Superradiance and Dipole Ordering of an $N$ Two-Level System Interacting with Optical Near Fields

Akira SHOJIGUCHI*, Kiyoshi KOBAYASHI, Suguru SANGU, Kazuo KITAHARA$^1$ and Motoichi OHTSU$^2$

ERATO Localized Photon Project, Japan Science and Technology, Tokyo 194-0004
$^1$Division of Natural Sciences, International Christian University, Tokyo 181-8585
$^2$Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology, Yokohama 226-8502

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A model is presented for a system of $N$ two-level excitons interacting with each other via optical near fields represented as localized photons. In a low exciton density limit, quantum dynamics of the dipole moments or quantum coherence between any two energy levels is linear. As the exciton density becomes higher, the dynamics becomes nonlinear, and the system has several kinds of quasi-steady states of the dipole distribution depending on the system parameters. These quasi-steady states are classified with the help of the effective Hamiltonian that is derived from the renormalization of degrees of freedom of localized photons with a unitary transformation. Among them there exist a “ferromagnetic” state (dipole-ordered state), in which all electric dipoles are aligned in the same direction, and an “anti-ferromagnetic” state, where all dipoles alternatingly change the direction. In addition, we show that an arbitrary state can be transformed into a dipole-ordered state by manipulating initial values of the population differences appropriately. For example, if we initially prepare a dipole-forbidden state, which is similar to the “anti-ferromagnetic” state and cannot be coupled with propagating far fields, and if we manipulate the distribution of the population differences properly, the initial state evolves into a dipole-ordered state. The radiation property of such dipole-ordered states is examined in detail. Neglecting energy dissipation by radiation, we find that some of the ordered states show strong radiation equivalent to Dicke’s superradiance. Then by introducing a radiation reservoir, the dissipative master equation is derived. Solving the equation with and without quantum correlations, we numerically show that multiple peaks in the radiation profile can survive in both cases. The mechanism of this phenomenon is discussed, and a brief comment on an application to photonic devices on a nanometer scale is given.

KEYWORDS: optical near field, localized photon, local coupling, global coupling, quantum dot, exciton, electric dipole, superradiance, Dicke model

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1. Introduction

1.1 Background and optical near fields

Recent progress in nano-fabrication and nano-manipulation has enabled us to explore the excitonic processes and to control quantum states in an electronically isolated semiconductor quantum dot (QD) or several coupled quantum dots (QDs) that are also electronically isolated from other QDs. Near-field optical approaches have achieved a breakthrough in nano-optics, in nano-science, and in nano-technology, utilizing the spatial localization and ultra-high speed originating from the strong coupling of matter and light on a nanometer scale. For example, scanning near-field optical microscopes (SNOM) have been developed as optical microscopes with resolution far beyond the diffraction limit of light. A variety of experiments have been carried out in order to control and to manipulate a single atom, molecule, or QD with accuracy beyond the diffraction limit. In the near future, the exciton dynamics will be experimentally investigated with the help of the near-field optical techniques by the local excitation of arbitrarily selected QDs. Therefore, optical near fields are essential for the exploration of excitation processes in a confined space. Optical near fields are localized around a material system, e.g., around a near-field probe tip or a QD system, depending on the size of them, and thus their noticeable effects manifest themselves when the two systems are close within a distance of the order of the size. The interaction between two material systems is even more dominated by optical near fields, not by propagating light or far fields, if the size of the two systems is smaller than the wavelength of light, and optical near fields and material polarization are mutually related. Therefore in nano-optics, one has to consistently describe both material systems and optical near fields, and needs to know the dynamical properties of the material systems interacting with optical near fields.

We give a few examples to understand the spatial localization of optical near fields. The evanescent field generated on a planar dielectric surface can be obtained by analytical continuation of the wave vector $k_\perp$ normal to the surface as $\exp(i k_\perp r - \gamma z)$ with $k_\perp = i\gamma$ ($\gamma$: real), whose localization range is about that of the wavelength of incident light. Similarly, optical near fields around a tiny dielectric sphere whose size is much smaller than the wavelength is described by the spherical Hankel function with analytic continuation of the radial component of the wave vector $k_\perp$ to the pure imaginary $k_\perp = i\gamma$; the first kind Hankel function of order 0 gives a typical form of optical near field around a sphere as

$$\phi = \frac{e^{-\gamma r}}{\gamma r}.$$  (1.1)

The fields satisfying the spherical boundary condition can be expanded in terms of plane waves, or can be expressed in the angular spectrum representation, and it follows from a numerical analysis that the distribution of the angular
spectrum has a peak at $\gamma = 1/\alpha$. Applying the result to eq. (1.1), we see that the localization range of optical near fields around a sphere is of the order of the radius $a$, i.e., the size of the material system. Here note that optical near fields should be derived from microscopic polarization of matter by using, for example, a nonlocal theory in a semi-classical way proposed by Cho,\textsuperscript{5} though we can formally obtain an optical near field from a solution of the Helmholtz’ equation by analytical continuation of the normal component of the wave vector to the pure imaginary.

We are interested in the dynamics of nanometric material systems interacting with optical near fields as well as quantum behaviors. However, it is very difficult to employ the nonlocal approach for the quantization of the electromagnetic fields and for treating the dynamics of the material systems and the optical fields because of the complexities of the self-consistent procedure in practice. Moreover, we have no formal or phenomenological theory to resolve the above problems. Therefore a new approach is required, which enables us to easily deal with the quantum dynamics of material systems interacting with optical near fields. In this paper, we address the issues and present a simple phenomenological model of optical near fields in order to discuss dynamical properties of a nanometric material system interacting with optical near fields.

1.2 Optical near fields and propagating fields

The main difference between a material system coupled with optical near fields and a material system coupled with propagating far fields, is that one is locally coupled and the other is globally coupled as schematically depicted in Fig. 1. Since the propagating field is usually expanded in terms of a plane wave basis, emitted photons as plane waves propagates from one site to far sites of the material system and can excite the far sites, as shown in Fig. 1(a). On the other hand, as shown in Fig. 1(b), “near field photons” or “localized photons” can only move to near sites and excite them. Generally speaking, the dynamics of a locally coupled system is different from that of a globally coupled system in respect to the relaxation speed towards equilibrium.\textsuperscript{6) 7) Therefore we can expect that the dynamics and equilibrium states for a locally coupled system are different from those for an ordinary globally coupled system such as a propagating field-matter system.\textsuperscript{7) From this point of view, it is significant to investigate a nanometric material system interacting with optical near fields.

In this paper we present a model for a material system, in particular, for a quantum dot system interacting with optical near fields, and discuss the dynamics of the electric dipole moment and the radiation property of the system in detail. For comparison a material system interacting with radiation field is also examined. We study the propagation of an initial excitation signal in a quantum dot which is locally prepared by optical near fields, and is described in terms of excitons in an $N$ two-level system. In a dilute limit, excitons are approximately treated as bosons, and a rigorous solution of the Heisenberg equation shows that the dipole moment representing quantum coherence between any two levels follows linear dynamics. In a non-dilute case, excitons at a site obey the fermion commutation relations while excitons at different sites satisfy the boson commutation relations, which results in nonlinear equations of motion. We predict a coherent oscillation of all the dipoles of the system (dipole ordering), and strong radiation from some of the dipole-ordered states, which are close to the Dicke’s superradiance.\textsuperscript{5)

1.3 Two-level system with propagating fields: Dicke’s superradiance

Superradiance, the cooperative emission of radiation from a collection of excited two-level systems was originally discussed by Dicke in 1954. He found that under a certain condition the radiation rate is proportional to the square of the number of two-level systems involved, and that an intense pulse is emitted.\textsuperscript{8) Superradiant phenomena have occupied considerable attention of a large number of authors in 1970’s.\textsuperscript{9–23) In a small system whose size is much smaller than the wavelength of radiation, the equations of motion for the two-level system coupled to the radiation field (Dicke model) is given as follows:

$$\frac{\partial \rho_\lambda}{\partial t} = -\gamma_0 (R_+ R_- \rho_\lambda - 2R_- R_+ \rho_\lambda + \rho_\lambda R_+ R_-),$$

where $\rho_\lambda$ is the density operator after eliminating radiation field’s degrees of freedom from total density operator of the system, $R_\pm$ is the collective raising and lowering operators of a two-level system,\textsuperscript{8) and $2\gamma_0$ is the inverse of the spontaneous emission lifetime of each two-level system (Einstein’s $A$-coefficient). From eq. (1.2) one obtains differential equations of the observables of the two-level system and a radiation pulse emitted under a certain initial condition whose height and width are proportional to $N^2$ and $1/N$, respectively. By using a semi-classical approximation of neglecting quantum correlations in the two-level system, an analytical solution\textsuperscript{10,13) for radiation intensity $I$ is written as

$$I \propto N^2 \text{sech}^2 \left[ \gamma_0 N (t - t_0) \right],$$

which shows a radiation profile of the cowbell shape. It should be noted that the semi-classical approximation does not correctly predict the dynamics starting from the completely inverted states\textsuperscript{13,14) with no dipole moments because the quantum fluctuations of the dipole moments are essential in the radiation process.

In a large system the superradiant state is affected by both inhomogeneity of the two-level system in a sample and reabsorption of emitted photons,\textsuperscript{15) which results in the decoherence; namely, some part of the coherence in the system is destroyed by the induced dipole–dipole interaction. It leads to reduction of the peak height and an extension

\[ B_{a} \rightarrow a \rightarrow e^{ikx} \rightarrow a \rightarrow B^{*} \]

\[ B_{a'} \rightarrow a' \rightarrow a \rightarrow B^{*} \]

Fig. 1. Schematic drawing for (a) a global coupling system and (b) a local coupling system. Each element of the system in (a) can directly interact with arbitrary elements at far site while elements at near site can only interact with each other in (b).
of the tail of the radiation profile. For a two-level system confined in a long cylindrical shape of sample, Bonifacio et al. predicted multiple peaks in the radiation profile that stem from the stimulated absorptions and emissions in the long active region. Experimental observation using a cylindrical sample was done by Skribanowitz and the experimental results were analysed by Bonifacio et al. Tokihiro et al. examined a linear excitonic system, where excitons can hop from one site to its nearest neighbors due to the dipole–dipole interaction, and showed that radiation from a totally inverted state as an initial condition exhibits a reduction of the peak intensity and the extension of the tail as discussed by Coffey et al. They also claimed that radiation from a partially excited state shows an oscillatory behavior of the radiation profile as indicated by Bonifacio et al. It is clear that the peak intensity reduction of radiation in both cases comes from the dipole–dipole decoherence, but it is not obvious whether the origin of the multiple pulse generation is the same or not. We also show multiple pulse emission in our model, and discuss the mechanism.

1.4 Outline

The paper is organized as follows. In §2 we present a model of an $N$ two-level quantum dot system interacting with optical near fields represented in terms of localized photons, and the model Hamiltonian. In §3 a rigorous solution of the equations of motion for the system is obtained within a boson approximation. With the help of the solution we investigate the propagation of the dipole moments of the system. In §4 without using the boson approximation we obtain the second-order perturbative solution to show the dynamics of the system. In §5 an effective Hamiltonian is introduced by renormalizing degrees of freedom of localized photons. With the help of the effective Hamiltonian we classify the quasi-steady states of the dipole distribution in order to discuss the origin of the dipole ordering. In §6 we examine the radiation property of the system in a weak limit, where the radiation field does not affect the dynamics of the system. In order to discuss the radiation property of the dissipative system, in §7, the master equation is derived after introducing a radiation reservoir, and is solved with and without including quantum correlations between excitons and localized photons. Finally in §8 concluding remarks are provided.

2. Model Hamiltonian

One of the most important features of optical near fields is the localization property, where it is not suitable to use a broad spreading wave as a basis function of quantization of the fields. Therefore, it is significant to find a good normal mode of the electromagnetic fields to satisfy a peculiar boundary condition, by which the optical near fields are produced and quantized. It is difficult, however, to find a general and appropriate normal mode satisfying any arbitrary boundary conditions. As an alternative approach, it is possible to model optical near fields and their important characters phenomenologically. Such an approach is adopted in this paper to formulate the problem and to discuss the dynamics of a nanoscale material system.

To describe the localizability of the optical near fields it is very effective to use a localized basis function such as a Wannier function or a delta function instead of a plane wave. However, at the same time we need to describe the property of a short range interaction, or a steep gradient of optical near fields. Taking into account such circumstances we model optical near fields phenomenologically in terms of localized photons which are described as a harmonic oscillator localized in each quantum dot site, and are only allowed to hop from one site to the nearest neighbors. Figure 2 schematically describes our model system. We suppose a closely located quantum dot chain as a nano-scale material system that is expressed as a one-dimensional $N$ two-level system, or an excitonic system with a periodic boundary condition. Since each exciton can only interact with localized photons in the same QD, each quantum dot indirectly interacts with one another via localized photons, as illustrated in Fig. 2. This model is based on the unique property of localization or non-propagation of optical near fields mentioned above, and is preferable for an intuitive understanding of coherent excitation transfer between the QDs and its manipulation by the localized photons. The model Hamiltonian of the system can be written as

$$H = H_a + H_b + H_{\text{int}},$$  \hspace{1cm} (2.1)

where $H_a$ describes localized photons, $H_b$ describes excitons, and $H_{\text{int}}$ represents the localized photon–exciton interaction. Each Hamiltonian can be expressed as

$$H_a = \sum_{n=1}^{N} \left[ e a_n^a a_n^a + V \left(a_n^a a_n^a + a_n^a a_{n+1}^a \right) \right],$$ \hspace{1cm} (2.2a)

$$H_b = E \sum_{n=1}^{N} b_n^b b_n^b,$$ \hspace{1cm} (2.2b)

$$H_{\text{int}} = U \sum_{n=1}^{N} \left( a_n^a b_n^b + b_n^b a_n^a \right),$$ \hspace{1cm} (2.2c)

where $n$ indicates the site number, and $a_n$ ($a_n^a$) and $b_n$ ($b_n^b$) represent annihilation (creation) operators of a localized photon and an exciton, respectively. The periodic boundary condition requires that the $(N + 1)$-th site corresponds to the first site. The constant energies of the localized photons and excitons are represented $\epsilon = \hbar \omega$ and $E = \hbar \Omega$, respectively. The hopping energy of the localized photons is represented...
as \( V = \hbar \nu \), and \( U = \hbar g \) gives the strength of the conventional dipolar coupling between the localized photons and the excitons in the rotating wave approximation.

We apply the boson commutation relations to the localized photons as
\[
[a_n, a_m^\dagger] = \delta_{mn}, \quad [a_n, a_m] = \dfrac{1}{C_1}a_n, a_m^\dagger = 0.
\]

(2.3)

The creation operator of an exciton, \( b_n^\dagger \), can be written in terms of the annihilation operator of a valence electron of \( n \)th site, \( c_{n0} \), and the creation operator of a conducting electron of \( n \)th site, \( c_{n}^\dagger \), as \( b_n^\dagger = c_{n0}^\dagger c_{n}^\dagger \). In the same way, the annihilation operator of an exciton, \( b_n \), can be written as \( b_n = c_{n0} c_n \). Assuming that only one exciton is generated in a quantum dot site, we obtain the relation as \( N_f = N_n \). With the help of these expressions and the fermi commutation relations for electrons, the commutation relations for excitons are derived as\(^{27,29}\)
\[
[b_n, b_m^\dagger] = \delta_{nm} (1 - 2b_n^\dagger b_n),
\]

(2.4)

which shows that excitons behave as fermions at intra-site and as bosons at inter-site. It follows from eq. (2.4) that excitons are approximated as bosons in a dilute limit of the exciton density, \( \langle b_n b_n^\dagger \rangle = (N_n) \ll 1 \).

With the boson approximation, the Heisenberg equations of motion can be rigorously solved. First, we obtain a rigorous solution of the Heisenberg equations for bosonic excitons to investigate the dynamical properties. Then we investigate the dynamics of fermionic excitons, solving the Heisenberg equations both perturbatively and numerically.

3. Dynamics of Bosonic Excitons

3.1 Boson approximation and diagonalization of the Hamiltonian

To solve the Heisenberg equation we introduce the spatial Fourier transformation for \( a_n \) and \( b_n \) as
\[
A_k = \dfrac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{i\kappa n} a_n,
\]

(3.1a)

\[
B_k = \dfrac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{i\kappa n} b_n,
\]

(3.1b)

where we set \( k = 2\pi l/N \) for \( l = 1, \ldots, N \) and the lattice constant as 1. Fourier transforms \( A_k \) satisfy the following commutation relations
\[
[A_k, A_{k'}^\dagger] = \delta_{kk'}, \quad [A_k, A_{k''}] = [A_{k'}, A_{k''}] = 0,
\]

(3.2)

and the commutation relations of Fourier transforms \( B_k \) are similarly obtained as
\[
[B_k, B_{k'}^\dagger] = \dfrac{1}{N} \sum_{n,m} e^{i\kappa (n-m')} [b_n, b_{m'}^\dagger] = \dfrac{1}{N} \sum_{n} e^{i\kappa n} (1 - 2b_n^\dagger b_n).
\]

(3.3)

It follows from this expression that when the expectation value of the number of excitons is small as \( \langle b_n^\dagger b_n \rangle \ll N \), the second term of the right hand side of eq. (3.3) can be neglected, and excitons are approximated as bosons as
\[
[B_k, B_{k'}^\dagger] = \delta_{kk'}, \quad [B_k, B_{k''}] = [B_{k'}, B_{k''}] = 0,
\]

(3.4)

where the following relation
\[
\dfrac{1}{N} \sum_{n} e^{i\kappa (n-k)} n = \delta_{kk'}
\]

(3.5)

is used. Fourier inverse transforms are also given as
\[
a_n = \dfrac{1}{\sqrt{N}} \sum_{k=1}^{N} e^{-i\kappa n} A_k,
\]

(3.6a)

\[
b_n = \dfrac{1}{\sqrt{N}} \sum_{k=1}^{N} e^{-i\kappa n} B_k,
\]

(3.6b)

where the summation of \( k \) runs over the first Brillouin zone (1BZ). Using eqs. (3.1a), (3.1b), and the following relation
\[
\dfrac{1}{N} \sum_{k=1}^{N} e^{i\kappa (n-k')} = \delta_{nm'},
\]

(3.7)

we can transform the Hamiltonian (2.1) into a Fourier transform representation
\[
H = \sum_{k} \{ (\varepsilon + 2V \cos k) A_k^\dagger A_k + E B_k^\dagger B_k + U (A_k B_k^\dagger + A_k^\dagger B_k) \} = \sum_{k} H_k.
\]

(3.8)

Note that the dispersion relation of localized photons is changed from \( \varepsilon \) to \( (\varepsilon + 2V \cos k) \). The transformed Hamiltonian \( H_k \) can be diagonalized into a simple quadratic form
\[
H_k = \langle A_k^\dagger, B_k^\dagger \rangle \begin{pmatrix} \varepsilon + 2V \cos k & U \\ U & E \end{pmatrix} \begin{pmatrix} A_k \\ B_k \end{pmatrix},
\]

\[
= \langle A_k^\dagger, B_k^\dagger \rangle \begin{pmatrix} e^U & U \\ U & E \end{pmatrix} \begin{pmatrix} A_k \\ B_k \end{pmatrix}
\]

\[
= \sum_{l=1}^{2} M_l X^l(k) X(k),
\]

(3.9)

where the abbreviations \( e = \varepsilon + 2V \cos k \) and \( X^l(k) = (A_k^l, B_k^l) \) are used. The matrix kernel of the Hamiltonian, \( M \), is diagonalized as
\[
S^T MS = \begin{pmatrix} \lambda_+ & 0 \\ 0 & \lambda_- \end{pmatrix},
\]

(3.10)

by an orthogonal matrix \( S \)
\[
S = \begin{pmatrix} L + K \sqrt{L - K} \\ \frac{2L}{\sqrt{L - K}} \frac{2L}{L + K} \end{pmatrix}.
\]

(3.11)

Here eigenvalues \( \lambda_\pm \) are written as
\[
\lambda_k = \dfrac{e + E}{2} \pm \dfrac{1}{2} \sqrt{(e - E)^2 + 4U^2}
\]

(3.12)

\[
= K \pm E \pm L
\]

with \( K = (e - E)/2 \) and \( L = \sqrt{K^2 + U^2} \). Using the polariton transformation as
\[
\begin{pmatrix} a_k \\ b_k \end{pmatrix} = S^T \begin{pmatrix} A_k \\ B_k \end{pmatrix},
\]

(3.13)

we can finally diagonalize the Hamiltonian (3.8) as follows:
3.2 Dipole dynamics driven by local excitation

With the help of the diagonalized Hamiltonian eq. (3.14), we can immediately solve the Heisenberg equation and can express the time evolution of exciton–polariton operators (Ak and Bk) as

\[
\begin{align*}
\alpha_k(t) & = e^{-iL/E t/2h} \alpha_k + \sum_k \left( A_k \alpha_k^\dagger + A^\dagger_k \alpha_k \right), \\
\beta_k(t) & = e^{-iL/E t/2h} \beta_k + \sum_k \left( B_k \beta_k^\dagger + B^\dagger_k \beta_k \right),
\end{align*}
\]  

(3.15)

where \( \alpha_k \) and \( \beta_k \) denote the operators in the Schrödinger representation. It is assumed in this paper that an operator with no indication of the time dependence is expressed in the Schrödinger picture as a time-independent operator. From the inverse transformation of eq. (3.13), we obtain a time-evolution of the Schrödinger picture as a time-independent operator. From eq. (3.21), where the total number of sites is eight, and the number operators of excitons \( b_n^\dagger b_n \) are the independent operators in this case.

In order to investigate the coherent excitation dynamics of the system, we examine the time evolution of the expectation value of the dipole moment at an arbitrary QD \( n \), \( \langle P_n(t) \rangle \), under a variety of initial conditions. The expression given by eq. (3.18) and its Hermitian conjugate provide a useful result as

\[
\langle P_n(t) \rangle = \frac{1}{N} \sum_{k=1}^{N} \left( \langle P_m \rangle \cos \xi_{mm} + \langle V_m \rangle \sin \xi_{mm} \right) \cos \frac{L}{\hbar} t
\]

(3.21)

where the operator \( \langle \cdots \rangle = \text{Tr} \rho \cdots \) means the expectation value of an arbitrary operator. The notation \( \xi_{mn} = k(m - n) - (\epsilon + E)/2\hbar \) is used. Localized photons are assumed to be initially in the vacuum state. In particular, it is intriguing to investigate the dynamics with the initial condition of a locally excited state, i.e., how the initial excitation prepared only at one site propagates in the system. Setting \( \langle P_a(0) \rangle = \delta_{a1} \langle P_1 \rangle \) and \( \langle V_a(0) \rangle = 0 \) for all \( n \), we obtain from eq. (3.21) an explicit solution as

\[
\langle P_n(t) \rangle = \frac{1}{N} \sum_{n} \left( \cos \xi_{1n} \cos \frac{L}{\hbar} t - K \sin \xi_{1n} \cos \frac{L}{\hbar} t \right) \langle P_1 \rangle.
\]

(3.22)

In Fig. 3 we present one of numerical results of the time evolution of the dipole moment distribution calculated from eq. (3.22), where the total number of sites is eight, and the parameter values \( E = 2, \epsilon = 1, \) and \( V = 1 \) are used in a unit of a typical exciton energy in QD. Although the dynamics seems complicated at first sight, we can find a characteristic behavior that the dipole of the first site moves to the fifth site (the opposite site) and returns to the first site again. To see the behavior more precisely we show plots of the time evolution of the dipole moments of the first site and the fifth site in Figs. 4(a) and 4(b), respectively. The result clearly shows as a dominant behavior that when the dipole of the first site is active, that of the fifth site is inactive and vice versa, or the dipole moves like a seesaw between the first site and the fifth site. The recurrence behavior is reasonable from the fact that the Heisenberg equation of the system is linear in the boson approximation. Since the dipole moment \( P_n \) is quantum coherence between the ground and excited states of a two-level system, quantum coherence is transported to the farthest site of the chain and then comes back to the original site. When the total number of sites is odd, the seesaw motion arises between an initially excited site and its farthest pair of sites of the chain. An interesting point in this case is that we can obtain two copies of coherence at the farthest pair of sites as illustrated in Fig. 5(b). Moreover, it might be possible to use the system as a nanophotonic device of transporting or splitting quantum coherence.

4. Dynamics of Fermionic Excitons

If we rigorously adopt the commutation relation for excitons given by eq. (2.4), the Heisenberg equations for the atom system. However, it should be noted that the population difference \( W_n^\dagger W_n \) is always \(-1\) for bosonic excitons, and the number operators of excitons \( b_n^\dagger b_n \) are the independent operators in this case.
system read

\[ \dot{b}_n(t) = \frac{i}{\hbar} [H, b_n(t)] = -\frac{i}{\hbar} E b_n(t) + \frac{i}{\hbar} U a_n(t) W_n(t), \quad (4.1a) \]

\[ \dot{a}_n(t) = \frac{i}{\hbar} [H, a_n(t)] = -\frac{i}{\hbar} \varepsilon a_n(t) - \frac{i}{\hbar} V(a_{n+1}(t) + a_{n-1}(t)) - \frac{i}{\hbar} U b_n(t). \quad (4.1b) \]

Since the higher-order terms are produced by a mode-mode coupling such as \( a_n W_n = a_n (b_n^* b_n - b_n b_n^*) \), the equations become nonlinear and are hardly solved analytically. Thus we first solve the equations perturbatively in order to investigate the dynamics discussed in the preceding section.

4.1 Perturbative expansion of time evolution operator

Noticing that the Hamiltonian for localized photons \( H_a \) given by eq. (2.2a) can be written in a quadratic form as

\[ H_a = (a_1^+, \ldots, a_N^+) \begin{pmatrix} \varepsilon & V & V & \cdots & V \\ V & \varepsilon & V & \cdots & V \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ V & V & V & \varepsilon & \cdots \\ a_N \end{pmatrix} \begin{pmatrix} a_1 \\ \vdots \\ \vdots \\ a_N \end{pmatrix}, \quad (4.2) \]

\[ \equiv \sum_{n,m} a_n^+ R_{nm} a_m, \quad (4.3) \]

we obtain an orthogonal matrix \( P \) to diagonalize the matrix.
kernel $R$ of the Hamiltonian as
\[
(P^{-1}RP)_{ij} = \hbar \lambda \delta_{ij}.
\]
(4.4)
Then new annihilation operators $v_j = \sum_n P_{nj} a_n$ and creation operator $v^+_j = \sum_n P_{nj} a^+_n$ of new modes of localized photons are introduced. Since all the elements of the diagonalization matrix $P$ are not zero, note that the new modes are extended over a whole system. The commutation relation of the new modes can be given as
\[
[v_j, v^+_i] = \delta_{ij}.
\]
With the help of eq. (4.4) and the modes $v_j$, the Hamiltonian in eq. (4.3), or eq. (2.2a) is diagonalized as
\[
H = \sum_j \hbar \lambda_j v^+_j v_j,
\]
(4.5)
and the localized photon–exciton interaction $H_{\text{int}}$ in eq. (2.2c) is written in terms of $v_j$ as
\[
H_{\text{int}} = \hbar g \sum_n P_{nj} (v^+_j b_n + v_j b^+_n).
\]
(4.6)
In order to derive a perturbative expansion of the time evolution of an arbitrary operator, we define the Liouvillians as
\[
L \cdots = \frac{1}{\hbar} [H, \cdots], \quad L_0 \cdots = \frac{1}{\hbar} [H_0, \cdots],
\]
(4.7)
Then the Heisenberg equation of an arbitrary operator $O$ is written as
\[
\dot{O} = \frac{1}{\hbar} [H, O] = iLO,
\]
(4.8)
and we can obtain a formal solution as
\[
O(t) = G(t)O(0),
\]
(4.9)
with the time evolution operator $G(t) = e^{itL}$ that satisfies the following equation
\[
\dot{G}(t) = iLG(t) = iL_0 G(t) + iL_{\text{int}} G(t).
\]
(4.10)
Treating the interaction term $H_{\text{int}}$ as a perturbation, we solve the eq. (4.10) perturbatively up to the second order of the perturbation as
\[
G_D(t) = G_0(t) + \int_0^t G_0(t-s)L_{\text{int}} G_0(s)ds - \int_0^t ds \times \int_0^t ds '\n
\times \int_0^t ds '' \int_0^s dudv G_0(t-s)L_{\text{int}} G_0(s-u)L_{\text{int}} G_0(u),
\]
(4.11)
where the notation $G_0(t) = e^{itL_0}$ is used. Substituting eq. (4.11) into eq. (4.9) and using eq. (4.7), we can obtain the time evolution of the exciton operator $b_n(t) = G_D^2(t)b_n$. Suppose that localized photons are initially in the vacuum and $\langle V_n \rangle = 0$, then the expectation value of $b_n(t)$ is expressed
\[
\langle b_n(t) \rangle = e^{-i\Omega t} \langle b_n \rangle + g^2 \sum_{j=1}^N \sum_{m=1}^N \Gamma_j(t) P_{nj} P_{mj}
\times (1 - \delta_{nm}) \langle V_n \rangle \langle b_m \rangle - P^2_{nj} \langle b_n \rangle.
\]
(4.12)
where the notation
\[
\Gamma_j(t) = e^{-i\Omega t} \int_0^t ds e^{i\Omega (t-s)} \int_0^s ds ' e^{-i\Omega (s-u)}\langle b_m \rangle.
\]
(4.13)
is used, and $\langle b_n \rangle$ and $\langle d_n \rangle$ represent the real part and the imaginary part of $\Gamma_j(t)$, respectively. The expression given by eq. (4.12) and its Hermitian conjugate provide the time evolution of the expectation value of the dipole at an arbitrary QD site $n$ as
\[
\langle P_n(t) \rangle = \langle b_n \rangle \left( \cos \Omega t - g^2 \sum_j \langle b_j \rangle \langle P_{nj} \rangle \right)
\]
(4.14)
+ $g^2 \sum_{j \neq n} \langle b_j \rangle \langle P_{nj} \rangle / \langle W_n \rangle$. 

Corresponding to the discussion developed in §3.2, we set initially $\langle P_n \rangle = \delta_{n1}$ and obtain the perturbative solution that describes the dipole dynamics driven by a local excitation as follows:
\[
\langle P_n(t) \rangle = \delta_{n1} \left( \cos \Omega t - g^2 \sum_j \langle b_j \rangle \right) / \langle W_n \rangle
\]
(4.15)
+ $\langle W_n \rangle$. 

In the next section we discuss the dynamical properties of the system, on the basis of the solution given by eq. (4.15).

4.2 Numerical results and dynamical properties

As discussed in §3.2, we numerically investigate the dynamics of the dipole of the system depending on the initial conditions. Suppose that localized photons are initially in the vacuum, $\langle V_n \rangle = 0$ for all $n$, the dipole is initially set only at the first site as $\langle P_n \rangle = \delta_{n1}$, and all the populations are in the ground states except for the first site as $\langle W_n \rangle = -(1 - \delta_{n1})$. As mentioned before, all the population differences are automatically $-1$ for bosonic excitons. In addition, we impose that the length of the Bloch vector $\langle P_n, V_n, W_n \rangle$ is normalized as
\[
\langle P_n(t) \rangle^2 + \langle V_n(t) \rangle^2 + \langle W_n(t) \rangle^2 = 1.
\]
(4.16)

In Fig. 6 the time evolution of the dipole distribution is plotted. The parameter values used are the same as in §3.2 ($E = 2$, $\varepsilon = 1$, and $V = 1$). It follows from the figure that the dipole excitation of the first site moves to the opposite site as shown in the boson case (see Fig. 3). The reason is that the initial exciton density is so dilute as to validate the boson approximation. As time advances, the amplitude of each dipole increases because the perturbative solution violates the unitarity.

To find a new feature of fermionic excitons, we vary the initial conditions for the population differences from $\langle W_n \rangle = -(1 - \delta_{n1})$. Since the initial population differences at site $n$ contributes to the perturbative solution of the dipole at the same site, eq. (4.15), in a product of $\langle W_n \rangle$, the following hypothesis is proposed.

**Proposition 4.1 (Flip hypothesis).** If the sign of the initial population difference of the $n$-th site, $\langle W_n \rangle$, is inverted,
then the direction of the dipole moment of the n-th site at arbitrary time, $P_n(t)$, is flipped.

Since the dipoles at site 4, 5 and 6 in Fig. 6 are directed opposite to the others, we invert the sign of $W_n$ for $n = 4, 5, 6$. Figure 7 shows the result that the direction of the dipoles at site 4, 5, and 6 are flipped, and that all the dipoles oscillate with a same phase but with a different amplitude. Thus we observe that the system is transferred from a locally excited state to a coherently oscillating state of the dipoles, in other words, to a dipole-ordered state. In the ordered state the total dipole is $N$ times larger than each single dipole. Moreover, since radiation from an oscillating dipole is proportional to the square of the dipole moment, we can expect a high intensity of radiation from the dipole-ordered state. The radiation property of the system will be discussed in §6.2.

4.3 Dynamics of dipole-forbidden states via optical near-field interaction

It is well known that the electric interaction between molecules with no dipole moments is weak when they are separated in a macroscopic distance, and that the interaction becomes strong when they are very close to each other. Taking it into account, we investigate the dynamics of the system driven by localized photons, where it is interesting to consider an initial state that alternating dipoles are set and thus the total dipole of the system vanishes for an even number of sites. Such a dipole-forbidden state can be manipulated by localized photons, not by propagating far fields.

As an initial condition we set $P_n = (-1)^n$, and for simplicity, localized photons are in the vacuum, $V_n = 0$, and $W_n = 0$. The system parameters $E = 2$, $\varepsilon = 1$, and $V = 1$ are used as before. Figure 9 shows the time evolution of such a dipole-forbidden state. From Fig. 9 it follows that the system oscillates as schematically shown in Fig. 8(a), and that it remains in the initial dipole-forbidden state. This kind of dynamics is achieved in the system whose initial distribution of the population differences are uniform. For example, if we set $P_n = (-1)^n/\sqrt{2}$ and $W_n = 1/\sqrt{2}$ for

![Fig. 6. Time evolution of the dipole distributions ($E = 2$, $\varepsilon = 1$, $V = 1$). Initially all of the sites without the first site are in the ground state, while the dipole moment is initially set only at the first site.](image)

![Fig. 7. Time evolution of the dipole distribution ($E = 2$, $\varepsilon = 1$, $V = 1$): the initial population differences of site 4, 5 and 6 are inverted from $W_n = -(1 - \delta_n)$.](image)

![Fig. 8. Schematic illustration of (a) an alternating dipole distribution (a dipole-forbidden state) and (b) a dipole-ordered state. The state with alternating dipoles that result in the total dipole of zero cannot be coupled by the radiation field with the dipole interaction.](image)

![Fig. 9. Time evolution of a dipole-forbidden state (alternating dipole distribution). All the population differences are initially set as 0. We observe an oscillation of the alternating dipoles. The system remains in the dipole-forbidden state.](image)
all \( n \), the result is the same as shown in Fig. 9 except for its amplitude of the oscillation.

Next, we manipulate the distribution of the population difference non-uniformly, so that the signs are set as opposite to those of the corresponding dipoles:

\[
\langle P_n \rangle = (\ldots, +, -, +, +, +, +, +)/\sqrt{2},
\]
\[
\langle W_n \rangle = (\ldots, +, -, +, +, +, +, -)/\sqrt{2}.
\]

Figure 10 presents the result that a dipole-forbidden state as shown in Fig. 8(a) is converted to a dipole-ordered state as illustrated in Fig. 8(b). Since the dipole ordering has occurred by manipulating the initial distribution of the population differences, the result can be interpreted by the “Flip hypothesis” proposed in §4.2. As a collective oscillation of the dipoles occurred, the system evolved from a non-radiative state to a radiative state through the localized photon–exciton interaction.

### 4.4 Semi-classical approximation

Here we should note that the second-order perturbative solutions break the unitarity, and that the long-time behaviors of the dynamics given by the solutions are not necessarily same as the exact ones. Thus we have to return to the Heisenberg equations, eqs. (4.1a) and (4.1b), to evaluate the time evolution of physical observables. The Heisenberg equations are first solved by neglecting the quantum correlations between excitons and localized photons such as \( \langle W_n \rangle \langle y_n \rangle = \langle W_n \rangle \langle y_n \rangle \), and later the quantum correlations are estimated in §7.2. In this semi-classical approximation, eqs. (4.1a) and (4.1b) can be converted to the following coupled differential equations

\[
\begin{aligned}
\langle P_n \rangle &= -\Omega\langle V_n \rangle + g\langle W_n \rangle \langle y_n \rangle, \\
\langle V_n \rangle &= \Omega\langle P_n \rangle - g\langle W_n \rangle \langle x_n \rangle, \\
\langle W_n \rangle &= g(\langle V_n \rangle \langle x_n \rangle - \langle P_n \rangle \langle y_n \rangle), \\
\langle x_n \rangle &= -\omega (y_n) - v(\langle y_{n-1} \rangle + \langle y_{n+1} \rangle) - g(V_n), \\
\langle y_n \rangle &= \omega \langle x_n \rangle + v(\langle x_{n-1} \rangle + \langle x_{n+1} \rangle) + g(P_n),
\end{aligned}
\]

where the notations \( x_n = a_n + a_n^\dagger \), \( y_n = i(a_n - a_n^\dagger) \), and \( v = V/\hbar \) are used.

To check the reliability of the semi-classical approximation, we again investigate the system examined in §4.2. Figure 11 shows the time evolution of the dipole distribution obtained from the semi-classical approach for the system corresponding to Fig. 6. Comparing Fig. 11 with Fig. 6, we find that both profiles are same, and that the amplitude of each dipole obtained from the Heisenberg equations is less than 1, which means that the unitarity of the time evolution is conserved.

In Fig. 12, we show the time evolution of the dipole distribution obtained from the semi-classical approach when the initial population differences of site 4, 5, and 6 are inverted. It follows from the figure that each dipole is ordered as the flip hypothesis predicts, and that the flip hypothesis is still valid in the numerical solution of the semi-classical Heisenberg equation.

Numerical results obtained from eqs. (4.17) and (4.18) with a semi-classical approximation predict the dipole
dynamics with qualitative similarities as shown from the perturbative solution eq. (4.15). The flip hypothesis proposed in §4.2 is also verified by using the numerical solutions of eqs. (4.17) and (4.18), or of the Heisenberg equations without quantum correlations. These results show that the approach discussed in this section, maintaining an advantage to conserve the unitarity, qualitatively describe the dipole dynamics of the system in a similar way as the other two approaches. The validity of the approximation employed here, which we will examine in §7.2, is reported in the Dicke model that superradiant phenomena can be described with a semi-classical approximation which neglects quantum correlations among the atoms,\(^\text{12}\) and that the contribution from the quantum correlations is of the order of $1/N$ for large $N$.\(^\text{13}\)

5. Effective Hamiltonian and the Dipole Ordering

In order to investigate the origin of the dipole ordering discussed above, we first introduce an effective Hamiltonian to renormalize degrees of freedom of localized photons. Using the effective Hamiltonian, we then classify quasi-steady states with respect to the dipole distribution.

5.1 Effective Hamiltonian

An unitary transformation with an anti-Hermitian operator $S$ is applied to the Hamiltonian eq. (2.1) as\(^\text{32}\)

$$
\hat{H} = e^{-S} \hat{H} e^S = H + [H, S] + \frac{1}{2} [[H, S], S] + \cdots
$$

(5.1)

If the Hamiltonian eq. (2.1) is divided into the unperturbed part $H_0 = H_u + H_b$ and perturbed part $H_{\text{int}}$, and if $S$ is chosen to make

$$
H_{\text{int}} + [H_0, S] = 0,
$$

(5.2)

the terms linear in the coupling constant $g = U/\hbar$ vanish as

$$
\hat{H} = H_0 + \frac{1}{2} [H_0, S] + O(g^3).
$$

(5.3)

The transformed Hamiltonian $\hat{H}$ contains the terms in the second order of the exciton-localized photon interaction that describe exciton–exciton interactions via localized photons. Taking the expectation value of $\hat{H}$ in terms of the vacuum of localized photons $|\text{vac}\rangle$, we obtain the effective Hamiltonian $H_{\text{eff}}$ as

$$
H_{\text{eff}} = \langle \text{vac} | \hat{H} | \text{vac}\rangle = H_b + H_{b-b},
$$

$$
H_b = \sum_n \hbar \hbar \Delta b_n^\dagger b_n,
$$

$$
H_{b-b} = \sum_{n,m} \left( \frac{\hbar g^2}{\Omega l - R/\hbar} \right) _{nm} b_n^\dagger b_m,
$$

(5.4)

where the exciton energy $E$ is denoted as $\hbar \Omega$. With the help of the components of the Bloch vector $(P_n, V_n, W_n)$ the effective Hamiltonian eq. (5.4) can be rewritten as

$$
H_{\text{eff}} = \hbar \sum_n (\Omega + \Delta \Omega_n) \frac{1 + W_n}{2}
$$

$$
+ \frac{\hbar}{4} \sum_n \sum_{m \neq n} \Delta \Omega nm (P_n P_m + V_n V_m),
$$

(5.5)

where the interaction energy or coupling strength between excitons $\Delta \Omega nm$ are given as

$$
\Delta \Omega nm = \left( \frac{g^2}{\Omega l - R/\hbar} \right) _{nm} = \sum \frac{g^2}{\Omega - \lambda_j} P_n P_m,
$$

(5.6)

and the abbreviation $\Delta \Omega_n = \Delta \Omega nn$ is used for a special case.

5.2 Classification of quasi-steady states

Using the effective Hamiltonian $H_{\text{eff}}$, we can obtain the time evolution of the dipole moment at site $n$ in a similar way as discussed in §4.1:

$$
\langle P_n(t) \rangle = \langle P_n \rangle \cos(\Omega + \Delta \Omega_n t)]
$$

$$
+ \sum_{m \neq n} \Delta \Omega nm (W_n)(P_m) \sin(\Omega t).
$$

(5.7)

If the dipole at site 1, in particular, is only excited at $t = 0$, eq. (5.7) reads

$$
\langle P_n(t) \rangle = \delta_{n1} \langle P_1 \rangle \cos(\Omega + \Delta \Omega_1 t)
$$

$$
+ (1 - \delta_{n1}) \Delta \Omega nm (W_n)(P_1) \sin(\Omega t).
$$

(5.8)

It follows from this expression that the sign of the dipole at site $n$ depends on the coefficient $\Delta \Omega_{n1}$ as well as the initial values of $(P_1(0))$ and $(W_n(0))$, and that the coefficient $\Delta \Omega_{n1}$ determines what kind of quasi-steady states of the dipole distribution the system reaches when the initial values are fixed. Therefore we examine the matrix $\Delta \Omega$ to classify the quasi-steady states of the dipole distribution of the system.

First, as an example, we show the matrix elements of $\Delta \Omega$ for the system examined in §3 and §4 whose material parameters are $E = 2$, $\varepsilon = 1$, and $V = 1$ as follows:

$$
\Delta \Omega = \left( \begin{array}{cccc}
\frac{1}{2} & \frac{1}{2} & \frac{1}{2} & \frac{1}{2} \\
\frac{1}{2} & \frac{1}{2} & \frac{1}{2} & \frac{1}{2} \\
\frac{1}{2} & \frac{1}{2} & \frac{1}{2} & \frac{1}{2} \\
\frac{1}{2} & \frac{1}{2} & \frac{1}{2} & \frac{1}{2} \\
\end{array} \right)
$$

The matrix is symmetric and has a cyclic structure as

$$
\Delta \Omega_{n+1,m+1} = \Delta \Omega_{n,m}.
$$

This is due to the fact that the periodic boundary condition is imposed on the system, and that the Hamiltonian given in eq. (2.1) has translational invariance. Owing to the symmetric structure as

$$
(\Delta \Omega_{22}, \Delta \Omega_{23}, \Delta \Omega_{24}, \ldots) = (\Delta \Omega_{11}, \Delta \Omega_{12}, \Delta \Omega_{13}, \ldots),
$$

the dipole distribution of the system initiated by the dipole excitation at site 2 can be discussed in terms of $\Delta \Omega_{11}$ that governs the problem driven by the site-one excitation. In general the problem is reduced to examine the structure of the coefficients $\Delta \Omega_{1n}$ that depend on the material parameters $\Omega$ and $\lambda_j$, or on $E$, $\varepsilon$, and $V$. Since the parameter sets crossing the resonant points $(\Omega = \lambda_j)$ change the pattern of the dipole distribution in the system, we can classify quasi-steady states of the dipole distribution into four groups in Table I. The system with $E = 2$, $\varepsilon = 1$, and $V = 1$, which has been
investigated in §4, belongs to the first group. Since the coefficients \( \Delta \Omega_{1n} \) in the first group have different signs at site 4, 5, and 6 from others, the quasi-steady state of the dipole distribution that is predicted by the effective Hamiltonian is the one described by the second-order perturbation solution [see eq. (4.15) and Fig. 6]. The third and fourth groups are, in particular, interesting among the four groups of the quasi-steady states. The third group corresponds to a “ferromagnetic” system, in which all the electric dipole moments are aligned to the same direction, and a dipole-ordered state appears.

Using numerical results of eqs. (4.17) and (4.18), we examine the dynamics of the third and fourth groups whether they behave as predicted by the effective Hamiltonian: “ferromagnetic,” or “anti-ferromagnetic.” The parameter set of \( E = 1, \varepsilon = 3.01, \) and \( V = 1 \) gives

\[
\Delta \Omega_{1n} = (-13.1, -12.7, -12.4, -12.2, -12.2, -12.2, -12.4, -12.7),
\]

and thus the system belongs to the third group. The results is shown in Fig. 13, which predicts the “ferromagnetic” behavior as expected. When the parameter set of \( E = 1, \varepsilon = 3.01, \) and \( V = 1 \) is chosen, the system corresponds to the fourth group. Figure 14 presents the time evolution of the dipole distribution of the system that shows alternating changes in signs, as expected to be similar to anti-ferromagnetics.

With the help of the flip hypothesis proposed in §4.2, we infer that the distribution of the dipoles can be changed by locally manipulating the initial population differences, and that an “anti-ferromagnetic” state can be transformed into a dipole-ordered state. To verify it, the initial population differences are set in the zigzag form as \( W_n = (0, -, +, -, +, +, +, -) \). According to the flip hypothesis, the alternating dipoles change their directions and are aligned to form a dipole-ordered state.

Table 1. Classification of quasi-steady states.

<table>
<thead>
<tr>
<th>Group</th>
<th>Signs of ( \Delta \Omega_{1n} ) for ( n = 1, 2, 3, \ldots, 8 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st</td>
<td>((-,-,-,+,-,-,-,+)), ((+,-,+,-,+,-,+,-))</td>
</tr>
<tr>
<td>2nd</td>
<td>((-,-,-,+,-,-,-,+)), ((+,-,+,-,+,-,+,-))</td>
</tr>
<tr>
<td>3rd</td>
<td>((-,+,-,+,-,+,-,+)), ((+,-,+,-,+,-,+,-))</td>
</tr>
<tr>
<td>4th</td>
<td>((-,+,-,+,-,+,-,+)), ((+,-,+,-,+,-,+,-))</td>
</tr>
</tbody>
</table>

radiation from the Dipole-Ordered States

In this section we examine the radiation property of the dipole-ordered states discussed in §4.2 and §5.2. Our model has excluded radiation fields or free photons from the preceding discussions about the dynamics because the interaction energies of excitons with optical near fields are...
which are simultaneous eigenstates of both and determines the collectivity of cooperative phenomena.

6.1 Dicke states and superradiance

The components of the Bloch vector of a two-level system as a representative of a collection of atoms or quantum dots, \((P_n, V_n, W_n)\), are defined by Pauli’s spin operators as

\[
R_i^{(n)} = \frac{1}{2}(b_n + b_n^\dagger) = \frac{1}{2} P_n, \\
R_2^{(n)} = \frac{1}{2}(b_n - b_n^\dagger) = \frac{1}{2} V_n, \\
R_3^{(n)} = \frac{1}{\sqrt{2}}(b_n^\dagger b_n - b_n b_n^\dagger) = \frac{1}{2} W_n,
\]

where the spin operators \(R_i^{(n)}\) satisfy the commutation relations

\[
\left[ R_i^{(n)}, R_j^{(n)} \right] = i\epsilon_{ijk} R_k^{(n)},
\]

with Levi–Civita’s symbol \(\epsilon_{ijk}\). For an \(N\)-two-level system we introduce the total spin operators as

\[
R_1 = \sum_{n=1}^{N} R_1^{(n)}, \quad R_2 = \sum_{n=1}^{N} R_2^{(n)}, \quad R_3 = \sum_{n=1}^{N} R_3^{(n)},
\]

which obey the commutation relations

\[
[R_i, R_j] = i\epsilon_{ijk} R_k.
\]

Let us consider a state in which \(N_1\) of the \(N\) two-level system are in the lower state \([1]\) and \(N_2\) are in the excited state \([2]\), where

\[
N = N_1 + N_2, \quad m = \frac{1}{2}(N_2 - N_1).
\]

Evidently \(m\) is a measure of the total population inversion, and it is an eigenvalue of the spin operator \(R_3\). Introducing the operator \(R^2 = R_1^2 + R_2^2 + R_3^2\) and using eq. (6.4), we obtain the commutation relation

\[
[R_i, R^2] = 0, \quad i = 1, 2, 3,
\]

so that the collective operators \(R_i\) obey the same algebra as the angular momentum. Thus there exist the states \([l, m]\) which are simultaneous eigenstates of both \(R_i^2\) and \(R^2\) as

\[
R_3[l, m] = m[l, m], \quad \text{for } -\frac{1}{2}N \leq m \leq \frac{1}{2}N, \quad (6.7a)
\]

\[
R_2^2[l, m] = l(l + 1)[l, m], \quad \text{for } |m| \leq l \leq \frac{1}{2}N. \quad (6.7b)
\]

The quantum number \(l\) is called the cooperation number \(^8\) and determines the collectivity of cooperative phenomena. For the raising and lowering operators as

\[
R_+ = \sum_{n=1}^{N} b_n = R_1 + iR_2, \quad R_- = \sum_{n=1}^{N} b_n = R_1 - iR_2,
\]

it follows from the well-known properties of angular momentum operators that

\[
R_{\pm}[l, m] = \sqrt{(l \pm m)(l \pm m + 1)}[l, m \pm 1], \quad (6.9)
\]

where the collective states \([l, m]\) are called Dicke states.

Here we suppose a transition from an initial state \([l, m]|\text{vac}\rangle\) to a final state \(|\psi\rangle\), where \([l, m]\) and \(|\text{vac}\rangle\) represent a Dicke state and the vacuum of the radiation field, respectively. Adopting the dipole interaction between the two-level system and radiation field as

\[
H_I(t) = -\mu_{\text{tot}}(t) \cdot E(t)
\]

with the total electric dipole of the system

\[
\mu_{\text{tot}} = \mu \sum_{n=1}^{N}(b_n + b_n^\dagger) = \mu(R_+ + R_-),
\]

we can write the transition probability as

\[
-\langle\psi|\mu \cdot E_{\ldots}(t)R_-(t)|l, m|\text{vac}\rangle^2
\]

\[
= \langle R_+(t)R_-(t)|A = (l + m)(l + m + 1)A,
\]

where eq. (6.9) is used for the final expression, and \(A = \langle\text{vac}|\mu \cdot E_{\ldots}(t)\mu \cdot E_{\ldots}(t)|\text{vac}\rangle\) is identified as the Einstein A-coefficient for each two-level system like an atom or a quantum dot.

If all quantum dots are in the ground state \([1]\), then \(m = -N/2\), and from eq. (6.7b) we must have \(l = N/2\). Hence, from eq. (6.12) the radiation rate \(w_{\text{emit}}\) is zero as expected. If all quantum dots are in the excited states \([2]\), then \(m = N/2\) and we again have \(l = N/2\). Hence \(w_{\text{emit}} = NA\), which is just what one would expect from a group of \(N\) independently radiating quantum dots. However, the situation is quite different if the initial state is not the completely excited state. Let us consider a state in which half of the quantum dots are excited and half are not, so that \(m = 0\). Then we have

\[
w_{\text{emit}} = l(l + 1)A,
\]

and \(l\) can have any value between \(0\) and \(N/2\). The larger the value of \(l\), the larger is collective rate of radiation of the system. In particular, if \(l = N/2\), then

\[
w_{\text{emit}} = \frac{1}{4} N(N + 2)A,
\]

which maximizes the value of cooperative spontaneous radiation. Since the maximum radiation probability is proportional to the square of the site number \(N\), it becomes very large for large \(N\). This phenomenon is called superradiance, and the state \([l = N/2, m = 0]\) is called a superradiant Dicke state. From the definition of the total Bloch vector \((P, V, W)\) as \(P = \sum_n(P_n), V = \sum_n(V_n), W = \sum_n(W_n)\), it follows that the Bloch vector for the superradiant state is on the \(P-V\) plane and its length reaches a maximum, as shown in Fig. 16.
6.2 Radiation property of the dipole-ordered states

According to the formulation developed in §6.1, we now investigate the radiation property of the dipole-ordered states discussed in §4.2 and §5.2. From the expression eq. (6.12) the radiation intensity at time $t$ is determined by the radiation factor, $\langle R_+ R_- \rangle = (l + m)(l - m + 1)$, i.e., the expectation value of the operator

$$ R_+ R_- = R_1^2 + R_2^2 + R_3 = \left( \frac{P}{2} \right)^2 + \left( \frac{V}{2} \right)^2 + \frac{W}{2}. $$

(6.15)

which indicates that there are two elements mainly contributing to the radiation factor; one is the collectiveness of the system measured by the cooperation number $l$, and the other is the total energy of the quantum dot system given by $\langle R_3 \rangle = m$. Since the dynamics of the system is driven in our system by localized photons, the evolution of the radiation factor is also described in terms of $l(t)$ and $m(t)$ that are developed according to the localized photon–exciton interaction.

Numerical results of eqs. (4.17) and (4.18) are shown in Figs. 17 and 18. The upper parts in Fig. 17 show the time evolution of the radiation factors, while the lower parts illustrate the dipole distribution when each radiation factor has a maximal value. The total dipole in Fig. 17(a) is smaller than those in Figs. 17(b) and 17(c), and the peak value of the radiation factor is also smaller. It follows from the figure that the radiation factor increases as the total dipole becomes larger. The peak values of the radiation factor in Figs. 17(b) and 17(c), 15 and 14, correspond to the value for the Dicke’s superradiance, which is obtained as 20 for $N = 8$ from eq. (6.14). We thus expect that quasi-steady states shown in Figs. 17(b) and 17(c) are close to the superradiant states, and that the total Bloch vectors for such states are on the $P—V$ plane. In order to check whether the total Bloch vectors for the states belonging to the third group in Table I [see Fig.

![Fig. 16. Total Bloch vector in a superradiant state. It is on the $P–V$ plane, and its length has a maximal value.](image)

![Fig. 17. Time evolution of the radiation factor $\langle R_+ R_- \rangle$ (upper) and the dipole distribution at time indicated by the arrows when the radiation factors take maximal values (lower) for (a) a system belonging to the first group with an initial population difference $W_0 = (0++---+++)$, (b) a system belonging to the third group with an initial population difference $W_0 = (0+-----+++)$, and (c) a system belonging to the fourth group with an initial population difference $W_0 = (0---+++--)$](image)

![Fig. 18. Time evolution of the radiation factor (upper) and the polar angle of the total Bloch vector (lower) for a system belonging to the third group. The polar angle is measured from the $-W$ axis.](image)
17(b)] are on the P–V plane or not, we examine the time evolution of the polar angles of the Bloch vectors as well as the radiation factors. As shown in Fig. 18, the polar angle of the Bloch vector takes 90°, that is, are on the P–V plane when the radiation factor has a maximal value. Therefore we conclude that the system belonging to the third group in Table I are in transition to a quasi-steady state close to the superradiant Dicke state, judging from its large radiation factor and the polar angle of the total Bloch vector. Figures 17(b) and 17(c) indicate that multiple peaks appear in the radiation, or that multiple pulses are emitted from the system. One may think, as a possible origin, that such a phenomenon stems from the recurrence inherent in an isolated system. However, such multiple pulses may survive even if the system becomes dissipative, which will be examined in detail in the next section.

7. Radiation from a Dissipative System

We have assumed in the previous section that the radiation field is so weak as not to disturb the exciton dynamics of a quantum dot system. When radiation pulses are emitted from the system, however, energy has to be dissipated, and it is interesting but not clear whether multiple pulses shown in Figs. 17(b) and 17(c) are emitted from the system or not. In this section we thus study the radiation profile, adding a radiation field to the system as a reservoir that does not affect the dynamics but makes the system a dissipative one.

Suppose the Hamiltonian

\[ H_2 = H_{Q\text{Deff}} + H_F + H_{\text{Fin}}, \]

(7.1)

where \( H_{Q\text{Deff}} \) is the effective Hamiltonian given by eq. (5.4) that describes the \( N \)-two-level quantum dot system interacting with localized photons. The Hamiltonians \( H_F \) and \( H_{\text{Fin}} \) describe the free radiation field and the exciton-free photon interaction, respectively. Explicit Hamiltonians in the interaction representation are written as

\[ H_F = \sum_{k,l} \hbar \omega_k a_{k\lambda}^\dagger a_{k\lambda}, \]

(7.2)

\[ H_{\text{Fin}}(t) = \sum_{k,l} \hbar g_{k\lambda} a_{k\lambda}^\dagger R_+ e^{i(\omega_k - \Omega)t} + \text{h.c.} \]

(7.3)

where creation and annihilation operators of a free photon with wave vector \( k \), polarization \( \lambda \), and frequency \( \omega_k \) are denoted as \( a_{k\lambda}^\dagger \) and \( a_{k\lambda} \), respectively. The coupling constant between the free photon and exciton is given as

\[ g_{k\lambda} = \frac{i\Omega}{\hbar \sqrt{V}} \sqrt{\frac{\hbar}{2 \omega_k \epsilon_0}} \cdot \epsilon_{k\lambda}, \]

(7.4)

where \( V \), \( \epsilon_{k\lambda} \), and \( \epsilon_0 \) represent the quantization volume of the radiation field, the unit polarization vector, and the dielectric constant in vacuum, respectively. Using the Hamiltonian \( H_2 \) and the density operator \( \hat{\rho}(t) \), we write the Liouville equation as

\[ \frac{\partial \hat{\rho}(t)}{\partial t} = \frac{1}{\hbar} [H_2, \hat{\rho}(t)] = -iL_2 \hat{\rho}(t), \]

(7.5)

and eliminate degrees of freedom of the radiation field with the help of a projection operator defined as

\[ P \cdots = |0\rangle\langle 0| \text{Tr} \cdots \]

(7.6)

with the vacuum of the radiation field \( |0\rangle \). Then, we obtain equations of motion for the density operator \( \hat{\rho}(t) \) as follows:

\[ \frac{\partial \hat{\rho}(t)}{\partial t} = -i[\hat{P}L_2(t)\hat{\rho}(t) - \hat{P}L_2(t)U(t, 0)(1 - \hat{P})\hat{\rho}(0)] \]

\[ - \hat{P}L_2(t) \int_0^t \text{d}t' U(t, t') (1 - \hat{P})L_2(t') \hat{P}(t'), \]

(7.7)

where \( L_2(t) \) is the Liouville operator associated with \( H_2(t) \), and the operator \( U(t, t') \) is defined as

\[ U(t, t') = \exp \left( -i(1 - \hat{P}) \int_t^{t'} L_2(t') \text{d}t' \right). \]

(7.8)

With the Born–Markov approximation applied to the third term of eq. (7.7), we obtain the following equation for \( \hat{\rho}_\lambda \) as

\[ \frac{\partial \hat{\rho}_\lambda}{\partial t} = -iL_{Q\text{Deff}}(t)\hat{\rho}_\lambda(t) - \text{Tr}_F L_{\text{Fin}}(t) \]

\[ \times \int_0^t \text{d}t' U_{Q\text{Deff}}(t - t')L_{\text{Fin}}(t - t')(0\rangle\langle 0|\hat{\rho}_\lambda(t')) \]

(7.9)

with

\[ U_{Q\text{Deff}}(t) = \exp (-iL_{Q\text{Deff}}), \]

(7.10)

where \( L_{Q\text{Deff}} \) and \( L_{\text{Fin}} \) are the Liouville operators associated with the Hamiltonian \( H_{Q\text{Deff}} \) and \( H_{\text{Fin}} \), respectively. Moreover, using the Born approximation that neglects the exciton operators of higher than the second order, \(^{31}\) we approximate

\[ U_{Q\text{Deff}}(t) \sim 1 \]

(7.11)

to obtain a compact equation as

\[ \frac{\partial \hat{\rho}_\lambda}{\partial t} = -iL_{Q\text{Deff}}(t)\hat{\rho}_\lambda(t) + \beta \{ [R_+ \hat{\rho}_\lambda(t), R_-] + [R_-, \hat{\rho}_\lambda(t)R_+] \} - i\gamma [R_+R_-, \hat{\rho}_\lambda(t)] \]

(7.12)

with

\[ \beta + i\gamma = \int_0^t \sum_{k,l} |g_{k\lambda}|^2 e^{i(\Omega - \omega_k)t} \text{d}t, \]

(7.13)

which is exactly the same form as the Lindblad’s master equation\(^ {33}\) describing a general Markovian dynamics for a dissipative quantum system. Here the real and imaginary parts of the right hand side of eq. (7.13) are designated as \( \beta \) and \( \gamma \), respectively. In the following we neglect the energy shift as \( \gamma = 0 \), for simplicity. Note that the second and the third terms of the right hand side of eq. (7.12) are known as the Dicke’s master equation.\(^ {12,15,17}\)

7.1 Semi-classical description with the effective Hamiltonian

Neglecting quantum correlation between excitons,\(^ {13}\) we approximate the total density operator \( \rho_\lambda \) as a direct product of the density operator \( \rho_n \) at each site \( n \)

\[ \rho_\lambda = \prod_n \rho_n, \]

(7.14)

and then solve eq. (7.12). Noticing that the dynamics governed by the original Hamiltonian eq. (2.1) is not rigorously identical to the one described by the effective
Hamiltonian eq. (5.4), we use an isolated system described by the effective Hamiltonian eq. (5.4) in order to clarify the dynamics governed by eq. (7.12) for a dissipative system whose relevant system is described by the same Hamiltonian eq. (5.4). By comparing the radiation factors for the isolated system and the dissipative system, the similarity and the difference are discussed.

Figure 19 shows the time evolution of the radiation factor for (a) a “ferromagnetic” system belonging to the third group in Table I, (b) an “anti-ferromagnetic” system that belongs to the fourth group in Table I and is turned to a dipole-ordered state after manipulating the initial population differences, and (c) a “dipole-forbidden” case discussed in §4.3. The parameters \( \beta = 0.05 \) and \( \gamma = 0 \) are used.

7.2 Quantum correlations

It is well known that superradiance in the Dicke model occurs from a state where all excited states of all sites are occupied. On the other hand, the semi-classical approach discussed above cannot predict the occurrence of superradiance of the system when the total dipole of the system is zero as an initial condition. This means that quantum fluctuations and correlations should be properly included so as to correctly describe the radiation properties of a system with no initial dipoles, and that the semi-classical approximation is not appropriate in this case. Thus we numerically solve the master equation (7.12), taking quantum correlations into account, and we investigate the radiation properties of the dissipative system. In particular, we compare the results obtained from our model, i.e., the localized photon model with those obtained from the Dicke model, for which the first term of the right hand side is dropped from eq. (7.12). Some of such results are shown in Figs. 20 and 21.

Figure 20(a) shows the time evolution of the radiation factor for the case that all the populations are completely in the excited states and there are no dipoles as an initial condition. The solid curves are the results for our model while the dashed curves represent the results for the Dicke model. The dissipative system is assumed, and the parameters \( N = 4 \), \( \beta = 0.05 \), and \( \gamma = 0 \) are used. In addition, the following initial conditions are used: (a) \( (P_n) = 1 \) and \( (W_n) = 1 \), (b) \( (P_n) = (-1)^n/\sqrt{2} \) and \( (W_n) = 1/\sqrt{2} \) which correspond to a dipole-forbidden state as shown in Fig. 10, and (c) \( (P_n) = (-1)^n/\sqrt{2} = -(W_n) \) which corresponds to a dipole-ordered state as shown in Fig. 10.
A collective dipole oscillation via localized photon–exciton originates from the occurrence of a dipole-ordered state or difference between the two models, as we expected, superradiantly from the dissipative system. Therefore the factor increases, and thus multiple pulses are emitted strongly. As a result, the oscillation frequency of the radiation as in each QD is set opposite to that of the corresponding dipole for the case that initially the alternating dipole is set as $g_0 = 0$. It is found that the frequency of the oscillation is different. It indicates that the semi-classical approach can describe qualitatively the radiation properties of both isolated and dissipative systems when the total dipole of the system is not zero. The strong radiation coming from the dipole-ordering phenomenon, or the non-linearity and the collective phenomena of the dynamics of the system considered in this paper can be qualitatively predicted by the semi-classical approach.

8. Conclusions

In order to investigate the dynamics of the system, we
presented a model of an $N$ two-level quantum dot system interacting with optical near fields, explicitly expressing localized photonic degrees of freedom that characterize the unique property of localization of optical near fields. In the low density limit of excitons ($N$ two-level system) are approximately treated as bosons, and a rigorous solution of the Heisenberg equation is obtained. Using the solution, we have examined the dynamics of the excitonic system to show that the dipole moments linearly propagate in the system. Since the dipole moments in the system represent the quantum coherence between any two levels, this phenomenon might be applied to a photonic device on a nanometer scale, or transportation of quantum information.

For fermionic excitons, the Heisenberg equation becomes nonlinear, and the dynamics is more complicated. We obtained a perturbative solution given by eq. (4.14) within the second order with respect to the localized photon–exciton interaction, to investigate the dipole dynamics. The study revealed that there exist several oscillating quasi-steady states depending on the material parameters. Using the effective Hamiltonian obtained from renormalization of localized photonic degrees of freedom, we classified such quasi-steady states into several groups, some of which are a “ferromagnetic” state with all the dipoles aligned to the same direction, and an “anti-ferromagnetic” state with the alternating dipoles, as shown in Table I.

Here it should be noted that if the sign of the population difference ($W_n(0)$) at arbitrary site $n$ and at time $t = 0$ is inverted, then the dipole of the same site at arbitrary time $t$, ($P_n(t)$), also changes the sign [see eq. (4.15)]. Using this flip hypothesis, we can transform an arbitrary dipole distribution of the system into a dipole-ordered state after manipulating the initial distribution of the population differences. This hypothesis is based on the perturbative solution eq. (4.15) that determines the sign of ($P_n(t)$) according to the sign of the product of ($P_r(0)$)($W_r(0)$). It also originates from the fermionic property of excitons, which gives Heisenberg equations of motion for $b_n$ ($\propto P_n$) as

$$[H_{b\cdots b}, b_n] = \sum_{m \neq n} \Delta \Omega_{nm} W_m b_m.$$  

(8.1)

The right hand side of eq. (8.1) is proportional to $\langle P_n \rangle (W_n)$ for fermions while it is proportional to $\langle P_m \rangle$ for bosons. Therefore the occurrence of this kind of nonlinearity for fermionic excitons is a possible origin of the flip hypothesis.

Solving the Heisenberg equations (4.17) and (4.18) with the semi-classical approximation, we examined the radiation property of our system as an isolated system. It was found from numerical analysis that dipole-ordered states, which have large total dipole moments, show a large radiation probability comparable to Dicke’s superradiance. In particular, it predicted that multiple pulses are superradiantly emitted from the system. In order to verify whether such a phenomenon is inherent in an isolated system or not, we solved master equations (7.12) for a dissipative system with the semi-classical approximation, and found that such multiple pulses in the radiation profile can survive even in a dissipative system coupled to a radiation reservoir. Multiple superradiant peaks have been experimentally observed in an atomic gas system,\(^1\) and the origin has been reported as the dipole–dipole interaction between a two-level system within the framework of the Dicke model.\(^1\) Since excitons in our model are indirectly coupled with one another through the interactions with localized photons, it may be concluded that the pulse oscillation in the radiation profile occurs in a similar mechanism as that of the Dicke model.

Finally we solved master equation (7.12), taking quantum correlations into account, and compared the results with those obtained from the Dicke model. When all the populations are initially in the excited states, similar radiation profiles for both models are obtained. The qualitative difference is that the peak value of the radiation pulse in our model is reduced and the tail is extended. This tendency has been examined from comparison between the atomic system and the Frenkel exciton system, where excitons can propagate via the dipole–dipole interaction, and the same qualitative difference as ours has been reported.\(^2\) Regarding to the multiple-pulse generation which we have obtained, the Frenkel exciton model has also predicted a possibility that the superradiance profile oscillates if the system is initially prepared with a partial population inversion.\(^2\) We concluded from our modeling that the superradiant peaks of multiple pulses correspond to the occurrence of a collective dipole oscillation, or a dipole-ordered state.

It is an open question why the dipole distribution of our system has several quasi-steady states, which was predicted in this paper by using the perturbative expressions, numerical solutions of the Heisenberg equations, and the effective Hamiltonian. Since our system considered here has several kinds of symmetries, we expect that the stability of such quasi-steady states is clarified from the viewpoint of symmetry. The size dependence of the radiation profile has been investigated in the Dicke model.\(^3\) In order to clarify the differences between optical near fields and propagating fields, it is interesting to examine such size dependence in our model. The semi-classical approach may be useful for such a qualitative study because it has an advantage over the quantum approach with respect to the computational time. One of our main goals was to clarify the inherent characteristics in optical near fields from the viewpoint of the coupling scheme: a local coupling system and a global coupling system. We have shown several differences between our model (local coupling system) and the Dicke model (global coupling system), but we require further efforts to answer the question more directly.

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