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Quantum-Coherence Effect in a Quantum Dot System Coupled by Optical Near Fields

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Dynamics of the exciton population in a quantum dot system coupled via optical near fields is theoretically investigated. We derive analytical solutions of the density matrix elements for one-exciton and two-exciton states. Based on the results, the transient response from a steady state induced by an applied light pulse is obtained as approximated solutions. The results show fast increase of the population due to the state filling of a sub-level and the oscillation behavior, which originate from coherence effects inherent in such a system. Moreover, slow population decay appears in the nanosecond regime, which is related to the effect of state filling and radiative relaxation. These characteristics well explain recent experimental data of time-resolved near-field spectroscopy, and clarify physics behind the phenomenon.
Key words: coupled quantum dot, optical near fields, coherence, population dynamics, relaxation

1. INTRODUCTION

Optical near fields provide a powerful tool to control the states of nanometric matter as well as to observe nanometric structures. The localization property of the optical near fields, which is free from the diffraction limit of light, allows selective access to nanometric objects. This induces interesting and inherent effects, such as non-radiative energy transfer between optically (dipolar) forbidden levels [1]. We previously estimated energy transfer time via optical near-field interaction, which is less than 100 ps [2]. Such characteristic features inherent in the optical near fields open up a way to realize a novel type of nanophotonic functional devices.

Since unidirectional energy transfer is an indispensable character for functional devices, we have considered a quantum dot system with intra-sublevel relaxation due to exciton-phonon interaction, where a few quantum dots are coupled via optical near-field interaction. In this paper, we assume a two-quantum dot system; one is two-level and the other is three-level. We theoretically derive the dynamics of exciton population in this system, and compare our results to data obtained in a recent time-resolved experiment [3]. Here, the key point is to control the exciton-population dynamics by applying a light pulse. In other word, transient dynamics from one-exciton states to two-exciton states determines the characteristics of device control.

In the case of resonant two-level quantum dots, the optical near-field interaction induces nutation of the population, or coherent oscillation, which is analogous to the Förster process describing molecular excitation [4]. We theoretically show that fluctuations of luminescence intensity in the experimental results are related to the coherent oscillation between resonant energy levels. Moreover the quantum coherence can realize fast energy transfer in the system. As another aspect, we demonstrate slower population decay that is a characteristic effect in two-exciton states.

This paper is organized as follows. Section 2 is devoted to describing a theoretical model and equations of motion of the population in a coupled quantum dot system with analytical solutions for typical initial conditions. In Sec. 3, transient variation from one-exciton states to two-exciton states is investigated analytically, by comparing to the experimental results. Finally, concluding remarks are given in Sec. 4.

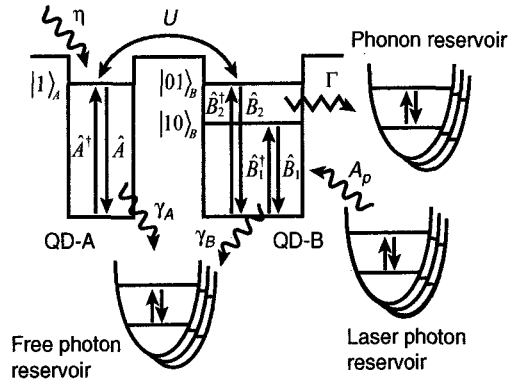

Fig. 1: A model of coupled quantum dot system. The reservoirs for phonons and free photons are introduced in order to represent the dissipation processes.

2. MODEL OF COUPLED QUANTUM DOT SYSTEM

2.1 Equations of motion

In order to evaluate luminescence intensity, we incorporate some reservoir systems as illustrated in Fig. 1. The energy levels $|1\rangle_A$ and $|10\rangle_B$ are coupled to a free photon reservoir, while the level $|01\rangle_B$ is not because the transition with even total quantum number is dipole forbidden by assuming cubic quantum dots. Temporal evolution of the density operator in this system can be described on the basis of the Born-Markov approximation [5] as

$$\begin{aligned}\dot{\hat{\rho}}(t) = &-\frac{i}{\hbar}[H_0 + H_{NF}, \hat{\rho}(t)] \\ &-\frac{\Gamma}{2}\left[\hat{B}_2^\dagger \hat{B}_1 \hat{B}_1^\dagger \hat{B}_2 \hat{\rho}(t) + \hat{\rho}(t)\hat{B}_2^\dagger \hat{B}_1 \hat{B}_1^\dagger \hat{B}_2 \right. \\ &\left. -2\hat{B}_1^\dagger \hat{B}_2 \hat{\rho}(t) \hat{B}_2^\dagger \hat{B}_1\right] \\ &-\frac{\gamma_A}{2}\left[\hat{A}^\dagger \hat{A}\hat{\rho}(t) + \hat{\rho}(t)\hat{A}^\dagger \hat{A} - 2\hat{A}\hat{\rho}(t)\hat{A}^\dagger\right] \\ &-\frac{\gamma_B}{2}\left[\hat{B}_1^\dagger \hat{B}_1 \hat{\rho}(t) + \hat{\rho}(t)\hat{B}_1^\dagger \hat{B}_1 - 2\hat{B}_1 \hat{\rho}(t)\hat{B}_1^\dagger\right],\end{aligned} \tag{1}$$

where

$$\begin{aligned}\hat{H}_0 &= E_1 \hat{A}^\dagger \hat{A} + E_1 \hat{B}_2^\dagger \hat{B}_2 + E_0 \hat{B}_1^\dagger \hat{B}_1, \\ \hat{H}_{NF} &= \hbar U\left(\hat{A}^\dagger \hat{B}_2 + \hat{B}_2^\dagger \hat{A}\right),\end{aligned} \tag{2}$$

where Γ, γ_A, and γ_B represent the relaxation constants of intra-sublevel transition via exciton-phonon coupling, and radiative transition in QD-A and B, respectively, which appear after tracing over both phonon and photon degrees of freedom. The strength of the optical near-field interaction is denoted as U. We have already derived the optical near-field interaction between two nanometric objects in our previous study [6] in which exciton-polariton degrees of freedom are eliminated. In this stage, we assume that phonon and photon reservoirs are in vacuum states, *i.e.*, zero temperature. Therefore, a process of stimulated absorption and emission for phonons and photons is not considered. What we focus on in this paper is the transition dynamics from one-exciton state to two-exciton state, and vice versa, and thus, we introduce six bases to present the state of the system as depicted in Fig. 2. Taking the expectation values by using these bases, we can evaluate the population dynamics in this system.

2.2 Dynamics driven from definite initial conditions

Before discussing the transient dynamics, we examine the dynamics driven from definite initial conditions. First, we pay attentions to the dynamics for one-exciton states where the initial conditions are set as $\rho_{A'B,A'B}(0) = 1$ and otherwise zero, where the subscripts are labeled as follows: $|0\rangle_A$ and $|1\rangle_A$ are abbreviated as A and A', and $|00\rangle_B$, $|10\rangle_B$, and $|01\rangle_B$ as B, B', and B''. In this case, two-exciton states are not excited, and the equations of motion are closed within one-exciton states. Analytical solutions in this problem

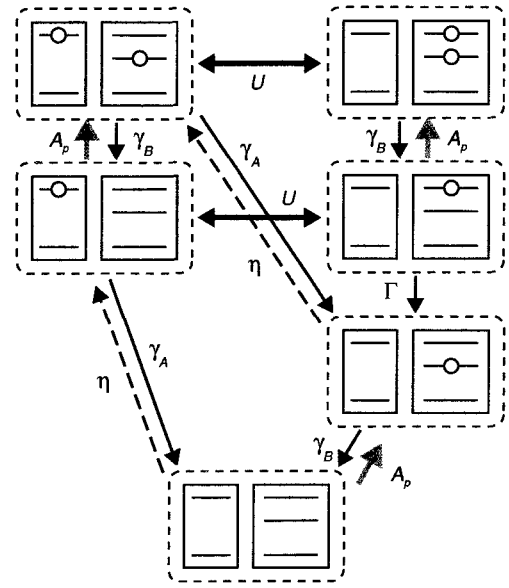

Fig. 2: Bases of the coupled quantum dot system with an exciton and two excitons.

can be obtained rigorously, and the population of the level $|1\rangle_A$ is given as

$$\begin{aligned}\rho_{A'B,A'B}(t) = &\exp\left[-\frac{\Gamma+\gamma_A}{2}t\right] \\ &\times\left[\cosh(Zt) + \frac{\Gamma-\gamma_A}{Z}\sinh(Zt)\right]^2,\end{aligned} \tag{3}$$

where $Z = \left[(\Gamma-\gamma_A)^2/4 - U^2\right]^{1/2}$. We theoretically estimate that the optical near-field interaction U is much stronger than the radiative relaxation constant γ_A. When we assume the condition $\gamma_A \ll U < \Gamma$, Eq. (3) is reduced to a simple form as

$$\rho_{A'B,A'B}(t) = \exp\left[-2U\left(\frac{2U}{\Gamma}\right)t\right], \tag{4}$$

Apparently, the decay time on the level $|1\rangle_A$ is determined by $\Gamma/(4U^2)$, and the ratio in the round brackets in Eq. (4) represents a balance of the optical near-field interaction and intra-sublevel transition. Figure 3(a) shows a plot of Eqs. (3) and (4) as a function of time. In this figure, we can observe fast energy transfer from $|1\rangle_A$ to $|10\rangle_B$ in the order of 200 ps.

On the other hand, when a two-exciton state is prepared in which the level $|10\rangle_B$ is initially filled, it follows from Eq. (1) that intra-sublevel transition is prohibited because of the Fermion-like feature of excitons. In this case, the excitation remains in the resonant level of either QD-A or B, leading to long decay time. The equations of motion for two-exciton states are completely decoupled from one-exciton states,

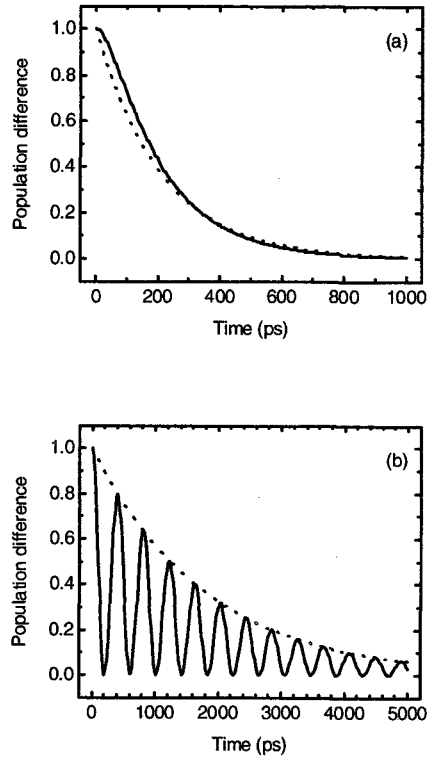

Fig. 3: Time evolution of the populations in QD-A. Initial conditions are set as follows: (a) completely excited on the level $|1\rangle_A$ in QD-A and (b) simultaneously excited on the levels $|1\rangle_A$ and $|10\rangle_B$. The fixed parameters are set as U^{-1}=130 ps, Γ^{-1}=20 ps, γ_A^{-1}=5.9 ns, and γ_B^{-1}=2.1 ns. The dotted curves in (a) and (b) represent exponential decay functions with decay constants of $2U/(2U/\Gamma)$ and $\gamma=\gamma_A/2+\gamma_B$, respectively.

and the analytic solutions are obtained by replacing the phonon relaxation constant Γ in Eq. (3) with zero and by multiplying the factor $e^{-\gamma_B t}$. Although two-exciton states are connected to one-exciton states with the radiative transition, the lifetime of population in the level $|1\rangle_A$ is in the order of 200 ps for one-exciton states as mentioned above. The population in two-exciton states has long decay time in the order of a few nanoseconds, while the population in one-exciton state is much smaller than that in two-exciton state. Therefore, the population of two-exciton state $\rho_{A'B',A'B'}(t)$ is dominant in the luminescence-intensity measurement. The result is shown in Fig. 3(b). Sinusoidal oscillation appears with an exponential decay whose decay constant is given by $\gamma=\gamma_A/2+\gamma_B$. Our further interests are transient transitions between above two situations in order to investigate excitation or signal control toward to nanophotonic devices.

3. TRANSIENT RESPONCE BY A LIGHT PULSE

Suppose to apply a light pulse to excite the level $|10\rangle_B$ in QD-B in order to demonstrate a change from a steady state. For the purpose, we introduce a coupling to a laser photon reservoir. Then, the following terms

$$\begin{aligned}&\eta\{[\hat{A}\hat{\rho}(t),\hat{A}^\dagger]+[\hat{A}^\dagger,\hat{\rho}(t)\hat{A}]\}\\&+A_p(t)\{[\hat{B}_1\hat{\rho}(t),\hat{B}_1^\dagger]+[\hat{B}_1^\dagger,\hat{\rho}(t)\hat{B}_1]\}\end{aligned} \tag{5}$$

are added to Eq. (1), where η and $A_p(t)$ are the rate of weak continuous excitation of QD-A to achieve a steady state, and that of strong pulse excitation for state filling in QD-B, respectively. These rates are proportional to the number of photons, and thus, the stimulated absorption and emission processes are involved. Here we assume incoherent excitations for simplicity. It would be valid if the period of the Rabi oscillation due to the laser pulse is longer than the applied pulse width. The light pulse couples one-exciton states to two-exciton states, and rigorous analytic solutions are hardly obtained. In the following, we show approximated solutions for the transient dynamics, separating the dynamics into two stages, an early stage and a later stage.

The steady state before applying the light pulse can be obtained from Eqs. (1) and (5) by setting the left hand side as zero. The populations in the steady state under the condition of $\gamma_A,\gamma_B \ll U < \Gamma$ are expressed as

$$\begin{aligned}&\rho_{A'B,A'B}(0_-)=\frac{\eta}{2U}\frac{\Gamma}{2U},\quad \rho_{AB',AB'}(0_-)=\frac{\eta}{\Gamma}(\sim 0),\\&\rho_{AB',AB'}(0_-)=\frac{\eta}{\gamma_B}.\end{aligned} \tag{6}$$

After the pulse excitation, each population is divided into two components. For example, the populations on the level $|1\rangle_A$ are given by $\rho_{A'B,A'B}(0_+)=R\rho_{A'B,A'B}(0_-)$ and $\rho_{A'B,A'B}(0_+)=(1-R)\rho_{A'B,A'B}(0_-)$, where R denotes the changing rate from one-exciton states to two-exciton states. The other populations in Eq. (6) change in a similar way.

In the early stage, two-exciton states have long decay time as mentioned in Sec.2, and thus, it is reasonable to be $\rho_{AB',AB'}(t)\approx\rho_{AB',AB'}(0_+)$. From this approximation, equations of motion for one-exciton and two-exciton states are decoupled, and an approximated solution is obtained as

$$\begin{aligned}\rho^{\text{early}}_{A'B',A'B'}(t)=\frac{R}{2}\Big\{&\rho_{A'B,A'B}(0_-)e^{-\gamma t}\\&+\rho_{AB',AB'}(0_-)(1-e^{-\gamma t})\\&+\Big[\rho_{A'B,A'B}(0_-)\cos(2Ut)\\&+i\frac{\Delta\rho_{A'B,AB'}(0_-)}{2}\sin(2Ut)\Big]e^{-\gamma t}\Big\}.\end{aligned} \tag{7}$$

The second term in Eq. (7) represents an

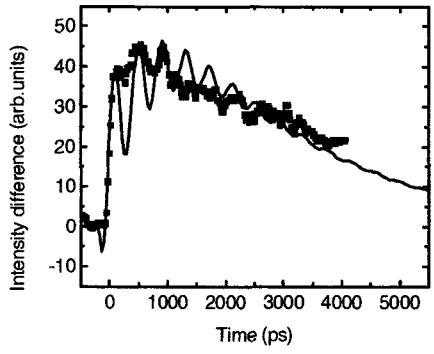

Fig. 4: Transient population dynamics from a steady state by applying a light pulse. The square dots represent the data of time-resolved experiment with CuCl quantum cubes embedded in a NaCl matrix. Analytical solution is depicted as the solid curve. The appropriate constant is multiplied by the analytical solution in order to compare the experimental data. The parameters are set as U^{-1}=130 ps, Γ^{-1}=20 ps, γ_A^{-1}=5.9 ns, and γ_B^{-1}=2.1 ns.

increase of the population that originates from the state filling of the level $|10\rangle_B$, and the terms in the rectangular brackets denote the coherence effect depending on the initial population.

On the other hand, in the later stage, the coherence in the two-exciton states disappears due to the radiative relaxation in both quantum dots. Therefore, the dynamics of the population follows a simple exponential curve as

$$\rho^{\text{later}}_{A'B',A'B'}(t) = R\rho_{A'B,A'B}(0_-) - (1 - R\rho_{A'B,A'B}(0_-))e^{-\gamma t}. \quad (8)$$

An approximated solution in the whole time can be given by multiplying the density matrix elements in the early and later stages as

$$\rho_{A'B',A'B'}(t) = \rho^{\text{early}}_{A'B',A'B'}(t)\rho^{\text{later}}_{A'B',A'B'}(t). \quad (9)$$

In Fig. 4, the temporal evolution obtained from Eq. (9) is plotted, together with experimental data. In first order approximation of η (weak excitation), the population increases with an exponent of $2\gamma_B$, and oscillates with a frequency of $2U$. The exponent reflects an increment due to continuous excitation and the components flowing into the lowest level $|10\rangle_B$ in the steady state. It follows the result that the fast state-filling time observed in the experiment comes from the effect of nutation or coherent energy transfer. With the radiative relaxation, the coherence effect is lost, and the excitation is occupied on the resonant energy levels, which have long decay time as same as in two-exciton states. The slow decay observed around t = 2 ns originates from the competition of the population increase due to the state filling and decrease due to the radiative relaxation. Hence, our approximated solution well describes the experimental results and clarifies physics behind the phenomenon.

4. CONCLUSIONS

In order to confirm a possibility to control excitation transfer in a coupled quantum dot system, we have investigated transition from a steady state to transient state by applying a light pulse. An approximated analytic solution of the dynamics well explains characteristic features of recent experimental results. Both fast increase of population in the early stage and the oscillation behavior originate from coherent effect between two quantum dots with sub-levels to be filled. This state-filling effect and the radiative relaxation cause the slow decay in the stage of a few nanoseconds. As a result, signal transfer and control in such a quantum dot system can be achieved, and we expect that this is a promising technique for future nanophotonic devices.

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