frac{1}{2}\omega_1\hat{\sigma}_1^z + \frac{1}{2}\omega_2\hat{\sigma}_2^z + g(\hat{\sigma}_1^{+}\hat{\sigma}_2^{-} + \hat{\sigma}_1^{-}\hat{\sigma}_2^{+}), \tag{25}$$

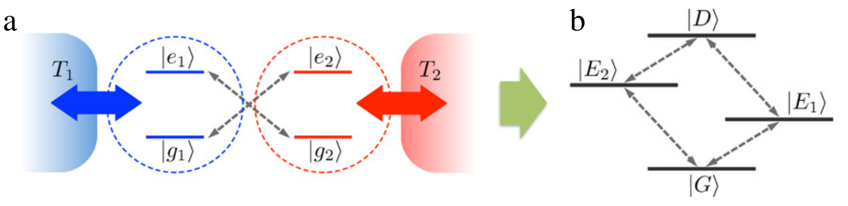

Fig. 5. (Color online) (a) Two coupled TLSs. The two TLSs exchange energy with each other. We denote the bare states of each TLS by $|e_i\rangle$, $|g_i\rangle$, and the eigenstates of the composite system are $|G\rangle$, $|E_i\rangle$, $|D\rangle$. The interaction with the environment gives rise to transitions between the four eigenstates, as shown by arrows in (b), where $|E_2\rangle \leftrightarrow |G\rangle \leftrightarrow |E_1\rangle$ can be regarded as a V-type structure, and $|E_2\rangle \leftrightarrow |D\rangle \leftrightarrow |E_1\rangle$ can be regarded as a Λ-type structure.

where $\hat{\sigma}_i^+ := |e_i\rangle\langle g_i|$, $\hat{\sigma}_i^- := |g_i\rangle\langle e_i|$, $\hat{\sigma}_i^z := |e_i\rangle\langle e_i| - |g_i\rangle\langle g_i|$, and $|e_{1,2}\rangle$, $|g_{1,2}\rangle$ are bare states of each TLS [see Fig. 5(a)]. And this Hamiltonian can be diagonalized as $\hat{H}_S = \sum_n E_n|E_n\rangle\langle E_n|$. The eigenenergies and the corresponding eigenstates are [35]

$$\begin{aligned}
E_G &= -\frac{1}{2}\overline{\Omega}, & |G\rangle &= |g_1g_2\rangle, \\
E_1 &= -\frac{1}{2}\sqrt{\Delta^2+4g^2}, & |E_1\rangle &= \sin\frac{\theta}{2}|e_1g_2\rangle - \cos\frac{\theta}{2}|g_1e_2\rangle, \\
E_2 &= \frac{1}{2}\sqrt{\Delta^2+4g^2}, & |E_2\rangle &= \cos\frac{\theta}{2}|e_1g_2\rangle + \sin\frac{\theta}{2}|g_1e_2\rangle, \\
E_D &= \frac{1}{2}\overline{\Omega}, & |D\rangle &= |e_1e_2\rangle,
\end{aligned} \tag{26}$$

where $\overline{\Omega} := (\omega_1+\omega_2)/2$, $\Delta := \omega_1 - \omega_2$ and $\cot\theta = \Delta/2g$.

Each TLS couples with an independent heat bath via interaction $\hat{H}_{SB} = \hat{H}_{SB}^{(1)} + \hat{H}_{SB}^{(2)}$, where

$$\hat{H}_{SB}^{(\alpha)} = \sum_{k_\alpha} g_{k_\alpha}\,\hat{\sigma}_\alpha^+\hat{b}_{k_\alpha} + g_{k_\alpha}^*\,\hat{\sigma}_\alpha^-\hat{b}_{k_\alpha}^\dagger. \tag{27}$$

Such interaction gives rise to a transition structure as shown in Fig. 5(b) [4,35]. We can regard the transition $|E_2\rangle \leftrightarrow |G\rangle \leftrightarrow |E_1\rangle$ as a V-type structure, and $|E_2\rangle \leftrightarrow |D\rangle \leftrightarrow |E_1\rangle$ as a Λ-type structure, and there exists quantum interference between the transitions. Such a non-equilibrium system can be realized in present experiments by, for example, interacting superconducting qubits [36], or double quantum dots [37,38], etc.

Now we consider the non-equilibrium flux flowing across, for example, the TLS-1. The dynamics of the population of TLS-1 $\overline{n}_1 := \langle e_1|\rho|e_1\rangle$ can be obtained by the Heisenberg equation,

$$\begin{aligned}
\dot{\overline{n}}_1 &= \frac{d}{dt}\langle\hat{\sigma}_1^z\rangle = -i\langle[\hat{\sigma}_1^z,\ \hat{H}_S + \hat{H}_{SB} + \hat{H}_B]\rangle \\
&= -i\langle[\hat{\sigma}_1^z,\ \hat{H}_S]\rangle - i\langle[\hat{\sigma}_1^z,\ \hat{H}_{SB}]\rangle := J_{1-2} + J_{1-B_1},
\end{aligned} \tag{28}$$

where $J_{1-2} := -i\langle[\hat{\sigma}_1^z,\ \hat{H}_S]\rangle$ means the internal flux between the two TLSs, while J_{1-B_1} means the flux flowing between TLS-1 and bath-1. We have

$$\begin{aligned}
J_{1-2} &= -i2g\langle\hat{\sigma}_1^+\hat{\sigma}_2^- - \hat{\sigma}_1^-\hat{\sigma}_2^+\rangle = -i2g\mathrm{Tr}\Big[\rho(|e_1g_2\rangle\langle g_1e_2| - |g_1e_2\rangle\langle e_1g_2|)\Big] \\
&= -i2g\mathrm{Tr}\Big[\rho(|E_2\rangle\langle E_1| - |E_1\rangle\langle E_2|)\Big] = 4g\Im\mathrm{m}\langle E_1|\rho|E_2\rangle.
\end{aligned}$$

The above calculation is completed with the help of Eq. (26). The above equation clearly shows that the non-equilibrium flux between the two TLSs J_{1-2} is exactly reflected by the imaginary part of the quantum coherence term $\langle E_1|\rho|E_2\rangle$ [21]. If the two heat baths have the same temperature, there is no net heat transfer, and the steady quantum coherence automatically vanishes, which is just consistent with what we have discussed above.

Therefore, the quantum coherence exactly reflects the non-equilibrium flux inside a composite system. If the steady quantum coherence is missed, the internal flux inside the composite system would also be omitted improperly, which leads to an unphysical conclusion that there is no net flux between the local sites even when the two heat baths have different temperatures.

6. Conclusion

In this paper, we studied the steady state of a three-level system in a non-equilibrium environment, which consists of two heat baths with different temperatures. We find that for the Λ-type and V-type systems, the interference between transitions can give rise to non-vanishing steady quantum coherence, if the two transitions couple to the two heat baths with different proportions of coupling strengths. And the amount of the steady quantum coherence increases with the temperature difference of the two heat baths. If the two heat baths have the same temperature, all the quantum coherence vanishes and returns to the equilibrium case. These transition structures are quite common in natural and artificial quantum systems. The non-equilibrium environment can be implemented via current noises with different effective temperatures in quantum circuits [36,39], or electron leads with different chemical potentials [37].

The interference between transitions play an essential role in the steady quantum coherence. But it was often omitted by secular approximation in previous literatures. We showed that indeed the secular approximation is consistent in the case of equilibrium environment, but for non-equilibrium environments, that would lead to the neglect of the steady quantum coherence.

We also show that the quantum coherence has a clear physical meaning, i.e., it exactly reflects the internal non-equilibrium flux inside a composite system, which is an important characterization of non-equilibrium systems.

Notice that many current investigations about non-equilibrium quantum thermodynamics are based on the rate equation of the energy level populations like Eq. (10), which does not include the quantum coherence. Our result implies that some further refinement, which takes into account the quantum coherence, should be made to the present study of non-equilibrium quantum thermodynamics [38].

Acknowledgments

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Appendix A. Derivation of the master equation

We show some detailed calculation of the derivation for the master equation. Denoting $\hat{\mathsf{B}}_i = \hat{B}_{i,L} + \hat{B}_{i,R}$, we rewrite the interaction Hamiltonian Eq. (3) as $\hat{H}_{SB} := \sum_i \hat{\varphi}_i^+ \cdot \hat{\mathsf{B}}_i + \hat{\varphi}_i^- \cdot \hat{\mathsf{B}}_i^\dagger$. In the interaction picture of $\hat{H}_S + \hat{H}_B$, we have

$$\hat{H}_{SB}(t) = \sum_i \hat{\varphi}_i^+(t) \cdot \hat{\mathsf{B}}_i(t) + \hat{\varphi}_i^-(t) \cdot \hat{\mathsf{B}}_i^\dagger(t) := V^+(t) + V^-(t). \tag{A.1}$$

Put it into Eq. (4) of $\dot{\rho}(t)$, and we have

$$\dot{\rho} = -\int_0^\infty ds\, \mathrm{Tr}_B \Big\{ \big[\hat{V}^+(t),\ [\hat{V}^-(t-s),\ \rho(t) \otimes \rho_B]\big] + \big[\hat{V}^-(t),\ [\hat{V}^+(t-s),\ \rho(t) \otimes \rho_B]\big] \Big\}. \tag{A.2}$$

The above equation is expanded as follows

$$\begin{aligned}\dot{\rho} = -\int_0^\infty ds \sum_{i,j=1}^{2} \Big\{ & e^{i(\varepsilon_i-\varepsilon_j)t}\cdot \hat{\varphi}_i^+\hat{\varphi}_j^-\rho\cdot e^{i\varepsilon_j s}\langle \hat{\mathsf{B}}_i(t)\hat{\mathsf{B}}_j^\dagger(t-s)\rangle \\ & - e^{i(\varepsilon_i-\varepsilon_j)t}\cdot \hat{\varphi}_i^+\rho\hat{\varphi}_j^-\cdot e^{i\varepsilon_j s}\langle \hat{\mathsf{B}}_j^\dagger(t-s)\hat{\mathsf{B}}_i(t)\rangle \\ & - e^{i(\varepsilon_i-\varepsilon_j)t}\cdot \hat{\varphi}_j^-\rho\hat{\varphi}_i^+\cdot e^{i\varepsilon_j s}\langle \hat{\mathsf{B}}_i(t)\hat{\mathsf{B}}_j^\dagger(t-s)\rangle + e^{i(\varepsilon_i-\varepsilon_j)t}\cdot \rho\hat{\varphi}_j^-\hat{\varphi}_i^+\cdot e^{i\varepsilon_j s}\langle \hat{\mathsf{B}}_j^\dagger(t-s)\hat{\mathsf{B}}_i(t)\rangle \\ & + e^{-i(\varepsilon_i-\varepsilon_j)t}\cdot \hat{\varphi}_i^-\hat{\varphi}_j^+\rho\cdot e^{-i\varepsilon_j s}\langle \hat{\mathsf{B}}_i^\dagger(t)\hat{\mathsf{B}}_j(t-s)\rangle - e^{-i(\varepsilon_i-\varepsilon_j)t}\cdot \hat{\varphi}_i^-\rho\hat{\varphi}_j^+\cdot e^{-i\varepsilon_j s}\langle \hat{\mathsf{B}}_j(t-s)\hat{\mathsf{B}}_i^\dagger(t)\rangle \\ & - e^{-i(\varepsilon_i-\varepsilon_j)t}\cdot \hat{\varphi}_j^+\rho\hat{\varphi}_i^-\cdot e^{-i\varepsilon_j s}\langle \hat{\mathsf{B}}_i^\dagger(t)\hat{\mathsf{B}}_j(t-s)\rangle + e^{-i(\varepsilon_i-\varepsilon_j)t}\cdot \rho\hat{\varphi}_j^+\hat{\varphi}_i^-\cdot e^{-i\varepsilon_j s}\langle \hat{\mathsf{B}}_j(t-s)\hat{\mathsf{B}}_i^\dagger(t)\rangle \Big\}.\end{aligned}$$

We apply the Born approximation that the two heat bath always stay at their canonical thermal state with temperature T_α respectively. Here we show the calculation of two terms as demonstration,

$$\begin{aligned}\int_0^\infty ds\, e^{i\varepsilon s}\langle \hat{\mathsf{B}}_i(t)\hat{\mathsf{B}}_j^\dagger(t-s)\rangle &= \int_0^\infty ds\, e^{i\varepsilon s}\Big(\langle \hat{B}_{i,L}(t)\hat{B}_{j,L}^\dagger(t-s)\rangle + \langle \hat{B}_{i,R}(t)\hat{B}_{j,R}^\dagger(t-s)\rangle\Big) \\ &= \int_0^\infty ds \int \frac{d\omega}{2\pi}\, e^{i(\varepsilon-\omega)s}\Big(\gamma_{ji}^{(L)}(\omega)[N_L(\omega)+1] + \gamma_{ji}^{(R)}(\omega)[N_R(\omega)+1]\Big) \\ &= \frac{1}{2}\gamma_{ji}^{(L)}(\varepsilon)[N_L(\varepsilon)+1] + \frac{1}{2}\gamma_{ji}^{(R)}(\varepsilon)[N_R(\varepsilon)+1] := \varGamma_{ji}^-(\varepsilon),\end{aligned} \tag{A.3}$$

$$\begin{aligned}\int_0^\infty ds\, e^{i\varepsilon s}\langle \hat{\mathsf{B}}_i^\dagger(t)\hat{\mathsf{B}}_j(t-s)\rangle &= \int_0^\infty ds\, e^{i\varepsilon s}\Big(\langle \hat{B}_{i,L}^\dagger(t)\hat{B}_{j,L}(t-s)\rangle + \langle \hat{B}_{i,R}^\dagger(t)\hat{B}_{j,R}(t-s)\rangle\Big) \\ &= \int_0^\infty ds \int \frac{d\omega}{2\pi}\, e^{i(\varepsilon-\omega)s}\Big(\gamma_{ij}^{(L)}(\omega)N_L(\omega) + \gamma_{ij}^{(R)}(\omega)N_R(\omega)\Big) \\ &= \frac{1}{2}\gamma_{ij}^{(L)}(\varepsilon)N_L(\varepsilon) + \frac{1}{2}\gamma_{ij}^{(R)}(\varepsilon)N_R(\varepsilon) := \varGamma_{ij}^+(\varepsilon),\end{aligned} \tag{A.4}$$

where we define the coupling spectrum with bath-α as $\gamma_{ij}^{(\alpha)}(\omega) := 2\pi\sum g_{i,k_\alpha}^* g_{j,k_\alpha}\delta(\omega-\omega_{k_\alpha}) = [\gamma_{ji}^{(\alpha)}(\omega)]^*$. Here we utilized the formula

$$\int_0^\infty ds\, e^{i(\varepsilon-\omega)s} = \pi\delta(\varepsilon-\omega) + i\mathbf{P}\frac{1}{\varepsilon-\omega}, \tag{A.5}$$

and omitted the principle integral terms. Then we obtain the master equation as

$$\begin{aligned}\dot{\rho} = -\sum_{i,j=1}^{2}\Big\{ & \varGamma_{ji}^-(\varepsilon_j)e^{i\Delta_{ij}t}\hat{\varphi}_i^+\hat{\varphi}_j^-\rho - \varGamma_{ji}^+(\varepsilon_j)e^{i\Delta_{ij}t}\hat{\varphi}_i^+\rho\hat{\varphi}_j^- - \varGamma_{ji}^-(\varepsilon_j)e^{i\Delta_{ij}t}\hat{\varphi}_j^-\rho\hat{\varphi}_i^+ \\ & + \varGamma_{ji}^+(\varepsilon_j)e^{i\Delta_{ij}t}\rho\hat{\varphi}_j^-\hat{\varphi}_i^+ + \varGamma_{ij}^+(\varepsilon_j)e^{-i\Delta_{ij}t}\hat{\varphi}_i^-\hat{\varphi}_j^+\rho - \varGamma_{ij}^-(\varepsilon_j)e^{-i\Delta_{ij}t}\hat{\varphi}_i^-\rho\hat{\varphi}_j^+ \\ & - \varGamma_{ij}^+(\varepsilon_j)e^{-i\Delta_{ij}t}\hat{\varphi}_j^+\rho\hat{\varphi}_i^- + \varGamma_{ij}^-(\varepsilon_j)e^{-i\Delta_{ij}t}\rho\hat{\varphi}_j^+\hat{\varphi}_i^-\Big\},\end{aligned}$$

where $\Delta_{ij} := \varepsilon_i - \varepsilon_j$. In Schrödinger picture, we can write down the master equation in the following time-independent Lindblad-like form,

$$\begin{aligned}\dot{\rho} = i[\rho,\ \hat{H}_S] + \sum_{i,j=1}^{2}\Big\{ & \varGamma_{ji}^+(\varepsilon_j)\cdot\big[\hat{\varphi}_i^+,\ \rho\hat{\varphi}_j^-\big] + \varGamma_{ji}^+(\varepsilon_i)\cdot\big[\hat{\varphi}_i^+\rho,\ \hat{\varphi}_j^-\big] \\ & + \varGamma_{ij}^-(\varepsilon_j)\cdot\big[\hat{\varphi}_i^-,\ \rho\hat{\varphi}_j^+\big] + \varGamma_{ij}^-(\varepsilon_i)\cdot\big[\hat{\varphi}_i^-\rho,\ \hat{\varphi}_j^+\big]\Big\},\end{aligned} \tag{A.6}$$

or equivalently,

$$\dot{\rho} = i[\rho,\ \hat{H}_S + \hat{H}_c] + \sum_{i,j=1}^{2}[\Gamma_{ji}^{+}(\varepsilon_i) + \Gamma_{ji}^{+}(\varepsilon_j)] \cdot \left(\hat{\varphi}_i^{+}\rho\hat{\varphi}_j^{-} - \frac{1}{2}\{\rho,\ \hat{\varphi}_j^{-}\hat{\varphi}_i^{+}\}_{+}\right)$$
$$+ [\Gamma_{ij}^{-}(\varepsilon_i) + \Gamma_{ij}^{-}(\varepsilon_j)] \cdot \left(\hat{\varphi}_i^{-}\rho\hat{\varphi}_j^{+} - \frac{1}{2}\{\rho,\ \hat{\varphi}_j^{+}\hat{\varphi}_i^{-}\}_{+}\right), \tag{A.7}$$

where

$$\hat{H}_c = \sum_{i,j=1}^{2} \frac{\Gamma_{ji}^{+}(\varepsilon_i) - \Gamma_{ji}^{+}(\varepsilon_j)}{2i}\hat{\varphi}_j^{-}\hat{\varphi}_i^{+} + \frac{\Gamma_{ij}^{-}(\varepsilon_i) - \Gamma_{ij}^{-}(\varepsilon_j)}{2i}\hat{\varphi}_j^{+}\hat{\varphi}_i^{-} \tag{A.8}$$

can be regarded as the non-diagonal Lamb shift resulted from interference between transitions [14].

Appendix B. Cross spectrum

Here we derive a relation of the cross spectrum. When we have two transitions contacting with the same environment, we need three coupling spectrums, i.e., two individual spectrum for each transition, $\gamma_{ii}(\omega)$, and another cross spectrum [22] for the cross transition, $\gamma_{12}(\omega)$. These spectrums are defined as

$$\gamma_{ii}(\omega) := 2\pi \sum_{k} \left|g_{i,k}\right|^2 \delta(\omega - \omega_k),$$
$$\gamma_{12}(\omega) := 2\pi \sum_{k} g_{1,k}^{*}\, g_{2,k}\, \delta(\omega - \omega_k). \tag{B.1}$$

From this definition, we have

$$\gamma_{11}(\omega)\gamma_{22}(\omega) = 4\pi^2 \sum_{k,q} g_{1,k}^{*}g_{1,k}\delta(\omega - \omega_k) \cdot g_{2,q}^{*}g_{2,q}\delta(\omega - \omega_q),$$
$$|\gamma_{12}(\omega)|^2 = 4\pi^2 \sum_{k,q} g_{1,k}^{*}g_{2,k}\delta(\omega - \omega_k) \cdot g_{1,q}g_{2,q}^{*}\delta(\omega - \omega_q).$$

From $|g_{1,k}g_{2,q} - g_{2,k}g_{1,q}|^2 \geq 0$, we have

$$g_{1,k}^{*}g_{1,k} \cdot g_{2,q}^{*}g_{2,q} + g_{1,q}^{*}g_{1,q} \cdot g_{2,k}^{*}g_{2,k} \geq g_{1,k}^{*}g_{2,k} \cdot g_{1,q}g_{2,q}^{*} + g_{1,k}g_{2,k}^{*} \cdot g_{2,q}^{*}g_{2,q}.$$

Thus, we have

$$\sum_{k,q}(g_{1,k}^{*}g_{1,k} \cdot g_{2,q}^{*}g_{2,q} + g_{1,q}^{*}g_{1,q} \cdot g_{2,k}^{*}g_{2,k}) \cdot \delta(\omega - \omega_k)\delta(\omega - \omega_q)$$
$$\geq \sum_{k,q}(g_{1,k}^{*}g_{2,k} \cdot g_{1,q}g_{2,q}^{*} + g_{1,k}g_{2,k}^{*} \cdot g_{2,q}^{*}g_{2,q}) \cdot \delta(\omega - \omega_k)\delta(\omega - \omega_q).$$

That is

$$\gamma_{11}(\omega)\gamma_{22}(\omega) \geq |\gamma_{12}(\omega)|^2. \tag{B.2}$$

From the above proof we see that the equality holds if and only if we have

$$g_{1,k} \cdot g_{2,q} = g_{1,q} \cdot g_{2,k}, \tag{B.3}$$

for any $k,\ q$ which satisfy $\omega_k = \omega_q$.

If $k \to \omega_k$ is a one-to-one map, we have $\omega_k = \omega_q \Leftrightarrow k = q$. Then the above relation holds and further we have

$$\gamma_{11}(\omega)\gamma_{22}(\omega) = |\gamma_{12}(\omega)|^2. \tag{B.4}$$

In more general cases, for example, the environment is the electromagnetic field or the phonon field, with the index $\mathbf{k}$ as a vector, the mode energy $\omega_{\mathbf{k}}$ has degeneracy in different directions of $\mathbf{k}$. If the amplitudes $|\mathbf{k}|$ are the same, the modes with different directions have the same energy $\omega_{\mathbf{k}}$. In these cases, if the coupling coefficients $g_{i,\mathbf{k}} = g_{i,|\mathbf{k}|}$ only depend on the amplitude $|\mathbf{k}|$, i.e., only depend on $\omega_{\mathbf{k}}$ equivalently, the relation (B.3) still holds, and we still have $\gamma_{11}(\omega)\gamma_{22}(\omega) = |\gamma_{12}(\omega)|^2$, as used in many literatures [35].

Except the above two cases, the equality (B.4) usually does not hold, and the general relation between the cross spectrum $\gamma_{12}(\omega)$ and the individual spectrums $\gamma_{ii}(\omega)$ appear as [14,23,24]

$$|\gamma_{12}(\omega)|^2 = f(\omega) \cdot \gamma_{11}(\omega)\gamma_{22}(\omega), \tag{B.5}$$

where $0 \le f(\omega) \le 1$ is a weight factor. In general cases, the concrete form of $f(\omega)$ depends on the form of coupling strength $g_{i,\mathbf{k}}$ in specific physical systems.

For example, two two-level atoms stay at position $\mathbf{r}_1$ and $\mathbf{r}_2$ in the same electromagnetic field, and they separate from each other for a distance $|\mathbf{r}_1 - \mathbf{r}_2| := d$. Their coupling strengths with the electromagnetic field have a relation $g_{1,\mathbf{k}} = g_{2,\mathbf{k}} \exp[i\mathbf{k} \cdot (\mathbf{r}_2 - \mathbf{r}_1)]$. The light emitted from the two atoms can interfere with each other. In this case, we can check that the weight factor has the form of $f(\omega d/c)$. It varies with the distance d, and depends on the dimensionality D of the electromagnetic field, i.e.,

$$f(x) = \begin{cases} \cos^2(x), & D = 1, \\ [J_0(x)]^2, & D = 2, \\ \mathrm{sinc}^2(x), & D = 3, \end{cases} \tag{B.6}$$

where $J_0(x)$ is the Bessel function of the first kind [23]. When the two atoms are quite near to each other, their interference effect achieve the maximum. When they are far from each other, their interference effect quickly decays with the distance (for $D = 2,\ 3$). Thus the weight function $f(\omega d/c)$ here describe the spatial correlation of the environment. The situation is similar when we consider the phonon bath [24].

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