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Superradiance of Frenkel excitons with any degree of excitation prepared by a short-pulse laser

Tetsuji Tokihiro, Yoshihiro Manabe, and Eiichi Hanamura
Department of Applied Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113, Japan

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Collective spontaneous emission from a one-dimensional mesoscopic exciton system interacting via static dipole moments is investigated by virtue of the analogy to the quantum XXZ spin chain. The rigorous expression for the initial state is obtained at any degree of excitation from the ground state by a short intense laser pulse. The emission profile from the initial state with partial population inversion exhibits oscillations and very rapid decay in comparison with that from the completely inverted state. The time-resolved emission spectra are also discussed.

I. INTRODUCTION

When a collection of dense atoms (or molecules) are prepared in the upper level of an electric transition, it starts to radiate spontaneously and coherently with much shorter-pulse width than the emission of independent atoms. This phenomenon is called superradiance[1] and has been studied both experimentally[2,3] and theoretically[4,5] since the pioneering work of Dicke.[1] He investigated the spontaneous emission from an ideal gas system composed of N two-level atoms and predicted this effect as the collective spontaneous emission due to a spontaneous phase locking of atomic dipole moments throughout the medium as long as the system size is small comparing with the relevant wavelength of the radiation. It is a quantum mechanical phenomenon in the sense that its intermediate transient states are described by the superpositions of the classical states of individual excitations. This is easily seen from the collective emission of two particles. We denote the upper (lower) level of the ith atom by $|e_i\rangle$ ($|g_i\rangle$). The two-particle excited state is expressed as $|e_1\rangle \otimes |e_2\rangle$. Classically it decays into either $|g_1\rangle \otimes |e_2\rangle$ or $|e_1\rangle \otimes |g_2\rangle$, while quantum mechanically any superposition of these two states is admissible and, in fact, it decays into the symmetric state: $\frac{1}{\sqrt{2}}$ $(|g_1\rangle \otimes |e_2\rangle + |e_1\rangle \otimes |g_2\rangle)$.

In order to describe the N particle (two-level atomic) system, it is convenient to use the analogy between a two-level atom and a quantum spin, that is, we regard the upper state of the ith atom $|e_i\rangle$ as the spin-up state $|\uparrow\rangle$ of the ith 1/2 quantum spin, and $|g_i\rangle$ as $|\downarrow\rangle$. Correspondingly, if we define the (pseudo-)spin operators,

$$\hat{s}_i^+ \equiv |e_i\rangle\langle g_i|, \quad \hat{s}_i^- \equiv |g_i\rangle\langle e_i|, \tag{1}$$

$$\hat{s}_i^z \equiv \frac{1}{2}(|e_i\rangle\langle e_i| - |g_i\rangle\langle g_i|), \tag{2}$$

they satisfy the usual commutation relations as

$$[\hat{s}_i^+, \hat{s}_j^-] = 2\delta_{i,j}\hat{s}_j^z, \quad [\hat{s}_i^z, \hat{s}_j^\pm] = \pm\delta_{i,j}\hat{s}_j^\pm. \tag{3}$$

The system Hamiltonian of the two-level atoms H_0 and the interaction Hamiltonian $H_{\rm int}$ are described with these operators as

$$H_0 = \sum_{j=1}^{N} \hbar\omega_0 \hat{s}_j^z \equiv \hbar\omega_0 \hat{S}^z,$$

$$\begin{aligned} H_{\rm int} &= -\sum_{j=1}^{N} (\mathbf{E}\cdot\boldsymbol{\mu}\, \hat{s}_j^+ + \mathbf{E}^\dagger\cdot\boldsymbol{\mu}^*\, \hat{s}_j^-) \\ &\equiv -\mathbf{E}\cdot\boldsymbol{\mu}\, \hat{S}^+ - \mathbf{E}^\dagger\cdot\boldsymbol{\mu}^*\, \hat{S}^-, \end{aligned} \tag{4}$$

where $\mathbf{E}$ is the electric field operator and $\boldsymbol{\mu}$ denotes the expectation value of the atomic dipole moment. If the initial state is in complete-population inversion, it is given by

$$\begin{aligned} |\Phi_0\rangle &= |\uparrow\rangle_1 \otimes |\uparrow\rangle_2 \otimes \cdots \otimes |\uparrow\rangle_N, \\ &= \left| S = \frac{N}{2}; M = \frac{N}{2} \right\rangle, \end{aligned}$$

where $S(S+1)$ and M are the quantum numbers of the total spin operator $\hat{S}^2 \equiv \frac{1}{2}(\hat{S}^+\hat{S}^- + \hat{S}^-\hat{S}^+) + (\hat{S}^z)^2$ and $\hat{S}^z$, respectively. Since $\hat{S}^2$ commutes with both H_0 and $H_{\rm int}$, the quantum number S, which is called the cooperation number, is a conserved quantity during the superradiant process so that the system evolves in the $N+1$ eigenstates: $|\frac{N}{2}; \frac{N}{2}\rangle, |\frac{N}{2}; \frac{N}{2}-1\rangle, \ldots, |\frac{N}{2}; -\frac{N}{2}\rangle$. The emission rate W_N is given with the spontaneous emission rate of an atom γ by

$$W_N = \gamma\langle \hat{S}^+\hat{S}^- \rangle,$$

which scales as $\sim N$ at the initial and final stage, while it scales as $\sim N^2$ at $M \simeq 0$. This squared N dependence of the radiation rate results in the "ideal" superradiance.

In realistic systems, however, the interaction between constituent atoms suppresses the superradiant processes.[6] Dipole-dipole interactions cause the dephasing of the symmetric states and frequency chirping of the emission light.[7–9] In the spin language, the total spin S is no longer a conserved quantity and smaller spin

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states participate in the superradiance. Frenkel exciton is the quantum for the coherent propagation of an excitation in an insulating or a molecular crystal. The excitation transfer is induced by the dipole-dipole interaction between the transition dipole moments of the molecules. Since the excitation (electron-hole pair) is localized at the molecular site and has no inner structure in contrast to Wannier exciton, we can adopt the system of two-level atoms with strong dipole-dipole interaction for the model of Frenkel excitons by regarding the constituent molecules as two-level atoms. Thus, the system of Frenkel excitons under strong excitation is equivalent to the two-level atomic system with population inversion and it will exhibit superradiance. This superradiance of Frenkel excitons in a linear system from the complete-population inversion has been investigated in detail in Refs. 7 and 8. Here, frequency chirping and slow emission tails due to the excitation transfer have been clarified by means of the numerical calculation of emission profile and time-resolved emission spectra.

The symmetry of the charge distribution of a constituent molecule in the excited state is, in general, different from that in the ground state and a static dipole moment is induced at the molecule. These static dipoles strongly affect the nature of Frenkel excitons at high excitation. For an example, excitonic strings which are the bound states of more than two excitons are generated due to the attractive interactions between them.[10] The superradiance of Frenkel excitons will also be influenced by them.

The recent development of laser technology has made it possible to generate short intense laser pulses of the order of $10 \sim 10^2$ femtoseconds. If we use such short pulses, we may generate rather dense and coherent Frenkel excitons instantaneously. The emission from them is collective spontaneous emission if we can neglect reabsorption of emitted light and we may observe the specific nature of superradiance of Frenkel excitons described above. In the actual experiments, especially for the aggregates of dyes and conjugated polymers, which are potentially the candidates of highly excited exciton systems, it would be almost impossible to excite the systems in the complete population inversion. The initial states of them would be in the partially excited states. Hence it seems important to clarify the features of collective spontaneous emission from the system of incomplete population inversion.

In the present paper, we wish to clarify how the static dipole moments modify the superradiance of Frenkel excitons and to investigate the effects of partial excitation by short intense laser pulses by giving the rigorous expression of the initial state. Some results about the former subject have already been published.[9] We shall give more detailed results and supplementary remarks. In Sec. II, we derive the master equation for a model system of Frenkel excitons under an external electromagnetic field by eliminating the radiation field operators from the system of atoms and radiation field. The elimination of low frequency modes and that of near-resonant modes yield, respectively, the static dipole-dipole interaction between atoms and the propagation of excitation through transition dipole moment, while that of the just resonant modes gives the coherent decay. The effective Hamiltonian for the atomic system is shown to be equivalent to that of quantum XXZ spin model.[10–12] We calculate the initial state distribution prepared by the short intense laser pulse in Sec. III. A rigorous expression of the density matrix for an arbitrary pulse is obtained in the short-pulse limit. In Sec. IV, we give the expressions of emission profile and that of time-resolved emission spectra. We discuss some important features of them and mention the methods for actual numerical calculation. The results of numerical calculations for one-dimensional mesoscopic systems are also shown. The drastic change of the emission profile and spectra due to the static dipole moments is presented. Pulse profile of the superradiance from partial degree of excitation is derived in Sec. V, where rapid decay and oscillation of the emitted pulse from the partial excitation is also found. Section VI is devoted to concluding remarks.

II. SUPERRADIANCE MASTER EQUATION

We start from a collection of N identical nonoverlapping two-level atoms, at positions $\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_N$, coupled to a radiation field. The treatment presented here is similar to the previous works by many authors,[13] except for the existence of static dipole-dipole couplings and an external electromagnetic field.

The total Hamiltonian is written as

$$H_{\text{tot}} = H_0 + H_{\text{rad}} + V, \tag{5}$$

$$H_0 \equiv \hbar\omega_0 \sum_{j=1}^{N} \hat{s}_j^z, \tag{6}$$

$$H_{\text{rad}} \equiv \sum_{\mathbf{k},\epsilon} \hbar\omega_{\mathbf{k}} a^\dagger_{\mathbf{k},\epsilon} a_{\mathbf{k},\epsilon}, \tag{7}$$

$$V \equiv -\sum_{j=1}^{N} [\hat{\mathbf{E}}^+(\mathbf{r}_j)\cdot\hat{\mathbf{P}}(\mathbf{r}_j) + \hat{\mathbf{E}}^-(\mathbf{r}_j)\cdot\hat{\mathbf{P}}^\dagger(\mathbf{r}_j)], \tag{8}$$

where

$$\hat{\mathbf{E}}^+(\mathbf{r}_j) \equiv (-i)\sum_{\mathbf{k},\epsilon}\sqrt{\frac{\hbar c|\mathbf{k}|}{2\epsilon_0\Omega}}\,\boldsymbol{\epsilon} a_{\mathbf{k},\epsilon}\exp[i\mathbf{k}\cdot\mathbf{r}_j],$$

$$\hat{\mathbf{E}}^-(\mathbf{r}_j) \equiv (\hat{\mathbf{E}}^+(\mathbf{r}_j))^\dagger,$$

and

$$\hat{\mathbf{P}}(\mathbf{r}_j) \equiv \boldsymbol{\mu}(\hat{s}_j^+ + \hat{s}_j^-) + \boldsymbol{\mu}'(\hat{s}_j^z + 1).$$

Here, $a^\dagger_{\mathbf{k},\epsilon}$ ($a_{\mathbf{k},\epsilon}$) is the creation (annihilation) operator for the plane wave mode of the radiation field with wave vector $\mathbf{k}$ and unit polarization vector $\boldsymbol{\epsilon}$, and $\boldsymbol{\mu} \equiv \mu\boldsymbol{\varepsilon} \equiv \langle e_j|e\mathbf{r}|g_j\rangle$ and $\boldsymbol{\mu}' \equiv \mu'\boldsymbol{\varepsilon}' \equiv \langle e_j|e\mathbf{r}|e_j\rangle$ are, respectively, the transition dipole matrix element and the static dipole matrix element. Although these values depend on the site index j in general, we assume, in this paper, that they do not. We choose $\mu = |\boldsymbol{\mu}|$ and $\mu' = |\boldsymbol{\mu}'|$ to be real, which is always possible by an appropriate gauge transformation. Thus we have $\hat{\mathbf{P}}^\dagger = \hat{\mathbf{P}}$. In the expression of $\hat{\mathbf{E}}^+(\mathbf{r}_j)$, Ω is the quantized volume of the radiation field, which is much larger than that of the atomic system. The pseudospin operators $\hat{s}_j^\pm$ and $\hat{s}_j^z$ are already defined in the Sec. I.

Let ρ_{tot} be the density matrix of the total system, which obeys the evolution equation:

$$\frac{d}{dt}\rho_{\mathrm{tot}}(t) = \frac{1}{i\hbar}[H_{\mathrm{tot}}, \rho_{\mathrm{tot}}(t)]. \tag{9}$$

Transforming it into the interaction representation:

$$\tilde{\rho_{\mathrm{tot}}}(t) \equiv \exp[i\{H_0 + H_{\mathrm{rad}}\}t/\hbar]\rho_{\mathrm{tot}}(t) \times \exp[-i\{H_0 + H_{\mathrm{rad}}\}t/\hbar],$$

$$\tilde{V}(t) \equiv \exp[i\{H_0 + H_{\mathrm{rad}}\}t/\hbar]V(t) \times \exp[-i\{H_0 + H_{\mathrm{rad}}\}t/\hbar],$$

we get

$$\frac{d}{dt}\tilde{\rho}_{\mathrm{tot}}(t) = \frac{1}{i\hbar}[\tilde{V}(t), \tilde{\rho}_{\mathrm{tot}}(t)]. \tag{10}$$

Iterative integration of Eq. (10) leads to

$$\tilde{\rho}_{\mathrm{tot}}(t + \Delta t) = \tilde{\rho}_{\mathrm{tot}}(t) + \frac{1}{i\hbar}\int_t^{t+\Delta t} d\tau[\tilde{V}(\tau), \tilde{\rho}_{\mathrm{tot}}(t)] + \frac{1}{(i\hbar)^2}\int_t^{t+\Delta t} d\tau_1 \int_t^{\tau_1} d\tau_2[\tilde{V}(\tau_1), [\tilde{V}(\tau_2), \tilde{\rho}_{\mathrm{tot}}(t)]] + \cdots. \tag{11}$$

The reduced density operator for the atomic system is defined (in the interaction representation) as

$$\tilde{\rho}(t) \equiv \mathrm{Tr}_{\mathrm{rad}}[\tilde{\rho}_{\mathrm{tot}}(t)]. \tag{12}$$

First, we consider the case of no external electromagnetic field. Under the superradiant configuration, that is, when the system size is small enough to neglect reabsorption of emitted photons and reflection at the edge of the system, we can approximate

$$\tilde{\rho}(t) \simeq \mathrm{Tr}_{\mathrm{rad}}[\tilde{\rho}_{\mathrm{tot}}(t)|\mathrm{vac}\rangle\langle\mathrm{vac}|]. \tag{13}$$

Here, $|\mathrm{vac}\rangle$ denotes the vacuum of the photon field. By the second Born approximation, we get

$$\frac{d}{dt}\tilde{\rho}(t) \simeq \frac{\tilde{\rho}(t + \Delta t) - \tilde{\rho}(t)}{\Delta t} \simeq \frac{1}{(i\hbar)^2 \Delta t}\int_t^{t+\Delta t} d\tau_1 \int_t^{\tau_1} d\tau_2 \langle\mathrm{vac}|[\tilde{V}(\tau_1), [\tilde{V}(\tau_2), \tilde{\rho}_{\mathrm{tot}}(t)]]|\mathrm{vac}\rangle_r, \tag{14}$$

where Δt is the coarse graining time, which is much longer than the time interval for light to pass through the sample and much shorter than the typical atomic relaxation times. Then we can regard the evolution on the atomic system as a Markov process by treating the effect of spontaneous emission as fluctuation of the atomic system (Markov approximation). Practically, this approximation is to replace the integral $\frac{1}{\Delta t}\int_t^{t+\Delta t} d\tau_1 \int_t^{\tau_1} d\tau_2$ in Eq. (14) by $\int_0^\infty d(\tau_1 - \tau_2)$. After some algebra,[4] we obtain the evolution equation:

$$\frac{d}{dt}\tilde{\rho}(t) = \frac{1}{i\hbar}[\tilde{H}_{XXZ}, \tilde{\rho}(t)] + \tilde{\Gamma}\tilde{\rho}, \tag{15}$$

where the first term includes the static dipole-dipole interactions and the transfer of Frenkel excitons, coming from the elimination of the low frequency modes of the radiation field and from that of the near-resonant modes, respectively. The second term describes the collective decay of the system and is obtained from the elimination of the just resonant modes. They are explicitly given as

$$\tilde{H}_{XXZ} \equiv \sum_{j,j'}\{\Omega_{j,j'}(\hat{s}_j^+\hat{s}_{j'}^- + \hat{s}_j^-\hat{s}_{j'}^+) + \Omega_{j,j'}^z(\hat{s}_j^z\hat{s}_{j'}^z)\},$$

$$\tilde{\Gamma}\tilde{\rho} \equiv -\frac{1}{2}\sum_{j,j'}\gamma_{j,j'}[\hat{s}_j^+\hat{s}_{j'}^-\tilde{\rho} - 2\hat{s}_j^-\tilde{\rho}\hat{s}_{j'}^+ + \tilde{\rho}\hat{s}_j^+\hat{s}_{j'}^-],$$

$$\Omega_{j,j'} \equiv -\frac{\mu^2}{16\pi^3\epsilon_0}\left[\int_0^\infty dk k^3 F(kr_{j,j'})\wp\frac{1}{k - k_0}\right],$$

$$\Omega_{j,j'}^z \equiv -\frac{\mu'^2}{16\pi^3\epsilon_0}\left[\int_0^\infty dk k^3 F'(kr_{j,j'})\wp\frac{1}{k}\right],$$

$$\gamma_{j,j'} \equiv \frac{\mu^2 k_0^3}{8\pi^2\epsilon_0\hbar}F(k_0 r_{j,j'}),$$

$$F(kr_{j,j'}) \equiv \int dS_k(\boldsymbol{\epsilon}\cdot\boldsymbol{\varepsilon})\exp[i\mathbf{k}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})],$$

and

$$F'(kr_{j,j'}) \equiv \int dS_k(\boldsymbol{\epsilon}\cdot\boldsymbol{\varepsilon}')\exp[i\mathbf{k}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})],$$

where $r_{j,j'} \equiv |\mathbf{r}_j - \mathbf{r}_{j'}|$, $k_0 \equiv \omega_0/c$ and we neglect small energy shift of the atomic excitation energy and several off-resonant terms. In deriving the above expressions, we replaced the summation over $\mathbf{k}$ by the integral over $\mathbf{k}$ in Eq. (14), i.e., $\sum_{\mathbf{k}} \to V/(2\pi)^3 \int_0^\infty k^2 dk \int dS_k$ and utilized the identity:

$$\int_0^\infty c\,d\tau \exp[\pm i(k - k_0)c\tau] = \pi\delta(k - k_0) \pm i\wp\frac{1}{k - k_0},$$

where $\wp$ denotes a principal value. If we take the off-resonant terms into account, $\tilde{H}_{XXZ}$ contains the term like $\hat{s}_j^+\hat{s}_{j'}^+ + \hat{s}_j^-\hat{s}_{j'}^-$.

Since we are interested in the linear mesoscopic system, whose length is small compared to an optical wavelength, we can take the limit $k_0 r_{j,j'} \longrightarrow 0$ in the above equations. We also keep only the nearest neighbor interactions, for the dipole-dipole interaction is short-ranged.

Then, transforming to the Schrödinger representation, Eq. (15) reduces to

$$\frac{d}{dt}\rho(t) = \frac{1}{i\hbar}[H_0 + H_{XXZ}, \rho(t)] + \hat{\Gamma}\rho(t), \tag{16}$$

$$H_{XXZ} \equiv -\hbar \sum_{j=1}^{N} \{ J_x (\hat{s}_j^+ \hat{s}_{j+1}^- + \hat{s}_j^- \hat{s}_{j+1}^+) + 2J_z \hat{s}_j^z \hat{s}_{j+1} \}, \tag{17}$$

$$\hat{\Gamma}\rho \equiv -\frac{1}{2}\gamma(\hat{S}^+\hat{S}^-\rho + \rho\hat{S}^+\hat{S}^-) + \gamma\hat{S}^-\rho\hat{S}^+, \tag{18}$$

where

$$2\hbar J_x = -\frac{\mu^2}{4\pi\epsilon_0 a^3}\left[1 - \frac{3(\boldsymbol{\varepsilon}\cdot\mathbf{a})^2}{a^2}\right],$$

$$2\hbar J_z = -\frac{\mu'^2}{4\pi\epsilon_0 a^3}\left[1 - \frac{3(\boldsymbol{\varepsilon}'\cdot\mathbf{a})^2}{a^2}\right],$$

and

$$\gamma \equiv \frac{k_0^3\mu^2}{3\pi\epsilon_0\hbar}, \tag{19}$$

where $\mathbf{a}$ is the primitive vector of the one-dimensional lattice and $a \equiv |\mathbf{a}|$. H_{XXZ} is exactly equal to the Hamiltonian of spin 1/2 quantum XXZ spin chain, which is one of the quantum many body systems, whose properties have been understood in full detail.[11] (If we include the off-resonant terms, the Hamiltonian becomes XYZ Hamiltonian in the interaction representation.) Since H_0 describes the interaction energy between the external magnetic field parallel to the z axis and the quantum spins for the spin system, we can conclude that "the Frenkel exciton system is equivalent to the quantum XXZ spin system under the magnetic field."

One may think that, because of this equivalence, we can immediately observe in Frenkel exciton system the same thermodynamic features as those of quantum XXZ spin, but this is not the case. The excitation energy $\hbar\omega_0$ is usually much larger than the transfer energy $|\hbar J|$ and interaction energy $|\hbar J_z|$. This is also the condition that the ground state of the Frenkel exciton system is stable. As a spin system, this condition means that it is in a strong external magnetic field. The thermodynamic properties of such a spin system are simple and we cannot expect a sign of any noble feature of a quantum spin chain. Under the strong excitation, however, some thermodynamic properties of quantum spin systems appear in the optical spectra.[12] Superradiance starts from the initial state with considerably dense excitons and decays rapidly to the ground state. It is neither stationary nor in thermal equilibrium. Thus we may characterize the superradiance of Frenkel excitons as an appearance of nonstationary and nonequilibrium properties of quantum spin, which are known very little for the actual spin systems.

Now we derive the master equation in an applied electromagnetic field. The effect of an applied field $\mathbf{E}_{\rm cl}(\mathbf{r}, t)$ can be represented by the coherent state of the radiation field prepared by the external field, which remains unchanged after the coarse graining time Δt just as the vacuum state in Eq. (13). The reduced density operator $\tilde{\rho}(t)$ is to be approximated as

$$\tilde{\rho}(t) \simeq \mathrm{Tr}_{\rm rad}[\tilde{\rho}_{\rm tot}(t)|\mathbf{E}(t)\rangle\langle\mathbf{E}(t)|], \tag{20}$$

where $|\mathbf{E}(t)\rangle$ is the coherent state with respect to the applied radiation modes but the vacuum state for the other modes:

$$\hat{\mathbf{E}}^+(\mathbf{r})|\mathbf{E}(t)\rangle = \mathbf{E}_{\rm cl}(\mathbf{r}, t)|\mathbf{E}(t)\rangle. \tag{21}$$

Note that Eq. (13) is a special case of Eq. (21), where $\mathbf{E}_{\rm cl}(\mathbf{r}, t) = \mathbf{0}$, i.e., $|\mathbf{E}(t)\rangle = |{\rm vac}\rangle$. Then the master equation becomes at the same level of approximation as in the case of no applied field:

$$\frac{d}{dt}\rho(t) = \frac{1}{i\hbar}[H_0 + H_{XXZ} + V_{\rm cl}(t), \rho(t)] + \hat{\Gamma}\rho(t), \tag{22}$$

where

$$V_{\rm cl}(t) \equiv -\sum_{j=1}^{N}[\mathbf{E}_{\rm cl}(\mathbf{r}_j, t) + \mathbf{E}_{\rm cl}^*(\mathbf{r}_j, t)] \cdot \hat{\mathbf{P}}(\mathbf{r}_j). \tag{23}$$

We shall use Eq. (22) to obtain the initial state in the next section. Then, according to the superradiance master equation, Eq. (16), we shall calculate the time evolution of the system.

III. INITIAL STATE BY SHORT-PULSE EXCITATION

The initial state of the superradiance can be prepared with a short intense laser pulse. Since the atomic system is in the ground state before the application of the pulse, we have to solve Eq. (22) for the external field $\mathbf{E}(\mathbf{r}, t)$, starting from the ground state distribution $\rho_0 \equiv |g\rangle\langle g|$. The ground state $|g\rangle$ is equal to $|S = \frac{N}{2}; M = -\frac{N}{2}\rangle$ in the spin state description. It is fairly difficult even for numerical calculation to solve Eq. (22) for a general case. When, however, the duration of the pulse t_f is short enough to satisfy: $t_f \ll \gamma^{-1}$, J_x^{-1}, J_z^{-1}, the evolution of the system is well approximated by

$$\frac{d}{dt}\rho(t) = \frac{1}{i\hbar}[H_0 + V_{\rm cl}(t), \rho(t)]. \tag{24}$$

Neglecting the spatial change of the electromagnetic field over the mesoscopic system and denoting $E(t) \equiv \frac{1}{\hbar}\boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{r}_j, t)$, we get from Eq. (23) and the definition of $\hat{\mathbf{P}}(\mathbf{r}_j)$,

$$H_0 + V_{\rm cl}(t) \simeq \hbar\omega_0\hat{S}^z - \hbar[E(t)\hat{S}^+ + E^*(t)\hat{S}^-]. \tag{25}$$

Then the reduced density operator at time t is given as a pure state:

$$\rho(t) = |\Psi(t)\rangle\langle\Psi(t)|, \tag{26}$$

$$|\Psi(t)\rangle \equiv \hat{F}(t)|g\rangle, \tag{27}$$

$$\hat{F}(t) \equiv T\exp\left[\int_0^t d\tau\{-i\omega_0\hat{S}^z + i[E(\tau)\hat{S}^+ + E^*(\tau)\hat{S}^-]\}\right], \tag{28}$$

where T denotes the time ordered products. We wish to reduce $\hat{F}(t)$ to a simpler form. Using the commutation relations of the spin operators, we can easily prove the following useful equalities for an arbitrary complex variable λ:

$$\exp[-\lambda\hat{S}^z]\;\hat{S}^{\pm}\;\exp[\lambda\hat{S}^z] = e^{\mp\lambda}\hat{S}^{\pm},$$

$$\exp[-\lambda\hat{S}^+]\;\hat{S}^-\;\exp[\lambda\hat{S}^+] = -\lambda^2\hat{S}^+ - 2\lambda\hat{S}^z + \hat{S}^-,$$

$$\exp[-\lambda\hat{S}^-]\;\hat{S}^+\;\exp[\lambda\hat{S}^-] = -\lambda^2\hat{S}^- + 2\lambda\hat{S}^z + \hat{S}^+,$$

$$\exp[-\lambda\hat{S}^{\pm}]\;\hat{S}^z\;\exp[\lambda\hat{S}^{\pm}] = \pm\lambda\hat{S}^{\pm} + \hat{S}^z. \tag{29}$$

With an initial condition $\hat{F}(0) = 1$, $\hat{F}(t)$ satisfies the differential equation:

$$\frac{d}{dt}\hat{F}(t) = \{-i\omega_0\hat{S}^z + i[E(t)\hat{S}^+ + E^*(t)\hat{S}^-]\}\hat{F}(t). \tag{30}$$

We find from the equalities (29) that the operator $\hat{G}(t) \equiv \exp[i\omega_0 t\hat{S}^z]\;\hat{F}(t)$ satisfies

$$\frac{d}{dt}\hat{G}(t) = i[E'(t)\hat{S}^+ + E'^*(t)\hat{S}^-]\hat{G}(t), \tag{31}$$

where $E'(t) \equiv E(t)e^{i\omega_0 t}$. Furthermore, the operator $\hat{H}(t)$, $\hat{G}(t) \equiv \exp[iX(t)\hat{S}^+]\hat{H}(t)$, satisfies

$$\frac{d}{dt}\hat{H}(t) = [2E'^*(t)X(t)\hat{S}^z + iE'^*(t)\hat{S}^-]\hat{H}(t), \tag{32}$$

if $X(t)$ is the solution of the following equation with the initial condition $X(0) = 0$:

$$E'(t) - \frac{d}{dt}X(t) + E'^*(t)X(t)^2 = 0. \tag{33}$$

Finally we find the operator $\hat{K}(t)$, $\hat{H}(t) \equiv e^{A(t)\hat{S}^z}\hat{K}(t)$, obeys

$$\frac{d}{dt}\hat{K}(t) = iE'^*(t)e^{A(t)}\hat{S}^-\hat{K}(t), \tag{34}$$

where

$$A(t) \equiv \int_0^t d\tau 2E'^*(\tau)X(\tau). \tag{35}$$

By integrating Eq. (34) and from the definition of $\hat{K}(t)$, $\hat{H}(t)$, and $\hat{G}(t)$, we get

$$\begin{aligned}\hat{F}(t) = {}&\exp[-i\omega_0 t\hat{S}^z]\exp[iX(t)\hat{S}^+]\\ &\times\exp[A(t)\hat{S}^z]\exp[iB(t)\hat{S}^-],\end{aligned} \tag{36}$$

where $B(t) \equiv \int_0^t d\tau E'^*(\tau)e^{A(\tau)}$. Thus, we find $|\Psi(t)\rangle$ is given by

$$|\Psi(t)\rangle = \exp[-i\omega_0 t\hat{S}^z]\exp[iX(t)\hat{S}^+]\exp\left[-\frac{N}{2}A(t)\right]|g\rangle. \tag{37}$$

Since $A(t)$ is determined by $X(t)$, we can obtain the concrete expression of the initial state by solving the Riccati-type equation, Eq. (33).

When we can approximate $E'(t)$ as a real function $E_0(t)$ up to an unimportant constant phase factor, we can solve Eq. (33) explicitly as

$$X(t) = \tan\left[\int_0^t d\tau E_0(\tau)\right]. \tag{38}$$

This essentially corresponds to the rotating wave approximation. By denoting $\Theta(t) \equiv \int_0^t d\tau E_0(\tau)$, we get from Eqs. (35) and (38),

$$A(t) = -2\ln|\cos\Theta(t)|. \tag{39}$$

Substituting Eqs. (38) and (39) to Eq. (37), we find

$$\begin{aligned}|\Psi(t)\rangle = {}&|\cos\Theta(t)|^N \sum_{M=-N/2}^{N/2} e^{-iM\omega_0 t}\frac{(i\tan\Theta(t))^{M+N/2}}{(M+N/2)!}\\ &\times\left\{\prod_{k=0}^{M-1+N/2}(N-k)^{1/2}(k+1)^{1/2}\right\}\left|\frac{N}{2};M\right\rangle.\end{aligned} \tag{40}$$

For an arbitrary shape of a pulse, we have to solve the nonlinear equation (33). This is not difficult because we can transform it into a second order linear differential equation by changing a variable from $X(t)$ to $u(t)$:

$$X(t) \equiv -E^{*-1}(t)\frac{d}{dt}\ln u(t), \tag{41}$$

with the initial condition $u(0) = 1$ and $\frac{d}{dt}u(0) = 0$. Then, by defining $\beta(t) \equiv \frac{d}{dt}\ln E'^*(t)$,

$$\frac{d^2}{dt^2}u(t) + \beta(t)\frac{d}{dt}u(t) + |E(t)|^2u(t) = 0. \tag{42}$$

IV. SUPERRADIANT PULSE PROFILE AND TIME-RESOLVED EMISSION SPECTRUM

The characteristics of superradiant pulse from the highly excited system of Frenkel excitons can be understood from its time dependence of the emission intensity [emission profile $I(t)$] and that of the frequency decomposition of the emitted light [time-resolved emission spectrum $I(\omega,t;\Delta t)$]. They are defined, respectively, as[8]

$$\begin{aligned}I(t) &\equiv -\frac{d}{dt}\mathrm{Tr}[H_0\rho(t)]\\ &= \gamma\omega_0\mathrm{Tr}[\hat{S}^+\hat{S}^-\rho(t)],\end{aligned} \tag{43}$$

$$\begin{aligned}I(\omega,t;\Delta t) = {}&\frac{\gamma\omega_0}{\Delta t}\int_{-\Delta t/2}^{\Delta t/2} d\tau_1 \int_{-\Delta t/2}^{\Delta t/2} d\tau_2 \langle\hat{S}^+(t+\tau_1)\\ &\times\hat{S}^-(t+\tau_2)\rangle e^{-i\omega(\tau_1-\tau_2)}.\end{aligned} \tag{44}$$

Here and hereafter we take $\hbar = 1$. The derivation of $I(\omega,t;\Delta t)$ is described in the Appendix.

For the numerical calculation, we utilize the fact that $\hat{S}^z$ commutes with H_0 and H_{XXZ} so that we can choose the simultaneous eigenstates of $\hat{S}_z$ and H_{XXZ} as the basis functions. We denote them by $\{|v_m\rangle\}$, where subscript m

is the eigenvalue of $\hat{S}^z$. We employ a periodic boundary condition for simplicity. Although we can obtain the exact eigenstates of H_{XXZ} by Bethe ansatz or quantum inverse scattering method, for the practical reason, we take $|v_m\rangle$ to be the simultaneous eigenstates of the translation operator, the reflection operator and the Ising model (the Hamiltonian for $J_x = 0$). For an example, if we denote

$$|1,2,4\rangle \equiv |e_1\rangle \otimes |e_2\rangle \otimes |g_3\rangle \otimes |e_4\rangle \otimes |g_5\rangle \otimes |g_6\rangle, \tag{45}$$

an unnormalized basis function for six particles is $|1,2,4\rangle\rangle$:

$$\begin{aligned} |1,2,4\rangle\rangle &\equiv |1,2,4\} + |6,5,3\}, \\ |1,2,4\} &\equiv |1,2,4\rangle + |2,3,5\rangle + |3,4,6\rangle \\ &\quad + |4,5,1\rangle + |5,6,2\rangle + |6,1,3\rangle. \end{aligned} \tag{46}$$

The time evolution of the matrix elements of the density operator is written down with these bases as

$$\begin{aligned} \frac{d}{dt}\langle v_m|\rho(t)|v'_{m'}\rangle &= \frac{\gamma}{2}\sum_{v''_m}\langle v_m|\hat{S}^+\hat{S}^-|v''_m\rangle\langle v''_m|\rho|v'_{m'}\rangle - \frac{\gamma}{2}\sum_{v''_{m'}}\langle v_m|\rho|v''_{m'}\rangle\langle v''_{m'}|\hat{S}^+\hat{S}^-|v'_{m'}\rangle \\ &\quad + \gamma\sum_{v''_{m+1}}\sum_{v'''_{m'+1}}\langle v_m|\hat{S}^-|v''_{m+1}\rangle\langle v''_{m+1}|\rho|v'''_{m'+1}\rangle\langle v'''_{m'+1}|\hat{S}^+|v'_{m'}\rangle \\ &\quad + \frac{1}{i}\left(\sum_{v''_m}\langle v_m|H_{XXZ}|v''_m\rangle\langle v''_m|\rho|v'_{m'}\rangle - \sum_{v''_{m'}}\langle v_m|\rho|v''_{m'}\rangle\langle v''_{m'}|H_{XXZ}|v'_{m'}\rangle\right). \end{aligned} \tag{47}$$

We find from this expression that the equation of the matrix element $\langle v_m|\rho|v'_{m'}\rangle$ is linear and couples with a matrix element $\langle v''_{m''}|\rho|v'''_{m'''}\rangle$ only if $m - m' = m'' - m'''$. Therefore, we can write

$$\begin{aligned} \langle v_m|\rho(t+\tau)|v'_{m+l}\rangle = \sum_{v''_{m'}}\sum_{v'''_{m'+l}} O^{v''_{m'}v'''_{m'+l}}_{v_m v'_{m+l}}(\tau) \\ \times\langle v''_m|\rho(t)|v'''_{m'+l}\rangle, \end{aligned} \tag{48}$$

where $O\cdots(\tau)$ is a function of τ determined by Eq. (47). Similarly, we can express the emission profile as

$$\begin{aligned} I(t) = \gamma\omega_0\sum_{v_m}\sum_{v'_{m-1}}\sum_{v''_m}\langle v_m|\hat{S}^+|v'_{m-1}\rangle\langle v'_{m-1}|\hat{S}^-|v''_m\rangle \\ \times\langle v''_m|\rho(t)|v_m\rangle. \end{aligned} \tag{49}$$

To calculate the time-resolved emission spectrum, we have to evaluate the time correlation function $\langle\hat{S}^-(t+\tau)\hat{S}^+(t)\rangle$. For that purpose, we use the quantum regression theorem which can be stated as followings.[14] If an expectation value of an operator $\hat{A}$ is expressed as

$$\langle\hat{A}(t+\tau)\rangle = \sum_\eta Q_\eta(\tau)\langle\hat{f}_\eta(t)\rangle, \tag{50}$$

then the correlation function $\langle\hat{B}(t)\hat{A}(t+\tau)\rangle$ is given by

$$\langle\hat{B}(t)\hat{A}(t+\tau)\rangle = \sum_\eta Q_\eta(\tau)\langle\hat{B}(t)\hat{f}_\eta(t)\rangle. \tag{51}$$

Using Eq. (48), $\langle\hat{S}^-(t+\tau)\rangle$ is written as,

$$\begin{aligned} \langle\hat{S}^-(t+\tau)\rangle &\equiv \mathrm{Tr}[\hat{S}^-\rho(t+\tau)] \\ &= \sum_{v_m}\sum_{v'_{m-1}}\langle v'_{m-1}|\hat{S}^-|v_m\rangle\langle v_m|\rho(t+\tau)|v'_{m-1}\rangle \\ &= \sum_{v_m}\sum_{v'_{m-1}}\sum_{v''_{m'}}\sum_{v'''_{m'-1}}\langle v'_{m-1}|\hat{S}^-|v_m\rangle \\ &\quad \times O^{v''_{m'}v'''_{m'-1}}_{v_m v'_{m-1}}(\tau)\langle v''{}_{m'}|\rho(t)|v'''_{m'-1}\rangle. \end{aligned} \tag{52}$$

Therefore, by the identification,

$$\begin{aligned} &\hat{A} = \hat{S}^-, \quad \hat{B} = \hat{S}^+, \\ &\eta = (v_m, v'_{m-1}, v''_{m'}, v'''_{m'-1}), \\ &\hat{f}_\eta = |v'''_{m'-1}\rangle\langle v'_{m-1}|\hat{S}^-|v_m\rangle\langle v''_{m'}|, \\ &Q_\eta(\tau) = O^{v''_{m'}v'''_{m'-1}}_{v_m v'_{m-1}}(\tau), \end{aligned} \tag{53}$$

we obtain

$$\begin{aligned} \langle\hat{S}^+(t)\hat{S}^-(t+\tau)\rangle &= \sum_{v_m}\sum_{v'_{m-1}}\sum_{v''_{m'}}\sum_{v'''_{m'-1}} O^{v''_{m'}v'''_{m'-1}}_{v_m v'_{m-1}}(\tau)\langle\hat{S}^+|v'''_{m'-1}\rangle\langle v'_{m-1}|\hat{S}^-|v_m\rangle\langle v''_{m'}|\rangle_t \\ &= \sum_{\tilde{v}_{m'}}\sum_{v_m}\sum_{v'_{m-1}}\sum_{v''_{m'}}\sum_{v'''_{m'-1}} O^{v''_{m'}v'''_{m'-1}}_{v_m v'_{m-1}}(\tau)\langle\tilde{v}_{m'}|\hat{S}^+|v'''_{m'-1}\rangle \\ &\quad \times\langle v'_{m-1}|\hat{S}^-|v_m\rangle\langle v''_{m'}|\rho(t)|\tilde{v}_{m'}\rangle. \end{aligned} \tag{54}$$

From these expressions (53) and (54), we find that only the block diagonal matrix elements of the density operator with respect to m are necessary for the emission profile and the time-resolved emission spectrum. One important consequence is "both of emission profile and time-resolved emission spectrum are independent of the (block) off-diagonal matrix elements of the initial state." In this respect, we cannot distinguish a pure state from mixed state by the emission profile and the time-resolved emission spectrum.

In this section, we show the numerical results for the superradiance from complete-population inversion system. We take 1/10 times spontaneous emission time, i.e., $1/10 \times \gamma^{-1}$, as a unit of time. Hence $10\hbar\gamma$ corresponds to the unit of the energy. The emission profile and the time-resolved emission spectrum have been studied under the constraint $J_x^2 + J_z^2 = 4$ with this unit. We note that the emission profile is invariant under the transformation: $(J_x, J_z) \longrightarrow (-J_x, -J_z)$. This feature is easily proven from the fact:

$$\begin{aligned} I(t) &= I(t)^* \\ &= \gamma\omega_0 \mathrm{Tr}[\hat{S}^+\hat{S}^-\rho(t)]^* \\ &= \gamma\omega_0 \mathrm{Tr}[\hat{S}^+\hat{S}^-\rho(t)^\dagger] \\ &= \gamma\omega_0 \mathrm{Tr}[\hat{S}^+\hat{S}^-\tilde{\rho}(t)^\dagger], \end{aligned} \tag{55}$$

and $\tilde{\rho}(t)^\dagger$ obeys the same equation as that of $\tilde{\rho}(t)$ with $H_{XXZ} \to -H_{XXZ}$. The similar arguments lead to the fact that the time-resolved emission spectrum is also invariant under $(J_x, J_z, \omega - \omega_0) \longrightarrow (-J_x, -J_z, \omega_0 - \omega)$. Thus we restricted ourselves to the values for $J_x \geq 0$. They are schematically shown in Fig. 1. The points I, H, and X correspond, respectively, to the Ising, Heisenberg and $XX0$ models as quantum spin systems. The emission profile $I(t)$ for several values of J_x and J_z are shown in Figs. 2(a)–(h). The system size $N = 8$. They show the appearance of slow emission tails except for the Heisenberg case ($J_x = J_z$). In fact, as we prove in Ref. 9, the superradiance from the Heisenberg case is exactly equal to the Dicke's superradiance ($J_x = J_z = 0$). In the Ising case ($J_x = 0$), we observe the oscillating nature. The period of the oscillation, Δt, depends on a value of $|J_z|$ as is shown in Fig. 3. This oscillation in $\Delta t = 2\pi/2J_z$ comes from the interference of the two transitions with frequency difference $2J_z$, because the transition dipole moments in the Ising limit have only three frequencies, ω_0 and $\omega_0 \pm 2J_z$.[9]

The time-resolved emission spectra corresponding to the cases of Figs. 2(a)–(f) have already been reported in Figs. 2(a)–(f) of Ref. 9. Here, we only show a stereographic presentation of the $XX0$ case in Fig. 4. The chirping effects clearly appear in these spectra. In the Ising case, the peak shifts from low frequency to high frequency side. On the other hand, it shifts from high to low frequency side in the $XX0$ case. In general, we observe blueshift to redshift for $J_z < J_x$ and the opposite for $J_x < J_z$. The reason is the following. When we reduce one exciton from the system with high density of excitons, except for the atomic excitation energy $\hbar\omega_0$, the kinetic energy due to the excitation transfer $(-\hbar J_x \hat{s}_j^+ \hat{s}_{j+1}^-$ + H.c.) becomes lower, for the other excitons can transfer more easily. On the other hand, the energy due to the interaction between excitons $(-\hbar J_z \hat{s}_j^z \hat{s}_{j+1}^z)$ becomes higher. Because, in the spin state description, the number of the down spin states increases by one in the system where most states are in up spin state and the number of adjacent pairs with different spins increases. At low density of excitons, the situation is just the opposite. Hence we observe above features of emission frequency.

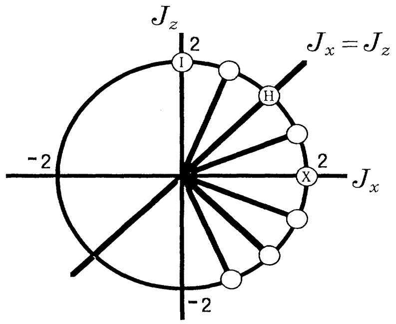

FIG. 1. The values of J_x and J_z we adopted for the numerical calculations are shown in the J_x-J_z plane. I, H, and X correspond, respectively, to Ising, Heisenberg, and $XX0$ models.

V. SUPERRADIANCE FROM PARTIALLY EXCITED SYSTEM

We discuss the emission profiles from the system partially excited by short intense laser pulse. We adopt the initial state given by Eq. (40) which is completely determined by the Bloch angle $\Theta(t_f)$. The results for $XX0$ model ($J_z = 0$) are shown in Fig. 5, where the Bloch angle $\Theta(t_f) =$ (a) π, (b) 0.8π, (c) 0.6π, (d) 0.5π, (e) 0.4π, and (f) 0.2π. The other parameters are the same as those in Fig. 2(e). The peak of the emission intensity appears at the beginning for $\Theta(t_f) \simeq 0.5\pi$, which is higher than that for the complete inversion $\Theta(t_f) = \pi$. This is because the initial state for $\Theta(t_f) = 0.5\pi$ has the largest total transition dipole moment with cooperation number $S = \frac{1}{2}N$, which gives the emission intensity proportional to N^2 just as in the Dicke's case, or the Heisenberg case, whereas the peak emission intensity of $XX0$ case is proportional to $N^{3/2}$ when γ is considerably small.[12] The initial state prepared by the short pulse pumping is described mainly as a superposition of $\{\pm\pi/8, \pm3\pi/8\}$ and $\{\pm\pi/8, \pm5\pi/8\}$ for the case of (d) $\Theta(t_f) = 0.5\pi$. Here, $\{\pm k_m, \pm k_m'\}$ means that four fermion states with wave numbers $\pm k_m$, $\pm k_m'$ are occupied and others unoccupied as in Ref. 8. For the case if the excitation degree (e) $\Theta(t_f) = 0.4\pi$, other configurations $\{0, \pm\pi/4\}$ and $\{0, \pm3\pi/4\}$ are also superposed on the above configurations. The emission intensity from the excitation $m = 4$ to $m = 3$ comes from two configurations of $\{\pm\pi/8, \pm3\pi/8\}$, and $\{\pm\pi/8, \pm5\pi/8\}$, with the energy difference of $2J_x \times 2\{\cos(3\pi/8) - \cos(5\pi/8)\} \simeq 2J_x \times 1.44$. They interfere constructively at $\Delta t = 2\pi/2.88J_x \simeq 1.1$ and destructively at $\Delta t' = \pi/2.88J_x \simeq 0.55$ in our time unit. For the transition from $m = 3$ to $m = 2$, the emission intensity interferes with the energy difference of $2J_x \cdot \cos(\pi/4) \simeq 2J_x \times 1.41$, i.e., in time $\Delta t = 2\pi/(2J_x \times 1.41) \simeq 1.1$. Under the full inversion $\Theta(t_f) = \pi$, a single initial state $\{\pm\pi/8, \pm3\pi/8, \pm5\pi/8, \pm7\pi/8\}$ is prepared and the emission process with the largest cooperation number is dominant. As a result, we have no interference for the case (a) $\Theta = \pi$. For larger N, the distribution of the frequencies of transition dipole moments becomes continuous and the interference smears

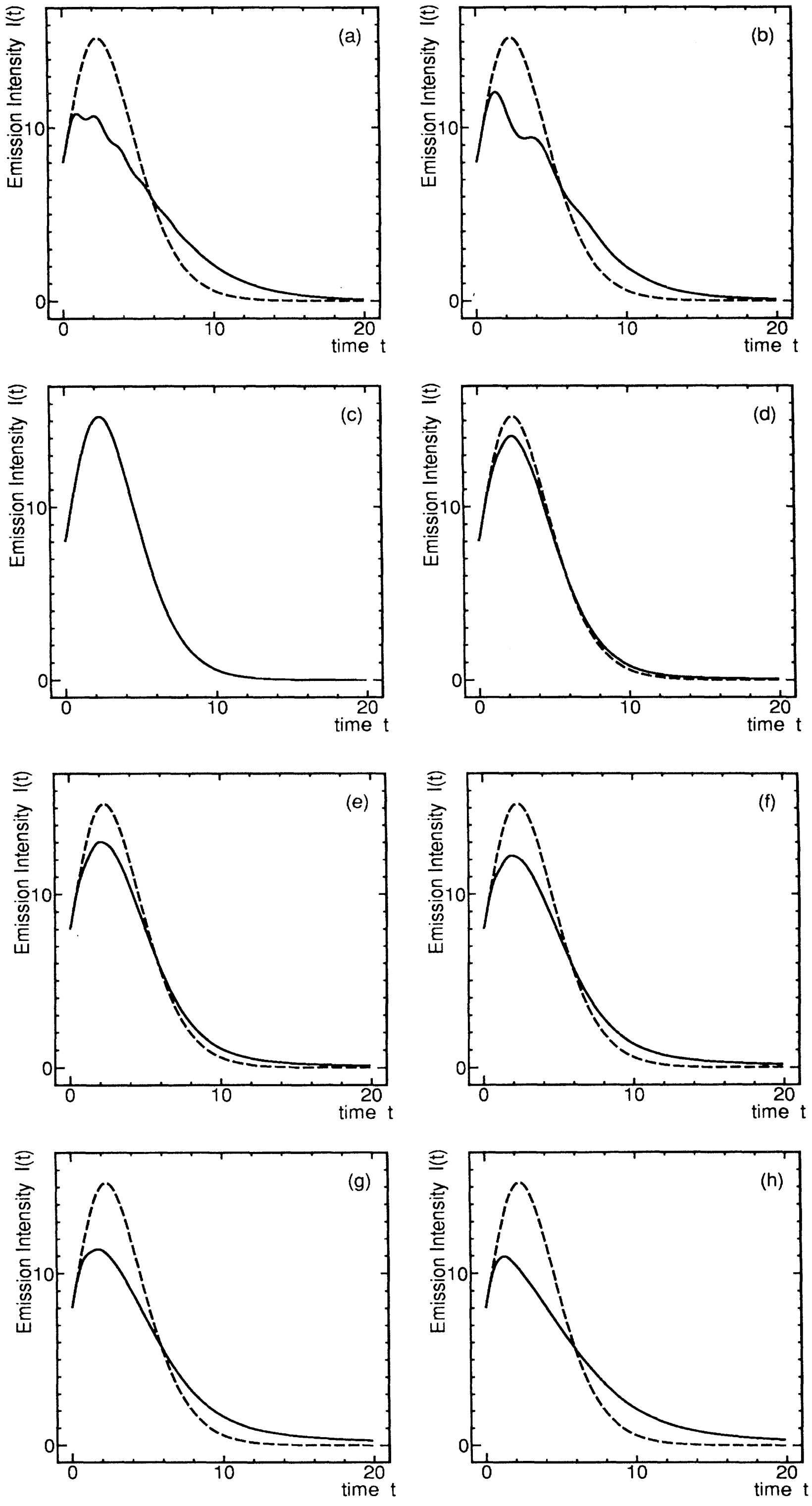

FIG. 2. Emission profile $I(t)$ of the superradiance from the initial state with complete population inversion. $N = 8$ and $\gamma = 0.1$. (a) $J_x = 0$, $J_z = 2.0$; (b) $J_x = 0.765$, $J_z = 1.85$; (c) $J_x = 1.41$, $J_z = 1.41$; (d) $J_x = 1.85$, $J_z = 0.765$; (e) $J_x = 2.0$, $J_z = 0$; (f) $J_x = 1.85$, $J_z = -0.765$; (g) $J_x = 1.41$, $J_z = -1.41$; (h) $J_x = 0.765$, $J_z = -1.85$. The dashed lines are the emission profile of Dicke superradiance.

out except for the Ising case, so that this phenomenon is due to the finite size effect and disappear in the limit $N \to \infty$. Therefore once the oscillation is observed, we can estimate the effective coherence length of the actual system by this oscillation.

The difference of the emission profile for several models are shown in Fig. 6. Here we choose $\Theta(t_f) = 0.5\ \pi$ and denote (a) the Ising case, (b) the Heisenberg case, (c) the $XX0$ case, and (d) $J_x = -J_z$. The oscillation is conspicuous in the Ising case. The emission lines in this case consist of three lines ω_0, and $\omega_0 \pm 2J_z$ and interfere constructively in every time interval $\Delta t = 2\pi/2J_z \simeq$

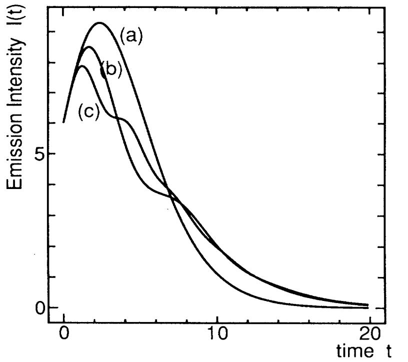

FIG. 3. The emission profile of the Ising case ($J_x = 0$). (a) $J_z = 0$, (b) $J_z = 0.5$, (c) $J_z = 1.0$. The oscillation period is π/J_z.

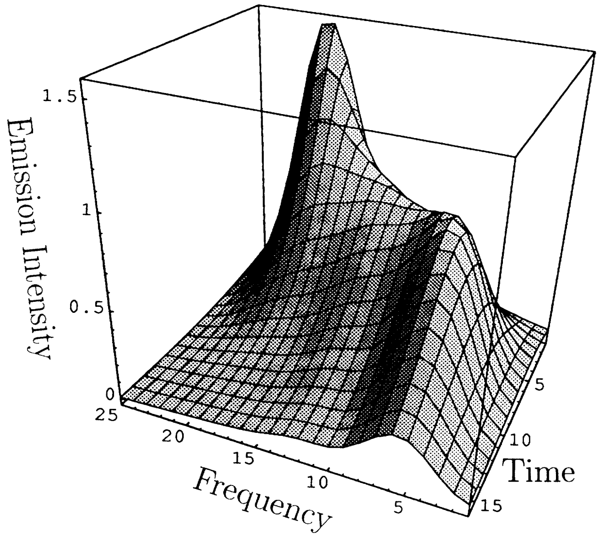

FIG. 4. The stereographic presentation of the time-resolved emission spectra $I(\omega, t; \Delta t)$ from the initial state with complete-population inversion for the $XX0$ case. These spectra correspond to Fig. 2(e).

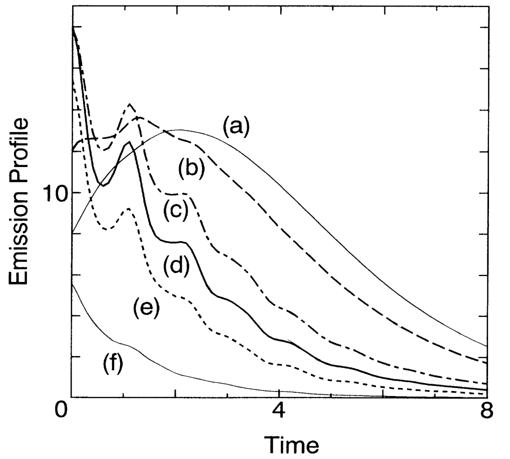

FIG. 5. Emission profile from the partially excited system of the $XX0$ case. $\Theta(t_f) =$ (a) π, (b) 0.8π, (c) 0.6π, (d) 0.5π, (e) 0.4π, and (f) 0.2π. $N = 8$, $\gamma = 0.1$, $J_x = 2.0$, and $J_z = 0$.

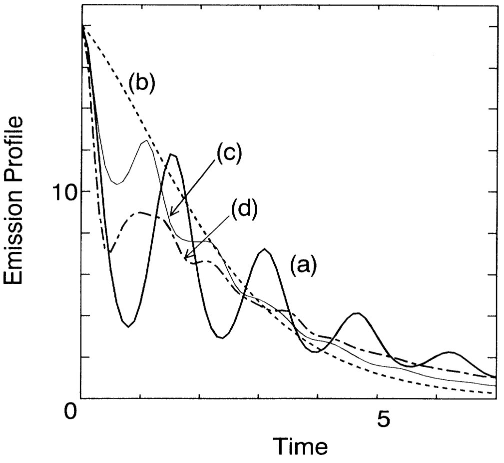

FIG. 6. Emission profile from the partially excited system for $\Theta(t_f) = 0.5$. (a) Ising case, (b) Heisenberg case, (c) $XX0$ case, and (d) $J_x = -J_z$.

1.57 in our time unit. No oscillation is observed in the Heisenberg case, for the dipole moments have a unique frequency ω_0. One important feature is that the initial emission decay of the Ising, $XX0$, and $J_x = -J_z$ cases is more rapid than that of the Heisenberg case, which is equal to the Dicke superradiance. This is due to the destructive interference around $\Delta t' = \pi/2J_z \simeq 0.79$ for the Ising model and at $\Delta t' = \pi/2.88J_x \simeq 0.55$ for the $XX0$ model. As a result, the emission intensity looks to decay very rapidly at the initial stage.

We have calculated the time-resolved emission spectra for superradiant processes from the state $\Theta(t_f) = 0.5\pi$. Figures 7(a), (b), (c), and (d) correspond, respectively, to the cases of Figs. 6(a), (b), (c), and (d). The slit-time width to observe the time-resolved emission spectrum was chosen to be $\Delta t = 2.56$, so that it is larger than the oscillatory periods 1.1 for the $XX0$ model and 1.57 for the Ising model. As a result, the oscillatory intensity observed in Fig. 6 has been partially smeared out in the time-resolved emission spectra of Fig. 7. In these cases, the time-resolved spectra under $\Theta(t_f) = 0.5\pi$ are very similar to those at the latter half in the superradiant pulse under the full population inversion. That is, for the Ising model of Fig. 7(a), the emission intensity on the high energy side decreases gradually in time while that at the central frequency is rather strong at the beginning and decays more rapidly. For the Heisenberg model of Fig. 7(b), the initial decay at $\Theta(t_f) = 0.5\pi$ is strongest reflecting the Dicke's superradiance. The observed spectra for the models of $XX0$ [Fig. 7(c)] and $J_x = -J_z$ [Fig. 7(d)] are very similar to those at the latter half under the complete population inversion.

On the other hand, the time-resolved emission spectra observed more carefully, e.g., at shorter time-intervals even with the same time-slit width $\Delta t = 2.56$, are found to oscillate in time as shown in Fig. 8 for the Ising model. This oscillatory behavior comes from the same origin as in Fig. 6. As a conclusion of this section, we will be able

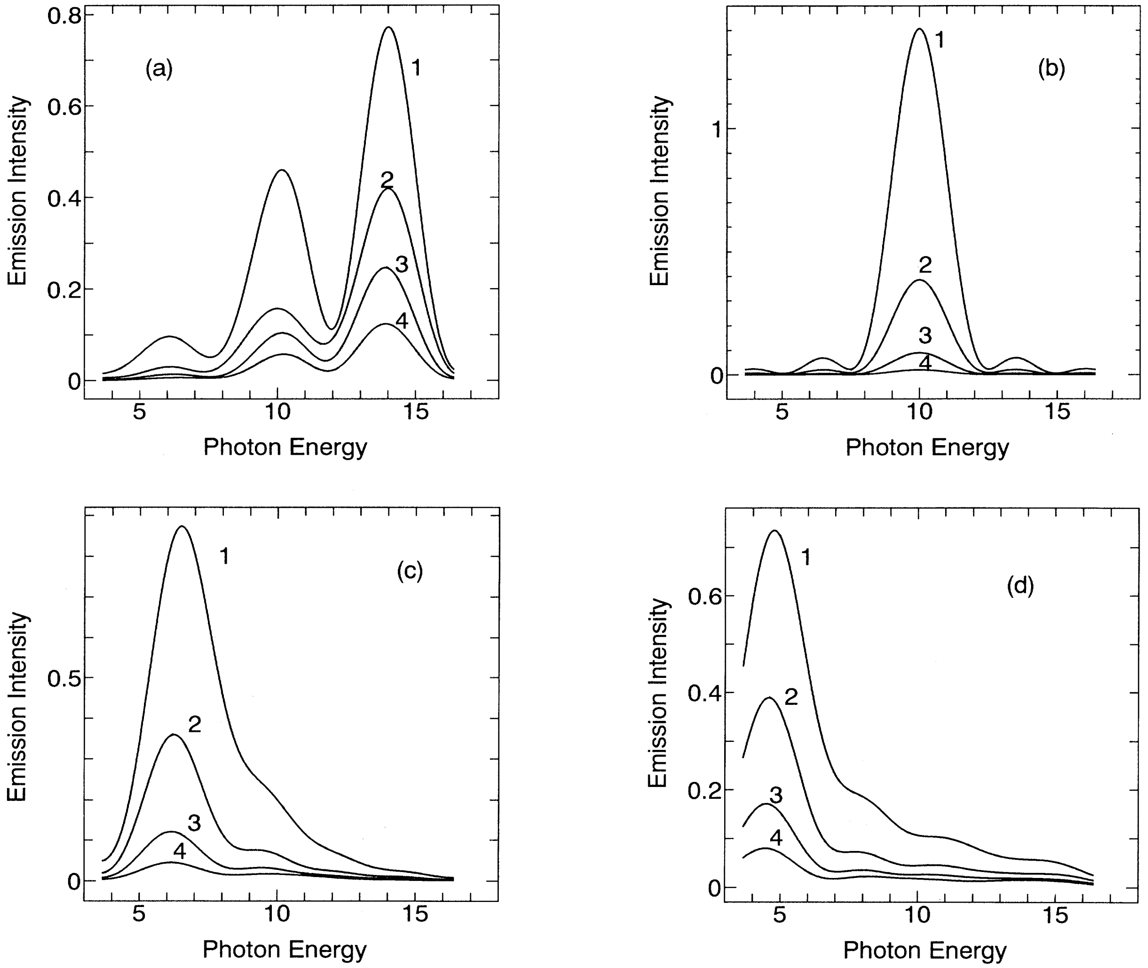

FIG. 7. The time-resolved emission spectra $I(\omega, t; \Delta t)$ from partially excited initial states. They correspond, respectively, to Figs. 6(a)–(d). The number in the figure means the time at which the spectrum is measured with the slit time width $\Delta t = 2.56$, i.e., (1) $t = 1.39$, (2) 2.39, (3) 3.39, and (4) 4.39.

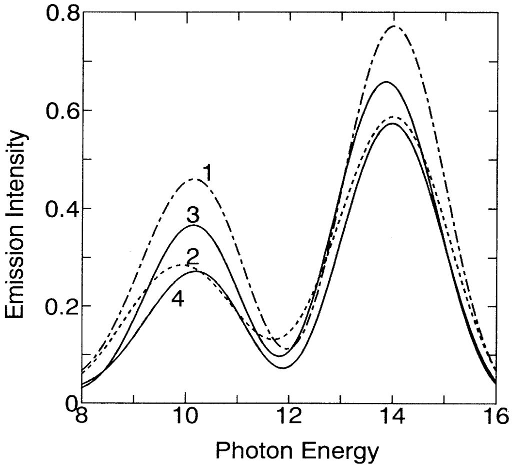

FIG. 8. The time-resolved emission spectra of the Ising case from the partially excited initial state. The number in the figure means the time at which the spectrum is measured with the slit time width $\Delta t = 2.56$, i.e., (1) $t = 1.39$, (2) 1.64, (3) 1.89, and (4) 2.14. The oscillation of intensity is observed.

to observe several phenomena in the superradiant decay also under the partial excitation as an initial electronic state.

VI. CONCLUSION

We have investigated the superradiance of Frenkel excitons in one-dimensional mesoscopic systems in terms of the equivalence to the XXZ spin chains. We have clarified that both emission profile and time-resolved emission spectrum are strongly dependent on the exciton transfer J_x and interaction J_z, due to the static dipole moments. In the Ising limit, the emission profile shows oscillation with period π/J_z. The superradiance of the Heisenberg limit is exactly equal to that of Dicke. We also gave the rigorous expression of the initial state by the short-pulse excitation. Using the expression, we have demonstrated that the superradiance from partially excited system of Frenkel excitons shows oscillation in time and more rapid decay at the initial stage than Dicke's superradiance.

We will discuss the candidate materials to study the superradiance phenomena described in this paper. Excitons in J and H aggregates of dyes have been well described as one-dimensional Frenkel excitons.[15–17] There are many kinds of dye molecules. These molecules constitute J aggregates in some cases and others do H aggregates. We can change the sign of J_x, the exciton-transfer matrix elements in our model. The actual magnitude of ω_0, J_x, and γ in Eq. (19) for BIC or PIC-Br J aggregates are $\hbar\omega_0 \simeq 2.0$ eV, $\hbar J_x \simeq 100$ meV, and $\gamma^{-1} \simeq 500$ ps.[15,16] In addition to this, decorating the dye molecule by many kinds of radicals, we can control the induced static dipole moment within the molecule. Consequently, we can change not only the sign but also the magnitude of J_z the exciton-exciton interaction. In fact, the value of $|J_x|$ is greater than that of $|J_z|$ for most J and H aggregates, but PIC-Br J aggregates is known to have much larger $|J_z|$ than $|J_x|$.[16] Therefore, it looks interesting to study experimentally the superradiant pulse profile and the time-resolved emission spectrum from the partially excited J or H aggregates of dyes. These observations will be used to check the present model and will be useful in determining the electric structure and dynamics of elementary excitations in aggregates of dyes. Another candidate is a weak charge-transfer exciton system such as anthracene-PMDA (pyromellic acid dianhydride),[10] where $\hbar J_x = 5 \sim 6$ meV, $\hbar J_z = 12 \sim 18$ meV, and $\gamma \simeq 0.1 J_x$. The magnitude of J_z can be controlled by an application of static electric field, so that we can expect fruitful emission spectra discussed in the present paper. From these numerical values, the superradiance pulse from these excitonic systems is estimated to have a pulse width of several tens of a picosecond for the system with coherent length of ten molecules or molecule pairs. Therefore, we need the pumping lasers with shorter than several tens picosecond to observe the superradiant emission. However, we must use the much shorter-laser pulses as a pumping source in order to satisfy the condition introduced in Sec. III, i.e., the pump pulse width $t_f \ll \gamma^{-1}$, J_x^{-1}, and J_z^{-1}. Therefore, we need several tens of a femtosecond pulses to pump the excitons in anthracene-PMDA and several femtosecond pulses for the excitons in dye aggregates.

The static dipole moment induced in the excited state comes from the different electronic distribution without any lattice deformation in the J and H aggregates of dyes. In the case of charge-transfer exciton, such as in anthracene-PMDA, the exciton formation and the induced static dipolemoment are carried out instantaneously just by photon absorption without lattice deformation. When these two or three excitons are created in the neighboring unit cells, two-, and three-excitonic strings are formed by these static dipolar interactions.[10,18] After the charge-transfer exciton is formed, the lattice may be deformed so that the eigenenergy of the exciton as well as the magnitude of induced static dipole moment are a little modified. This effect is observed as appearance of the emission line with a small Stokes shift in addition to that of the original charge-transfer exciton.[19] As a result, the static dipole moment as well as the dipolar interaction works just after the formation of the Frenkel as well as charge-transfer excitons. This may be shorter than a picosecond when we use the femtosecond laser pulses to prepare the initial state. On the other hand, the superradiant decay time will be longer than a picosecond. As a result, we may expect the present theoretical predictions to be observable.

ACKNOWLEDGMENT

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APPENDIX: TIME-RESOLVED EMISSION SPECTRUM

In this appendix, we derive Eq. (44), the expression of the time-resolved emission spectrum $I(\omega, t; \Delta t)$. The time-resolved emission spectrum $I(\omega, t; \Delta t)$ measures the photon energy with the angular frequency between $\omega - \Delta\omega$ and $\omega + \Delta\omega$, observed for the time interval from $t - \Delta t$ to $t + \Delta t$. Here $\Delta\omega$ is the resolving power of the device for the measurement, so that Δt must satisfy the inequality $\Delta\omega\Delta t \geq \pi$ due to the uncertainty principle. If we regard the radiation field at the position of device $\mathbf{r}$ as a classical field $\mathbf{E}_{\rm cl}(\mathbf{r}, t)$, the time-resolved emission spectrum $I(\omega, t; \Delta t)$ may be considered to be proportional to $|\mathbf{E}^{\omega}_{\rm cl}(\mathbf{r}, t)|^2$, where $\mathbf{E}^{\omega}_{\rm cl}(\mathbf{r}, t)$ is a Fourier component of $\mathbf{E}_{\rm cl}(\mathbf{r}, t)$:

$$\mathbf{E}^{\omega}_{\rm cl}(\mathbf{r}, t) \equiv \int_{-\Delta t/2}^{\Delta t/2} d\tau \mathbf{E}_{\rm cl}(\mathbf{r}, t + \tau) e^{i\omega(t+\tau)}, \qquad (A1)$$

and

$$|\mathbf{E}^{\omega}_{\rm cl}(\mathbf{r}, t)|^2 = \int_{-\Delta t/2}^{\Delta t/2} d\tau_1 \int_{-\Delta t/2}^{\Delta t/2} d\tau_2 \mathbf{E}^{*}_{\rm cl}(\mathbf{r}, t + \tau_1) \mathbf{E}_{\rm cl}(\mathbf{r}, t + \tau_2) e^{-i\omega(\tau_1 - \tau_2)}. \qquad (A2)$$

Since the measurements are quantum mechanical ones, we have to replace $\mathbf{E}^{*}_{\rm cl}(\mathbf{r}, t + \tau_1)\mathbf{E}_{\rm cl}(\mathbf{r}, t + \tau_2)$ by the time correlation function of the field operators $\langle \hat{\mathbf{E}}^{-}(\mathbf{r}, t + \tau_1)\hat{\mathbf{E}}^{+}(\mathbf{r}, t + \tau_2)\rangle$, that is, the time-resolved emission spectrum $I(\omega, t; \Delta t)$ can be written except for some constant C as

$$I(\omega, t; \Delta t) = C \times \int_{-\Delta t/2}^{\Delta t/2} d\tau_1 \times \int_{-\Delta t/2}^{\Delta t/2} d\tau_2 \langle \hat{\mathbf{E}}^{-}(\mathbf{r}, t + \tau_1)\hat{\mathbf{E}}^{+}(\mathbf{r}, t + \tau_2)\rangle \times e^{-i\omega(\tau_1 - \tau_2)}. \qquad (A3)$$

Since the radiation field obeys adiabatically equations of motion for the electronic system in the superradiant configuration, it is intuitively obvious that $\langle \hat{\mathbf{E}}^{-}(\mathbf{r}, t + \tau_1)\hat{\mathbf{E}}^{+}(\mathbf{r}, t + \tau_2)\rangle$ is linearly proportional to the correlation function of the electronic polarization operators $\langle \hat{S}^{+}(t + \tau_1)\hat{S}^{-}(t + \tau_2)\rangle$. The derivation of this fact is, however, not so easy. Here we adopt the approach developed by Bonifacio *et al.*,[5] and give a formal proof.

A time correlation function can be obtained from equal time correlation functions by virtue of the quantum regression theorem[14] as is described in Sec. IV, so that it is enough to obtain the expression of the equal time correlation function of the field operators:

$$\begin{aligned}\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle &\equiv \mathrm{Tr}[(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rho_{\mathrm{tot}}(t)]\\ &= \mathrm{Tr}[(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\Delta\rho(t)]. \end{aligned}\tag{A4}$$

Here, $\Delta\rho(t)$ is defined as

$$\begin{aligned}&\Delta\rho(t) \equiv \rho_{\mathrm{tot}}(t) - \rho_0(t),\\ &\rho_0(t) \equiv |\mathrm{vac}\rangle\langle\mathrm{vac}| \otimes \rho(t).\end{aligned}\tag{A5}$$

In deriving the superradiance master equation (16) for the atomic system, we could neglect this deviation $\Delta\rho(t)$ of the density operator $\rho(t)$. We need, however, $\Delta\rho(t)$ for the calculation of the evolution of the field.

We define the projection operator $\hat{P}$:[5]

$$\hat{P} \equiv |\mathrm{vac}\rangle\langle\mathrm{vac}| \otimes \mathrm{Tr}_{\mathrm{rad}},\tag{A6}$$

which satisfies

$$\begin{aligned}&\hat{P}\rho_{\mathrm{tot}} = \rho_0(t),\\ &\Delta\rho(t) = (1-\hat{P})\rho_{\mathrm{tot}}(t)(\equiv \hat{Q}\rho_{\mathrm{tot}}(t)).\end{aligned}\tag{A7}$$

We also define Liouville operator $-\hat{L}$ as

$$\begin{aligned}&-\hat{L} \equiv -\hat{L}_0 - \hat{L}',\\ &-\hat{L}_0\rho \equiv \frac{1}{i\hbar}[H_0 + H_{\mathrm{rad}}, \rho],\\ &-\hat{L}'\rho \equiv \frac{1}{i\hbar}[V,\rho],\end{aligned}\tag{A8}$$

where H_0, H_{rad}, and V are defined in Eqs. (6)–(8) in the text. By noticing the equality $-\hat{L}_0\rho_0(t) = 0$, we obtain

$$\Delta\rho(t) = -\int_0^t ds e^{-(1-\hat{P})\hat{L}s}(1-\hat{P})\hat{L}\rho_0(t-s).\tag{A9}$$

Thus, we can express $\Delta\rho(t)$ in terms of $\rho_0(t)$.

If we define $\hat{U}(s) \equiv \exp[-(1-\hat{P})\hat{L}_0 s]$, Eq. (A9) can be expanded as

$$\Delta\rho(t) = \sum_{j=0}^{\infty}(-1)^{j+1}\int_0^t ds\int_0^s ds_1\cdots\int_0^{s_{j-1}} ds_j\hat{U}(s-s_1)\hat{Q}\hat{L}'\hat{U}(s_1-s_2)\hat{Q}\hat{L}'\cdots\hat{U}(s_{j-1}-s_j)\hat{Q}\hat{L}'\hat{U}(s_j)\hat{Q}\hat{L}'\rho_0(t-s).\tag{A10}$$

With this expression of $\Delta\rho(t)$, the time correlation function Eq. (A4) is given as

$$\begin{aligned}\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle = &\sum_{j=0}^{\infty}(-1)^{j+1}\int_0^t ds\int_0^s ds_1\cdots\int_0^{s_{j-1}} ds_j\\ &\times\mathrm{Tr}[(\hat{\mathbf{E}}^-(\mathbf{r}))^m(\hat{\mathbf{E}}^+(\mathbf{r}))^l\hat{U}(s-s_1)\hat{Q}\hat{L}'\cdots\hat{U}(s_{j-1}-s_j)\hat{Q}\hat{L}'\hat{U}(s_j)\hat{Q}\hat{L}'\rho_0(t-s)].\end{aligned}\tag{A11}$$

Since $(\hat{\mathbf{E}}^-(\mathbf{r}))^m(\hat{\mathbf{E}}^+(\mathbf{r}))^l$ is a linear combination of the operators with $ma_*^\dagger$ and la_* and $\mathrm{Tr}[\hat{O}_1\hat{L}'\hat{O}_2] = -\mathrm{Tr}[(\hat{L}'\hat{O}_1)\hat{O}_2]$, we can show the lowest $m+l$ terms in the expansion Eq. (A11) vanish. Therefore, by taking the largest contribution, $(m+l-1)$th term, in that expansion and denoting $n \equiv m+l-1$, we obtain

$$\begin{aligned}\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle \simeq &(-1)^{n+1}\int_0^t ds\int_0^s ds_1\cdots\int_0^{s_{n-1}} ds_n\\ &\times\mathrm{Tr}[(\hat{\mathbf{E}}^-(\mathbf{r}))^m(\hat{\mathbf{E}}^+(\mathbf{r}))^l\hat{U}(s-s_1)\hat{Q}\hat{L}'\cdots\hat{U}(s_{n-1}-s_n)\hat{Q}\hat{L}'\hat{U}(s_n)\hat{Q}\hat{L}'\rho_0(t-s)].\end{aligned}\tag{A12}$$

In this expression, $\hat{Q} = 1 - |\mathrm{vac}\rangle\langle\mathrm{vac}| \otimes \mathrm{Tr}_{\mathrm{rad}}$ can be replaced by 1, because any term with $|\mathrm{vac}\rangle\langle\mathrm{vac}|$ vanishes. For the same reason, $\hat{U}$ can be replaced by $\hat{U}_0$:

$$\hat{U}_0(t) \equiv \exp[-\hat{L}_0 t].\tag{A13}$$

Thus, Eq. (A12) is written as

$$\begin{aligned}\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle \simeq &(-1)^{n+1}\int_0^t ds\int_0^s ds_1\cdots\int_0^s {}_{n-1}ds_n\\ &\times\mathrm{Tr}[(\hat{\mathbf{E}}^-(\mathbf{r}))^m(\hat{\mathbf{E}}^+(\mathbf{r}))^l\hat{U}_0(s-s_1)\hat{L}'\cdots\hat{U}_0(s_{n-1}-s_n)\hat{L}'\hat{U}_0(s_n)\hat{L}'\rho_0(t-s)].\end{aligned}\tag{A14}$$

Using the interaction representation as in Sec. II, we rewrite Eq. (A14) as

$$\begin{aligned}\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle \simeq &(-1)^{n+1}\int_0^t ds\int_0^s ds_1\cdots\int_0^{s_{n-1}} ds_n\\ &\times\mathrm{Tr}[\tilde{\hat{L}}'(t-s)\tilde{\hat{L}}'(t-s+s_n)\cdots\tilde{\hat{L}}'(t-s+s_1)(\tilde{\hat{\mathbf{E}}}^-(\mathbf{r},t))^m(\tilde{\hat{\mathbf{E}}}^+(\mathbf{r},t))^l\tilde{\rho}_0(t-s)].\end{aligned}\tag{A15}$$

Here, we used the relation $\tilde{\rho}(t) = \hat{U}_0(-t)\ \rho(t)$.

Because $\mathrm{Tr}[\cdots\rho_0(t)] = \mathrm{Tr}_{\mathrm{atom}}\,[\mathrm{Tr}_{\mathrm{rad}}\cdots|\mathrm{vac}\rangle\langle\mathrm{vac}|]\;\tilde{\rho}(t)]$, we first evaluate

$$\mathrm{Tr}_{\mathrm{rad}}[\tilde{\hat{L}}'(t-s)\tilde{\hat{L}}'(t-s+s_{n+1})\cdots\tilde{\hat{L}}'(t-s+s_1)(\tilde{\hat{\mathbf{E}}}^-(\mathbf{r},t))^m(\tilde{\hat{\mathbf{E}}}^+(\mathbf{r},t))^l|\mathrm{vac}\rangle\langle\mathrm{vac}|]$$

$$= \langle\mathrm{vac}|\tilde{\hat{L}}'(t-s)\tilde{\hat{L}}'(t-s+s_{n+1})\cdots\tilde{\hat{L}}'(t-s+s_1)(\tilde{\hat{\mathbf{E}}}^-(\mathbf{r},t))^m(\tilde{\hat{\mathbf{E}}}^+(\mathbf{r},t))^l|\mathrm{vac}\rangle. \tag{A16}$$

Substituting the concrete expression of $\tilde{V}(t)$ and the formula $\tilde{\hat{L}}(t)X = \frac{1}{i\hbar}[\tilde{V}(t),X]$, Eq. (A16) is rewritten as

$$\left(\frac{i}{\hbar}\right)^{n+1}(-1)^l{\sum_\sigma}'\sum_{j_1,j_2,\cdots,j_{n+1}}\left[\prod_{\alpha=1}^{m}\tilde{\hat{\mathbf{P}}}^+(\mathbf{r}_{j_\alpha},t-s+s_{\sigma(\alpha)})\right]\left[\prod_{\beta=m+1}^{n+1}\tilde{\hat{\mathbf{P}}}^-(\mathbf{r}_{j_\beta},t-s+s_{\sigma(\beta)})\right]$$

$$\times\left\langle\mathrm{vac}\left|\left[\prod_{\alpha=1}^{m}\tilde{\hat{\mathbf{E}}}^+(\mathbf{r}_{j_\alpha},t-s+s_{\sigma(\alpha)})\right](\tilde{\hat{\mathbf{E}}}^-(\mathbf{r},t))^m(\tilde{\hat{\mathbf{E}}}^+(\mathbf{r},t))^l\left[\prod_{\beta=m+1}^{n+1}\tilde{\hat{\mathbf{E}}}^-(\mathbf{r}_{j_\beta},t-s+s_{\sigma(\beta)})\right]\right|\mathrm{vac}\right\rangle, \tag{A17}$$

where we neglect the off-resonant coupling between the field and the polarization, and ${\sum_\sigma}'$ denotes all the permutation under the constraint: $\sigma(1) > \sigma(2) > \cdots > \sigma(m)$ and $\sigma(m+1) < \sigma(m+2) < \cdots < \sigma(n+1)$, and $\tilde{\hat{\mathbf{P}}}^\pm(\mathbf{r}_j,t)$ is the interaction representation of the jth polarization operator $\boldsymbol{\mu}\hat{s}_j^\pm$.

The field operators satisfy the commutation relation:

$$[\tilde{\hat{\mathbf{E}}}^+(\mathbf{r},t),\tilde{\hat{\mathbf{E}}}^-(\mathbf{r}',t')] = \sum_{\mathbf{k},\boldsymbol{\epsilon}}\boldsymbol{\epsilon\epsilon}\frac{\hbar ck}{2\varepsilon\Omega}e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')-ick(t-t')}. \tag{A18}$$

Hence Eq. (A17) is transformed by the Wick's theorem to

$$\left(\frac{i}{\hbar}\right)^{n+1}(-1)^l\,l!\,m!{\sum_\sigma}'\sum_{j_1,j_2,\cdots,j_{n+1}}\sum_{\mathbf{k}_1,\boldsymbol{\epsilon}_1,\mathbf{k}_2,\boldsymbol{\epsilon}_2,\cdots\mathbf{k}_{n+1},\boldsymbol{\epsilon}_{n+1}}\left[\prod_{\alpha=1}^{m}\boldsymbol{\epsilon}_\alpha\cdot\hat{\mathbf{P}}^+(\mathbf{r}_{j_\alpha})\right]\left[\prod_{\beta=m+1}^{n+1}\boldsymbol{\epsilon}_\beta\cdot\hat{\mathbf{P}}^-(\mathbf{r}_{j_\beta})\right]$$
$$\times\exp\left[i\omega_0\left\{(m-l)(t-s)+\sum_{\alpha=1}^{m}s_{\sigma(\alpha)}-\sum_{\beta=m+1}^{n+1}s_{\sigma(\beta)}\right\}\right]$$
$$\times\exp\left[\left\{-ic\sum_{\alpha=1}^{m}k_\alpha(-s+s_{\sigma(\alpha)})+ic\sum_{\beta=m+1}^{n+1}k_\beta(-s+s_{\sigma(\beta)})\right\}\right]$$

$$\times\boldsymbol{\epsilon}_1\boldsymbol{\epsilon}_2\cdots\boldsymbol{\epsilon}_{n+1}\left(\frac{\hbar c}{2\epsilon\Omega}\right)^{n+1}\prod_{j=1}^{n+1}k_j\exp\left[i\left\{\sum_{\alpha=1}^{m}\mathbf{k}_\alpha(\mathbf{r}-\mathbf{r}_{j_\alpha})-\sum_{\beta=m+1}^{n+1}\mathbf{k}_\beta(\mathbf{r}-\mathbf{r}_{j_\beta})\right\}\right]. \tag{A19}$$

We take the origin of the coordinates at the center of the system and assume, as in the text, that the length of the system is short in comparison with the relevant wavelength of the radiation field, i.e., we take $\mathbf{r}-\mathbf{r}_j \simeq \mathbf{r}$. Then, using the second Born and Markov approximation just as in Sec. II and neglecting a time delay $|\mathbf{r}|/c$, we obtain

$$\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle = C_0(\mathbf{r})\times e^{-i\omega_0(m-l)t}\sum_{j_1,j_2,\ldots,j_{n+1}}\mathrm{Tr}[\hat{P}^+(\mathbf{r}_{j_1})\cdots\hat{P}^+(\mathbf{r}_{j_m})\hat{P}^-(\mathbf{r}_{j_{m+1}})\cdots\hat{P}^-(\mathbf{r}_{j_{n+1}})\rho(t)]. \tag{A20}$$

Here, $\hat{P}^\pm(\mathbf{r}_j) = \mu\hat{s}_j^\pm$ and $C_0(\mathbf{r})$ is a constant which is essentially the product of the strength of the radiation from the total transition dipole moments at the position of the device $\mathbf{r}$. We, however, do not need its explicit form here. The important result is that the field-field correlation function $\langle(\hat{\mathbf{E}}^-(\mathbf{r},t))^m(\hat{\mathbf{E}}^+(\mathbf{r},t))^l\rangle$ is proportional to $\langle(\hat{S}^+(t))^m(\hat{S}^-(t))^l\rangle$. From the quantum regression theorem, we can easily show the time correlation function of the field $\langle\hat{\mathbf{E}}^-(\mathbf{r},t+\tau_1)\hat{\mathbf{E}}^+(\mathbf{r},t+\tau_2)\rangle$ is also proportional to $\langle\hat{S}^+(t+\tau_1)\hat{S}^-(t+\tau_2)\rangle$. Thus we find

$$I(\omega,t;\Delta t) \propto \int_{-\Delta t/2}^{\Delta t/2}d\tau_1\int_{-\Delta t/2}^{\Delta t/2}d\tau_2\langle\hat{S}^+(t+\tau_1)\times\hat{S}^-(t+\tau_2)\rangle e^{-i\omega(\tau_1-\tau_2)}. \tag{A21}$$

The proportional constant should be so chosen that the following relation holds:

$$I(t) = \lim_{\Delta t\to 0}\int d\omega I(\omega,t;\Delta t). \tag{A22}$$

Thus, finally, we obtain the expression of $I(\omega,t;\Delta t)$ as Eq. (44).

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