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Superradiant multiple pulses from excitons interacting with optical near fields

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A model of an N two-level system interacting with optical near fields is presented. Varying the initial conditions, we examine the dynamics of the electric dipoles of the system to predict a collective dipole oscillation, *i.e.*, manifestation of a dipole-ordered state. Multiple pulses superradiantly emitted from such a state is also shown, and the origins of the phenomena are discussed, on the basis of a localized photon model using both semi-classical and quantum master equations. Brief comments on an application to a nanometric photonic component are given.

Key words: localized photon, dipole ordering, superradiance, Dicke model

1. INTRODUCTION

Nano-fabrication and its application to nanophotonic devices have been actively investigated, and special attention has been paid to optical near-field approaches because of their potential for ultra-high speed operation and miniaturization[1, 2]. It is now essential to clarify the inherent phenomena of optical near fields that are applicable to nano-fabrication and devices of a nanometer scale. In this context, a localized phton model has been proposed to predict a collective dipole oscillation of an N two-level quantum dots (QD) system after local manipulation of the initial quantum states [3]. However, the origin and features of this dipole-ordered state have not been fully explored. In this paper we report an intriguing phenomenon of multiple pulses superradiantly emitted from the dipole-ordered state on the basis of a localized photon model using both semi-classical and quantum master equations. We discuss its mechanism, features, and a possible nanophtonic application.

2. MODEL HAMILTONIAN

In this section we review the localized photon model which has been introduced in Ref. [3]. The system consists of N two-level QDs closely configured in a ring and localized photons coupled to each QD. The model Hamiltonian H can be written in the following form:

$$H = H_{\mathrm{a}} + H_{\mathrm{b}} + H_{\mathrm{int}}, \tag{1a}$$

$$H_{\mathrm{a}} = \varepsilon \sum_{n=1}^{N} a_n^{\dagger} a_n + V \sum_{n=1}^{N} \left(a_{n+1}^{\dagger} a_n + a_n^{\dagger} a_{n+1} \right), \tag{1b}$$

$$H_{\mathrm{b}} = E \sum_{n=1}^{N} b_n^{\dagger} b_n, \tag{1c}$$

$$H_{\mathrm{int}} = U \sum_{n=1}^{N} \left(a_n^{\dagger} b_n + a_n b_n^{\dagger} \right), \tag{1d}$$

where H_{a} and H_{b} describe localized photons and excitons in QDs, respectively, and the intradot interaction between the localized photons and the excitons is denoted as H_{int}. Creation and annihilation operators for a localized photon and an exciton in each QD labeled by n are expressed as $a_n^{\dagger}, a_n$ and $b_n^{\dagger}, b_n$ respectively. We apply the boson commutation relations to the localized photons as

$$[a_n, a_{n'}^{\dagger}] = \delta_{nn'}, \quad [a_n, a_{n'}] = [a_n^{\dagger}, a_{n'}^{\dagger}] = 0.$$

The excitons on a site obey the fermion commutation relations while excitons at different sites satisfy the boson commutation relations as

$$[b_n, b_{n'}^{\dagger}] = \delta_{nn'}(1 - 2b_n^{\dagger} b_n).$$

The constant energies of the localized photons and excitons are assumed to be $\varepsilon = \hbar\omega$ and $E = \hbar\Omega$ respectively. The hopping energy of the localized photons is represented as $V = \hbar v$, and $U = \hbar g$ is for the conventional dipolar coupling between the localized photons and the excitons in the rotating wave approximation. We show a illustration of our system in Fig. 1.

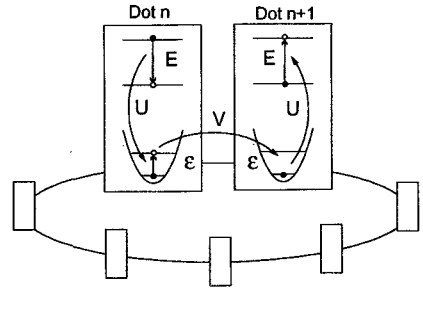

Fig. 1. Model of a quantum dots system interacting with localized photons. Each quantum dot interacts with the adjacent dots via localized photons.

3. DIPOLE-ORDERED STATE

Using the Hamiltonian (1), we can obtain the second-order perturbation solution for temporal evolution of the dipole moment at the n-th site [3] $\langle P_n(t)\rangle = \langle b_n(t) + b_n^\dagger(t)\rangle$ as

$$\langle P_n(t)\rangle = \langle P_n(0)\rangle \left(\cos\Omega t - g^2 \sum_j c_j(t) P_{nj}^2 \right) + g^2 \sum_j \sum_{m\neq n} c_j(t) P_{nj} P_{mj} \langle P_m(0)\rangle \langle W_n(0)\rangle . \quad (2)$$

Here P_{nj} is the diagonalization matrix element of the Hamiltonian of the localized photons H_a, and $c_j(t)$ is the time dependent coefficient depending on the exciton energy $\hbar\Omega$ and j-th eigenvalue $\hbar\lambda_j$ of the Hamiltonian H_a. The initial dipole moment and the population difference at site n are denoted as $\langle P_n(0)\rangle$ and $\langle W_n(0)\rangle = \langle b_n^\dagger b_n - b_n b_n^\dagger\rangle$, respectively. From Eq. (2) it follows that the dipole distribution of the system at time t can be controlled by the initial population differences, which indicates a possibility of a local manipulation of dipole moment of an arbitrary site which results in a collective behavior of the dipoles, or in a dipole ordered state.

On the basis of Eq. (2), we numerically investigate the dynamics of the dipoles depending on initial conditions. Figure 2 illustrates an intriguing non-radiative case, where the alternating dipoles $\langle P_n(0)\rangle = (-1)^n$ are initially set, *i.e.*, the total dipole of the system is vanishing.

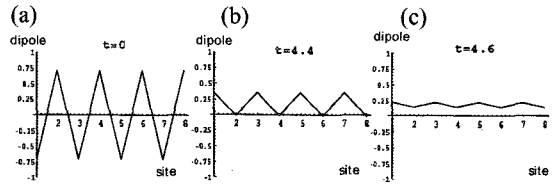

Fig. 2. Temporal evolutions of the dipole moments for the system size N=8 at time (a) t=0, (b) t=4.4 (c) t=4.6 with a unit of time $\hbar/E$. A unit of energy of our system is a typical excitation energy E of a quantum dot which is of the order of 1 eV. The vertical and horizontal axes represent the dipole and the site number, respectively. From (b) and (c), one can see that a collective oscillation is growing, which is originated from a local manipulation of the initial distribution of the population difference as $\langle W_n(0)\rangle = -\langle P_n(0)\rangle$.

Figurers 2(b) and 2(c) show that a collective oscillation of the dipoles is growing, *i.e.*, a dipole-ordered state is manifested. The point of the dipole ordering is a local manipulation in which the sign of the population difference in each QD is set opposite to that of the corresponding diopole as $\langle W_n(0)\rangle = -\langle P_n(0)\rangle$.

4. RADIATION PROPERTY

We examine the radiation properties of the dipole-ordered state discussed above, for which we expect that intense radiation is emitted. For simplicity, we renormalize the localized photonic degrees of freedom into an effective Hamiltonian, $H_{\text{QD-eff}}$, with the help of the unitary transformation [4, 5]:

$$H_{\text{QD-eff}} = \sum_n \hbar\Omega b_n^\dagger b_n + \sum_n \sum_{m\neq n} \hbar J_{nm} b_n^\dagger b_m , \quad (3)$$

where the hopping energy of excitons is expressed as

$$J_{nm} = \sum_j \frac{g^2}{\Omega - \lambda_j} P_{nj} P_{mj} .$$

In addition, a radiation field weakly interacting with the QDs is included in the system as a reservoir that makes the system dissipative. Then, equations of motion for the density operator $\rho_{\text{QD}}(t)$ of the QD system can be written as

$$\frac{\partial \rho_{\text{QD}}(t)}{\partial t} = -iL_{\text{eff}}\rho_{\text{QD}}(t) + \gamma([R_-\rho_{\text{QD}}(t), R_+] + [R_-, \rho_{\text{QD}}(t)R_+]). \quad (4)$$

where L_{eff} is the Liouville operator associated with $H_{\text{QD-eff}}$, and the relaxation constant due to the elimination of the radiation field's degrees of freedom is denoted as γ. The raising and lowering operators $R_\pm$ are defined, respectively in terms of the creation and annihilation operators of excitons in the QDs as

$$R_- = \sum_n b_n , \quad R_+ = \sum_n b_n^\dagger .$$

To study the emission properties of the radiation, we solve Eq. (4) and calculate the radiation intensity

$$I(t) = \langle R_+(t) R_-(t)\rangle A ,$$

where A is the Einstein's A coefficient which represents the probability of spontaneous emission of an excited single two-level system. We first solve Eq. (4) semi-classically without taking account of quantum correlations. This method has an advantage over the full quantum method to enable us to easily handle a relatively large number N system. However, it should be noted that the Dicke model shows superradiance from the initial condition of a perfect inversion population, while semi-classical descriptions in the same condition do not predict superradiance because the system has initially no dipoles [6, 7]. We consider the case discussed in Sec. 3 that is initially non-radiative and subsequently dipole-ordered. Figure 3 shows that multiple pulses can be emitted from the system superradiantly (while the Dicke model shows no superradiance under such conditions), and indicates that the ordered total dipole moment of the system plays an important role in the oscillating radiation profile.

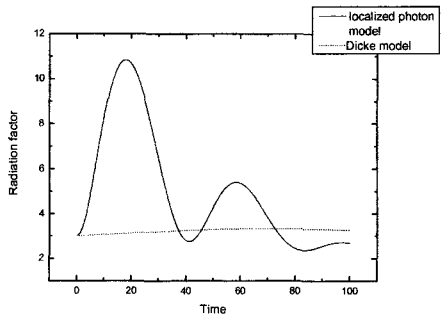

Fig. 3. Temporal evolution of the radiation intensity for the system size N=8 obtained semi-classically without a quantum correlation among excitons. The solid curve is the result for the localized photon model while the dotted curve represents the result for the Dicke model. The initial condition is the same with Fig. 2. Three radiation peaks are seen for the dipole-ordered state when γ=0.005 is used. The coupling strength of exciton-localized photon interaction is assumed to be weak, g=0.2.

Considering the quantum correlation, we then solve Eq. (4) with the same initial conditions and system size N=4 to further investigate the origin and mechanism of the multiple pulse generation and quantum effects. As shown in Fig. 4, a supperradiant pulse is emitted in our case, while the Dicke model shows no superradiance under such conditions.

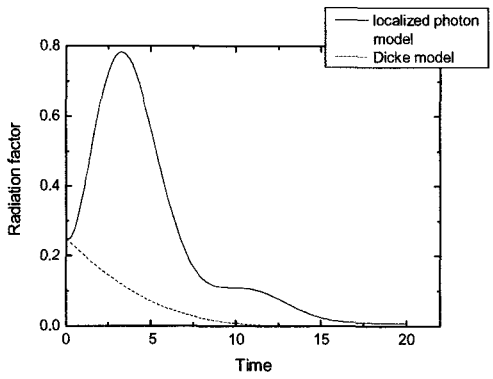

Fig. 4. Temporal evolution of the radiation intensity for the system size N=4 obtained with quantum correlations. The solid curve is the result for the localized photon model while the dotted curve represents the result for the Dicke model. The parameters are γ=0.05 and g=0.5.

The difference is due to the following fact: the dipole-ordered state appears in our case while it does not in the Dicke model. This is qualitatively predicted in the semi-classical approach (see Fig 3). This feature is seen more clearly in Figs. 5(a)-(b) when the coupling strength of the exciton-localized photon interaction is changed.

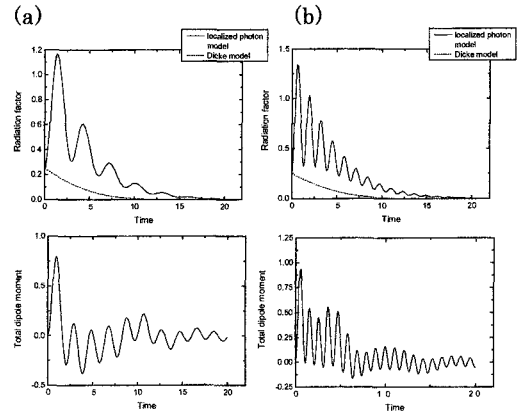

Fig. 5. Temporal evolution of radiation intensity (upper) and the total dipole of the system (lower) obtained with quantum correlations. The solid curves are the result for the localized photon model while the dotted curves represent the result for the Dicke model. The parameters and the system size are the same as in Fig. 4, except (a) g=0.8 and (b) g=1.2.

As the coupling becomes stronger, the oscillation frequency of the total dipole moment increases because of the nonlinearity of the first term on the right hand side of Eq. (4) (see lower of Fig 5). It results in collective multiple pulse generation (see upper of Fig. 5). This phenomenon can be applied to a nanometric photonic source whose radiation intensity is proportional to the square of the total number of QDs [1].

Comparing the semi-classical results shown in Fig. 3 with the quantum results in Fig. 5, we find a similar behavior of the radiation profiles between them. In Fig. 6 we show the radiation intensities calculated (a) semi-classically and (b) with quantum correlation under the identical conditions.

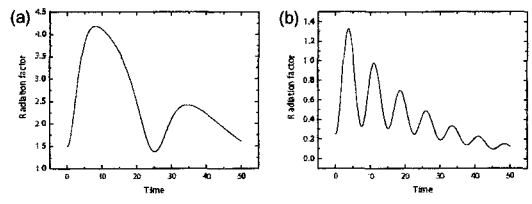

Fig. 6. Temporal evolution of the radiation intensity obtained (a) semi-classically without a quantum correlation among excitons and (b) with quantum correlations. We use the parameters γ=0.05 and g=0.5 for the system N=4.

Although the frequencies and the amplitudes are different, both results have qualitatively a similar tendency, that is, occurrence of multiple peaks. One can thus infer that when the total dipole moment is initially not zero the semi-classical treatment can qualitatively describe the radiation property of the system. Note that when there is initially no dipole moment, the quantum

fluctuation is essential as a trigger of radiation. On the other hand, without quantum fluctuation, the initial non zero dipole moments can develop an ordering process due to the non-linear dynamics of the system, and it leads to strong radiation from the system. Since the semi-classical approach does not require a large scale computation, it has an advantage that one can easily examine the system of a relatively large number of sites. Our discussion indicates that the semi-classical approach is powerful for qualitative investigation of radiation properties of a large system. However, for quantitative investigation and for classification of the approximation it is important to have a clear criterion for the applicability of the semi-classical approach.

5. CONCUSION AND DISCUSSION

We have predicted superradiant multiple pulse emission from the dipole-ordered state prepared by the exciton-localized photon interaction, and discussed the origin of the phenomenon, on the basis of the localized photon model using the dissipative master equation. One expects that this kind of phenomenon can be applied to a component of nanophotonics. The superradiant multiple pulse generation has been discussed in the Dicke model for a large system [8] and a Frenkel exciton system with dipole-dipole interaction [9]. In the former multiple pulses are the results of the stimulated emission and absorption of emitted photons stayed inside the large system. In the latter the oscillation in the radiation originates from the hopping of the excitation due to the dipole-dipole interaction. The origin of the multiple pulse generation in our system is similar to the latter case. However, our system includes a dipole-ordered state, and the mechanism is not completely same as that of Ref. [9]. Moreover, we have clarified that the multiple pulsation is a direct consequence of the ordering of the total dipole.

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