

Mesoscopic cooperative emission from a disordered system

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We study theoretically the cooperative light emission from a system of $N \gg 1$ classical oscillators confined within a volume with spatial scale L much smaller than the radiation wavelength $\lambda_0 = 2\pi c/\omega_0$. We assume that the oscillator frequencies are randomly distributed around a central frequency ω_0 with some characteristic width $\Omega \ll \omega_0$. In the absence of disorder, that is, $\Omega = 0$, the cooperative emission spectrum is composed of a narrow subradiant peak superimposed on a wide superradiant band. When $\Omega \neq 0$, we demonstrate that if N is large enough, the subradiant peak is not simply broadened by the disorder but rather *splits into a system of random narrow peaks*. We estimate the spectral width of these peaks as a function of N , L , Ω , and λ_0 . We also estimate the amplitude of this mesoscopic structure in the emission spectrum.

I. INTRODUCTION

The study of cooperative phenomena in optics was initiated by the pioneering work of Dicke.¹ The underlying physics of the cooperative emission can be readily understood using a classical approach. Suppose that a large number N of identical oscillators with frequency ω_0 are confined within a small volume with characteristic size $L \ll 2\pi c/\omega_0 = \lambda_0$, where λ_0 is the radiation wavelength; this is referred to as a ‘‘point’’ sample.² If τ is the radiative lifetime of an isolated oscillator, then according to Dicke, the N eigenmodes of the system of oscillators consist of one mode with a short lifetime $\tau/N \ll \tau$, and $N-1$ modes with lifetimes much longer than τ [by a factor $\sim (\lambda_0/L)^2$]. Correspondingly, the emission spectrum of this system consists of superimposed broad (superradiant) bands and a narrow (subradiant) band. The intensity ratio of these bands is determined by the details of the excitation. This type of lifetime redistribution is caused by the interactions among the oscillators through their radiation fields.

Certainly, the classical picture does not describe all aspects of the cooperative emission. In fact, the original work of Dicke¹ primarily addressed the time evolution of the radiation emission, provided that at the initial moment, $t=0$, all the oscillators are coherently excited. For this situation, the classical picture helps in understanding that the radiation is released during a short time, τ/N ; understanding of the initial stages of the emission process (the delay time statistics) requires, however, a quantum description.^{3,4} The original treatment in Ref. 1 also ignored dipole-dipole interactions, which give rise to a spread in the oscillator frequencies (dephasing). The question whether or not this dephasing would completely destroy the cooperative emission is very nontrivial and was addressed in a number of later works.⁵⁻⁷

In the previous considerations of cooperative emission, it was assumed that all N oscillators (atoms, molecules, or excitons) have the same frequencies. Such a restriction was adequate for the experimental situation in both gases and single crystals. To the best of our knowledge, the only ac-

count of disorder in the frequencies of the oscillators was given in Ref. 8, which addressed the transient behavior of the cooperative emission from molecular aggregates. The case of J aggregates corresponds to a symmetrical arrangement of oscillators in a circle. The authors⁸ treated the disorder within perturbation theory and averaged the second-order correction to the decay rates (the first-order correction vanishes upon averaging) with a Gaussian distribution. The advantage of the work in Ref. 8 is that the nearest-neighbor dipole-dipole interactions were taken into account exactly. The drawback is in the perturbative approach, which rules out certain *qualitative* physical effects (see below).

Whereas Ref. 8 addressed a rather particular situation, the following *general* questions might be asked. Suppose that the oscillator frequencies are randomly distributed with a characteristic width Ω . Obviously, as Ω increases, it would eventually destroy the cooperative features in the emission spectrum. Then what is the critical magnitude of Ω ? How does this magnitude depend on the parameters of the system N , L , and λ_0 ? What is the structure of the emission spectrum when disorder is smaller than critical?

These questions have become not purely academic due to the recent advances in the field of laser action in π -conjugated polymers.⁹⁻¹⁵ Some experiments provide strong evidence for cooperative emission from an ensemble of excitons in these materials for excitation intensities exceeding a certain characteristic threshold.¹¹⁻¹⁵ On the other hand, it is well known that the films of π -conjugated polymers are strongly disordered¹⁶ (in the absence of disorder, cooperative emission by polymer chain was considered in Ref. 17). They contain impurities and defects that break the polymer chains into segments of relatively short conjugation length with a distribution depending on the film quality.¹⁸ This has a direct effect on the exciton energy $\hbar\omega$, since it has been found that $\hbar\omega$ directly depends on the chain conjugation length.¹⁹

The questions formulated above are addressed in the present paper. We study here the effect of disorder on the cooperative emission spectrum of the system of classical os-

cillators. We consider the situation of incoherent excitation, which is most relevant to experiment.^{11–15} In contrast to Ref. 8, we are interested in the nonaveraged (but universal) properties of the emission spectrum. In other words, our goal is to assess the *mesoscopic*²⁰ aspects of the cooperative emission. By mesoscopic we mean that in the presence of a disorder, the emission spectrum of a large number of oscillators develops a fine structure. The actual shape of this spectral structure represents the *fingerprints* of the distribution of the oscillator frequencies and positions for a given realization. At the same time, the characteristic period and amplitude of the fine structure are determined by the net parameters of the system: N , L , and Ω .

The paper is organized as follows. In Sec. II we derive the expression for the emission spectrum of a system of classical oscillators coupled by their radiation fields. In Sec. III we study in detail a simplified model in which the coupling among the oscillators is independent of distance. The eigenmodes of a “point” sample in the presence of disorder are analyzed in Sec. IV. The universal properties of the mesoscopic structure in the emission spectrum for small and large (but still smaller than λ_0) sizes L are discussed in Secs. V and VI, respectively. In Sec. VII the effect of the dipole-dipole interactions is addressed. The conclusions are given in Sec. VIII.

II. THE BASIC EQUATIONS

We consider a system of N oscillators located at random points \mathbf{r}_i , with frequencies ω_i randomly distributed around a central frequency ω_0 with a characteristic width Ω . Each oscillator is driven by the radiation field $\mathbf{E}(\mathbf{r}, t)$ produced by all oscillators. The equation of motion for the displacement u_i of a given oscillator i reads

$$\ddot{u}_i + \omega_i^2 u_i = \frac{e}{m} \mathbf{n}_i \cdot \mathbf{E}(\mathbf{r}_i, t), \quad (1)$$

where e and m are the dipole characteristics (effective charge and mass) and \mathbf{n}_i is a unit vector in the direction of the dipole moment.

The current density, associated with the oscillators motion, can be written as

$$\mathbf{J}(\mathbf{r}, t) = e \sum_i \mathbf{n}_i \dot{u}_i \delta(\mathbf{r} - \mathbf{r}_i). \quad (2)$$

The current \mathbf{J} plays the role of a source, which generates the electric field $\mathbf{E}(\mathbf{r}, t)$ according to

$$\Delta \mathbf{E} - \frac{1}{c^2} \ddot{\mathbf{E}} = \frac{4\pi}{c^2} \mathbf{J}, \quad (3)$$

where c is the speed of light.

Within the classical approach, the emission spectrum of the system should be calculated as follows. We assume that at the initial moment, $t=0$, all oscillators are excited with different phases ϕ_i , and that the radiation field at the initial moment is zero, $\mathbf{E}(\mathbf{r}, 0) = \mathbf{0}$. The evolution of \mathbf{E} with time can be then obtained by solving Eqs. (1)–(3). After taking the limit $r \rightarrow \infty$ and expanding the field into harmonics, the spectral intensity can be obtained as $I(\omega) = |\mathbf{E}(\infty, \omega)|^2$.

To carry out this program, it is convenient to employ the Laplace transformation. The transformed functions $\bar{u}_i(p)$ and $\bar{\mathbf{E}}(\mathbf{r}, p)$ satisfy the following system of equations:

$$(\omega_i^2 + p^2) \bar{u}_i(p) = \frac{e}{m} \mathbf{n}_i \cdot \bar{\mathbf{E}}(\mathbf{r}_i, p) + u_0(p \cos \phi_i - \omega_i \sin \phi_i), \quad (4)$$

$$\Delta \bar{\mathbf{E}}(\mathbf{r}, p) - \frac{p^2}{c^2} \bar{\mathbf{E}}(\mathbf{r}, p) = \frac{4\pi e}{c^2} \sum_i \mathbf{n}_i [p^2 \bar{u}_i(p) - u_0(p \cos \phi_i - \omega_i \sin \phi_i)] \delta(\mathbf{r} - \mathbf{r}_i), \quad (5)$$

where $u_0 \sin \phi_i$ and $\omega_i u_0 \cos \phi_i$ are the respective initial displacement and velocity of the i th oscillator. The solution of Eq. (5) for $\bar{\mathbf{E}}(\mathbf{r}, p)$ can be presented as a superposition of eigenmodes $\mathbf{E}_\nu(\mathbf{r})$ of the wave equation for the electromagnetic field,

$$\Delta \mathbf{E}_\nu(\mathbf{r}) + \frac{\omega_\nu^2}{c^2} \mathbf{E}_\nu(\mathbf{r}) = \mathbf{0}, \quad (6)$$

where ω_ν is the eigenfrequency. Assuming that the modes are normalized, $\int d\mathbf{r} \mathbf{E}_\nu^2(\mathbf{r}) = 1$, we obtain the following expression for $\bar{\mathbf{E}}(\mathbf{r}, p)$:

$$\bar{\mathbf{E}}(\mathbf{r}, p) = -4\pi e \sum_{i\nu} [p^2 \bar{u}_i(p) - u_0(p \cos \phi_i - \omega_i \sin \phi_i)] \frac{\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{r}_i)}{\omega_\nu^2 + p^2} \mathbf{E}_\nu(\mathbf{r}). \quad (7)$$

Substituting Eq. (7) into Eq. (4), we get a system of coupled equations for the amplitudes $\bar{u}_i(p)$:

$$\begin{aligned} (\omega_i^2 + p^2) \bar{u}_i(p) = & -\frac{4\pi e^2}{m} \sum_{j\nu} \frac{[\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{r}_i)][\mathbf{n}_j \cdot \mathbf{E}_\nu(\mathbf{r}_j)]}{\omega_\nu^2 + p^2} \\ & \times [p^2 \bar{u}_j(p) - u_0(p \cos \phi_j - \omega_j \sin \phi_j)] \\ & + u_0(p \cos \phi_i - \omega_i \sin \phi_i). \end{aligned} \quad (8)$$

To simplify Eq. (8), it is convenient to introduce new variables $v_i(p)$:

$$v_i(p) = \frac{p^2 \bar{u}_i(p)}{u_0} - (p \cos \phi_i - \omega_i \sin \phi_i). \quad (9)$$

Then Eq. (8) takes the form

$$(\omega_i^2 + p^2) v_i + \sum_j S_{ij} v_j = \omega_i^2 (\omega_i \sin \phi_i - p \cos \phi_i), \quad (10)$$

where the coefficients

$$S_{ij}(p) = \frac{4\pi e^2 p^2}{m} \sum_\nu \frac{[\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{r}_i)][\mathbf{n}_j \cdot \mathbf{E}_\nu(\mathbf{r}_j)]}{\omega_\nu^2 + p^2} \quad (11)$$

describe the coupling between oscillators i and j via the radiation field.

Let us now express the intensity $I(\omega)$ in terms of the variables $v_i(p)$. The expression for $\bar{\mathbf{E}}(\mathbf{r}, p)$ follows from Eqs. (7) and (9):

$$\bar{\mathbf{E}}(\mathbf{r}, p) = -4\pi e \sum_{i\nu} v_i(p) \frac{\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{r}_i)}{\omega_\nu^2 + p^2} \mathbf{E}_\nu(\mathbf{r}). \quad (12)$$

The Fourier transform of the electric field is obtained by replacing p by $i\omega$ in Eq. (12). In the limit $r \rightarrow \infty$, only the pole $\omega_\nu = \omega$ contributes to the sum over ν , so that

$$\mathbf{E}(\mathbf{r}, \omega)|_{r \rightarrow \infty} \propto \sum_{i\nu} v_i [\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{r}_i)] \mathbf{E}_\nu(\mathbf{r}) \delta(\omega_\nu^2 - \omega^2). \quad (13)$$

This corresponds to taking the continuum limit for electromagnetic modes. The terms proportional to $\mathbf{E}_\nu(\mathbf{r}) \cdot \mathbf{E}_\mu(\mathbf{r})$, which appear after calculating $|\mathbf{E}(\mathbf{r}, \omega)|^2$ from Eq. (13), oscillate rapidly if $\mu \neq \nu$. Therefore, only the terms with $\mu = \nu$ survive at large r . These terms contain products of the form $[\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{r}_i)] [\mathbf{n}_j \cdot \mathbf{E}_\nu(\mathbf{r}_j)]$. Note that the same products enter into the coupling coefficients S_{ij} , defined by Eq. (11). This allows us to present the final expression for the spectral intensity in a compact form,

$$I(\omega) \propto \sum_{ij} v_i (\text{Im} S_{ij}) v_j^*, \quad (14)$$

where v_i and S_{ij} are calculated at $p = i\omega$.

We assume that the spread of the oscillator frequencies due to the disorder is much smaller than the central frequency, $\Omega \ll \omega_0$. This means that the frequency dependence of the coupling constants is weak, so that $S_{ij}(i\omega)$ can be evaluated at $\omega = \omega_0$. The real part of S_{ij} , which comes from the principal value of the sum over modes in Eq. (11), diverges for $i = j$. This divergency is the manifestation of the Lamb shift, well known in quantum electrodynamics, and can be absorbed into ω_i . At the same time, the imaginary part of S_{ii} , which results from the pole $\omega_\nu = \omega_0$, is finite. It determines the radiative lifetime τ of an individual oscillator via the relation $\text{Im} S_{ii}(i\omega_0) = 2\omega_0/\tau$. For a single oscillator in vacuum, the modes \mathbf{E}_ν are simply plane waves, and the summation over ν in Eq. (11) recovers the textbook result

$$\tau = \frac{3mc^3}{e^2 \omega_0^2}. \quad (15)$$

For $i \neq j$, the coupling S_{ij} between two oscillators depends on the ratio r_{ij}/λ_0 , where r_{ij} is the distance between the oscillators, and $\lambda_0 = 2\pi c/\omega_0$ is the radiation wavelength. For $r_{ij} \gg \lambda_0$, both real and imaginary parts of S_{ij} oscillate rapidly with r_{ij} , and the effect of coupling is negligibly small for a large ensemble of oscillators.²¹ For $r_{ij} \ll \lambda_0$, the real part of S_{ij} represents the dipole-dipole interaction of the oscillators i and j . It is convenient to present S_{ij} in the form

$$S_{ij} = \frac{2\omega_0}{\tau} (\beta_{ij} + i\alpha_{ij}), \quad (16)$$

where β_{ij} and α_{ij} are the dimensionless matrices of coupling between the oscillators, defined as

$$\beta_{ij} = \left(\frac{\lambda_0}{2\pi r_{ij}} \right)^3 \left[(\mathbf{n}_i \cdot \mathbf{n}_j) - \frac{3(\mathbf{n}_i \cdot \mathbf{r}_{ij})(\mathbf{n}_j \cdot \mathbf{r}_{ij})}{r_{ij}^2} \right] \quad (17)$$

and

$$\alpha_{ij} = \mathbf{n}_i \cdot \mathbf{n}_j - \frac{1}{5} \left(\frac{2\pi r_{ij}}{\lambda_0} \right)^2 \left[(\mathbf{n}_i \cdot \mathbf{n}_j) - \frac{(\mathbf{n}_i \cdot \mathbf{r}_{ij})(\mathbf{n}_j \cdot \mathbf{r}_{ij})}{2r_{ij}^2} \right]. \quad (18)$$

Turning back to Eq. (10), we note that since the distribution of oscillators frequencies is relatively narrow, that is, $\Omega \ll \omega_0$, we can make some simplifications. Namely, for $p = i\omega$, the factor $\omega_i^2 + p^2$ in the left-hand side (lhs) can be replaced by $2\omega_0(\omega_i - \omega)$, and the right-hand side (rhs) can be written as $-i\omega_0^3 e^{-i\phi_i}$. Finally, after rescaling v_i by factor ω_0^2 , Eq. (10) takes the form

$$(\omega_i - \omega)v_i + \frac{1}{\tau} \sum_j (\beta_{ij} + i\alpha_{ij})v_j = -\frac{i}{2} e^{-i\phi_i}. \quad (19)$$

Equation (19) together with Eqs. (14) and (16)–(18) allows us to calculate the spectral intensity $I(\omega)$ for any set of initial oscillators phases. For $\alpha_{ij} = \beta_{ij} = 0$ ($i \neq j$), the eigenfrequencies of the system are simply the frequencies of individual oscillators, and the emission spectrum represents a superposition of Lorentzian peaks centered at ω_i . In the presence of nondiagonal coupling, the eigenfrequencies are those of *cooperative eigenmodes* which, in turn, are determined by the imaginary part of the coupling α_{ij} . In the experiment, the measured spectrum represents the result of averaging over many excitation pulses.¹³ In order to simulate the experimental situation, we will assume the phases ϕ_i to be uncorrelated random numbers and average the result for the spectral intensity over all ϕ_i .

III. A SIMPLE MODEL

In this section we consider a simplified situation, in which Eq. (19) with random frequencies ω_i can be solved exactly, and the expression for the spectral intensity can be obtained in a closed form. Following Dicke,¹ we disregard the dipole-dipole interactions by setting $\beta_{ij} = 0$. Although this approximation is rather common, later on we will discuss it in more detail. Turning to α_{ij} , we note that since $L^2/\lambda_0^2 \ll 1$, the second term in Eq. (18) is a small correction to the first term. We therefore approximate the *nondiagonal* elements of α_{ij} by replacing r_{ij}^2/λ_0^2 with its average,

$$\alpha_{ij} = \alpha \mathbf{n}_i \cdot \mathbf{n}_j, \quad \alpha_{ii} = 1, \quad (20)$$

where the coupling constant α , with a typical value $(1 - \alpha) \sim L^2/\lambda_0^2 \ll 1$, is the same for *all* pairs. Note however, that the disorder coming from random orientations of \mathbf{n}_i is still included. Later we will use this model for the analysis of the system (19) with realistic α_{ij} .

A. General solution

For the model coupling (20), the system of equations (19) takes the form

$$\left[\omega_i - \omega + \frac{i}{\tau}(1 - \alpha) \right] v_i + \frac{i}{\tau} \alpha \mathbf{n}_i \cdot \mathbf{s} = -\frac{i}{2} e^{-i\phi_i}, \quad (21)$$

with vector \mathbf{s} defined as

$$\mathbf{s} = \sum_{i=1}^N v_i \mathbf{n}_i. \quad (22)$$

A closed equation for \mathbf{s} can be obtained by multiplying v_i , found from Eq. (21), by \mathbf{n}_i and taking the sum over i . This yields

$$\begin{aligned} \mathbf{s} + \frac{i\alpha}{\tau} \sum_i \frac{\mathbf{n}_i (\mathbf{n}_i \cdot \mathbf{s})}{\omega_i - \omega + i(1 - \alpha)/\tau} \\ = -\frac{i}{2} \sum_i \frac{\mathbf{n}_i e^{-i\phi_i}}{\omega_i - \omega + i(1 - \alpha)/\tau}. \end{aligned} \quad (23)$$

Solving Eqs. (21) and (23) for v_i and substituting the result into Eq. (14), we obtain for the spectral intensity after some algebra

$$I(\omega) \propto -\text{Im} \left[f(\omega) - \frac{i\alpha}{\tau} \sum_{\mu\nu} g_{\mu\nu}^- \left(1 + \frac{i\alpha}{\tau} F \right)_{\mu\nu}^- g_{\nu}^+ \right], \quad (24)$$

where we introduced a function

$$f(\omega) = \sum_i \frac{1}{\omega_i - \omega + i(1 - \alpha)/\tau}, \quad (25)$$

a vector

$$g_{\mu}^{\pm}(\omega) = \sum_i \frac{e^{\pm i\phi_i} n_{i\mu}}{\omega_i - \omega + i(1 - \alpha)/\tau}, \quad (26)$$

and a tensor

$$F_{\mu\nu}(\omega) = \sum_i \frac{n_{i\mu} n_{i\nu}}{\omega_i - \omega + i(1 - \alpha)/\tau}, \quad (27)$$

where $n_{i\mu}$ are the components of \mathbf{n}_i .

B. Identical oscillators

Let us first consider the case of N identical oscillators having the same frequencies $\omega_i = \omega_0$, and dipole momenta all aligned in the same direction. Then we find from Eq. (27),

$$F_{\mu\nu}(\omega) = \delta_{\mu\nu} f(\omega) = \frac{\delta_{\mu\nu} N}{\omega_i - \omega + i(1 - \alpha)/\tau}, \quad (28)$$

and after averaging over the initial phases ϕ_i , we obtain from Eq. (24)

$$\begin{aligned} I(\omega) \propto \left[\frac{(N-1)(1-\alpha)/\tau}{(\omega_0 - \omega)^2 + (1-\alpha)^2/\tau^2} \right. \\ \left. + \frac{(1-\alpha + \alpha N)/\tau}{(\omega_0 - \omega)^2 + (1-\alpha + \alpha N)^2/\tau^2} \right]. \end{aligned} \quad (29)$$

The emission spectrum is a superposition of a wide and a narrow Lorentzians with spectral widths $\Gamma \approx N/\tau$ and $\gamma = (1 - \alpha)/\tau$, respectively. In accordance with the classical

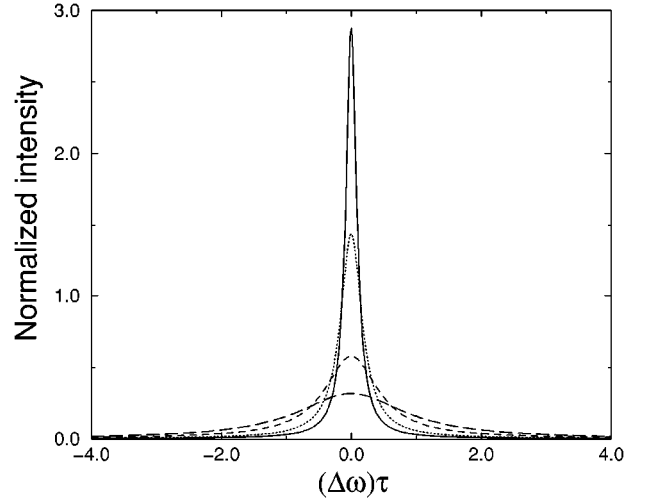


FIG. 1. Spectral intensity of N identical oscillators calculated from Eq. (29) plotted vs $\Delta\omega = \omega - \omega_0$ for $N=10$, and $\alpha=0$ (long-dashed line), $\alpha=0.5$ (dashed line), $\alpha=0.8$ (dotted line), and $\alpha=0.9$ (solid line).

result,¹ the eigenmodes of the system of N identical oscillators coupled via their radiation field represent a single superradiant mode with short radiation time τ/N , and $N-1$ subradiant modes with radiation time much longer than that for an isolated oscillator, $\tau/(1-\alpha) \gg \tau$. The superradiant mode is a symmetric superposition of oscillator states and is strongly coupled to the radiation field, whereas the coupling of the subradiant modes to the radiation field is suppressed. In this case, the frequencies of all $N-1$ subradiant modes are degenerate, and the spectrum consists of a single narrow peak of width γ on top of much broader band of width Γ , as shown in Fig. 1. As can be seen, with decreasing $1-\alpha$, the height of the subradiant peak increases, whereas the amplitude of the superradiant band diminishes.

C. Random frequencies

Consider now the case when the oscillator frequencies are random, but orientational disorder is still absent, i.e., all dipoles are aligned in one direction. Again we have $F_{\mu\nu}(\omega) = \delta_{\mu\nu} f(\omega)$, with $f(\omega) = f'(\omega) + i f''(\omega)$ given by Eq. (25). Then a straightforward evaluation of Eq. (24) yields (after averaging over the phases)

$$I(\omega) \propto \left[\frac{\frac{\alpha}{\tau} f_1' \left(1 - \frac{\alpha}{\tau} f'' \right) + \left(\frac{\alpha}{\tau} \right)^2 f_1'' f_1'}{\left(1 - \frac{\alpha}{\tau} f'' \right)^2 + \left(\frac{\alpha}{\tau} f_1' \right)^2} - f'' \right], \quad (30)$$

where the function $f_1(\omega) = f_1'(\omega) + i f_1''(\omega)$ is defined as

$$f_1(\omega) = \sum_i \frac{1}{[\omega_i - \omega + i(1 - \alpha)/\tau]^2}. \quad (31)$$

In order to clarify the underlying physics, it is useful to express the spectral intensity in terms of the system eigenmodes. The eigenfrequencies $\tilde{\omega}_k$ are determined by the equation

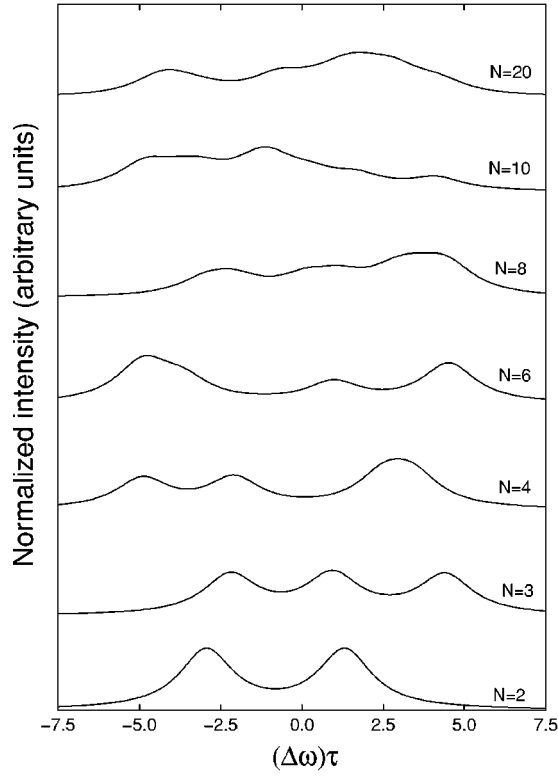


FIG. 2. Uncoupled oscillators: Spectral intensity $I(\omega)$ plotted vs $\Delta\omega = \omega - \omega_0$ for several sets of random oscillator frequencies with $\Omega\tau = 5.0$ and $\alpha = 0$.

$$1 + \frac{i\alpha}{\tau} f(\tilde{\omega}_k) = 0. \quad (32)$$

Then the intensity (30) can be simply rewritten as

$$I(\omega) \propto \sum_k \frac{\tilde{\omega}_k''}{(\omega - \tilde{\omega}_k')^2 + \tilde{\omega}_k''^2}, \quad (33)$$

where $\tilde{\omega}_k' = \text{Re } \tilde{\omega}_k$ is the eigenmode frequency and $\tilde{\omega}_k'' = \text{Im } \tilde{\omega}_k$ characterizes its width. Note that for $\omega_i = \omega_0$, we have $N-1$ degenerate eigenmodes with $\tilde{\omega}_k' = \omega_0$, and Eq. (33) turns into Eq. (29).

D. Disorder in orientations

In the presence of the orientational disorder, the spectral intensity (24) depends, in principle, on the direction of each \mathbf{n}_i . However, for large N , one can replace the product $n_{i\mu}n_{i\nu}$ in Eq. (27) for $F_{\mu\nu}$, with its average,

$$\langle n_{i\mu}n_{i\nu} \rangle = \frac{1}{3} \delta_{\mu\nu}. \quad (34)$$

Thus, we have $F_{\mu\nu}(\omega) = \frac{1}{3} \delta_{\mu\nu} f(\omega)$, so that the expression for the spectral intensity is similar to Eq. (30) with the only difference that in the first term, the functions $f(\omega)$ and $f_1(\omega)$ are now multiplied by $1/3$. This results in a shrinkage of the superradiant emission band by the same factor. At the same time, the width of subradiant peak increases by a factor of 3. Thus, the orientational disorder has *no* qualitative effect on the cooperative emission spectrum. The reason is that the

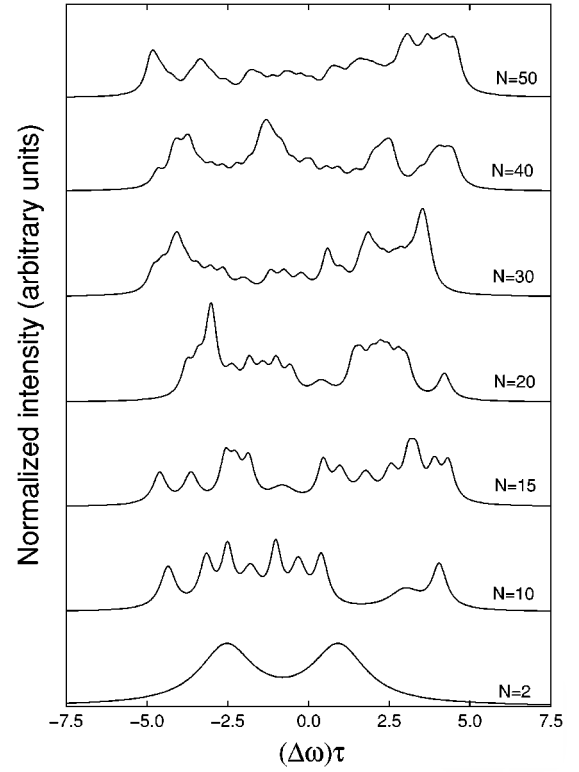


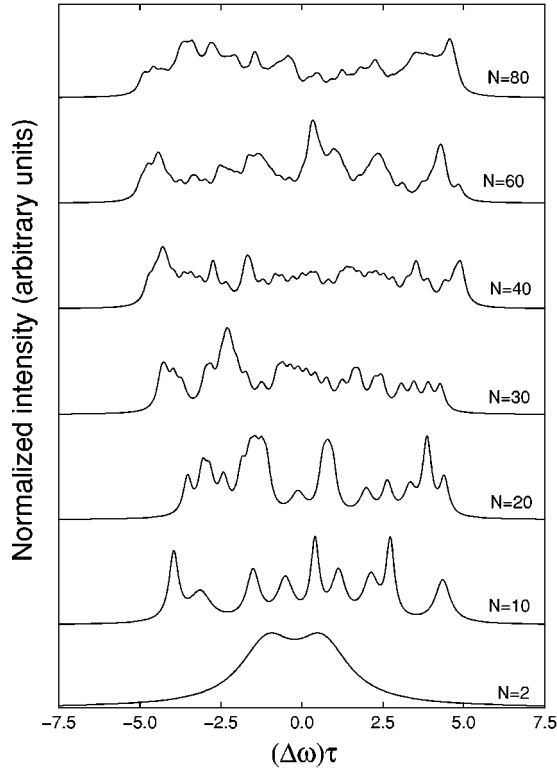
FIG. 3. Coupled oscillators: Spectral intensity $I(\omega)$ calculated from Eq. (30) for several sets of random oscillator frequencies with $\Omega\tau = 5.0$ and $\alpha = 0.8$.

coupling (20) is *separable*, that is, it depends on orientations via the product $\mathbf{n}_i \cdot \mathbf{n}_j$. Furthermore, for realistic α_{ij} given by Eq. (18), the main (first) term has the same separable form; therefore, the orientational disorder does not qualitatively affect the cooperative emission spectrum and will be disregarded in the rest of the paper.

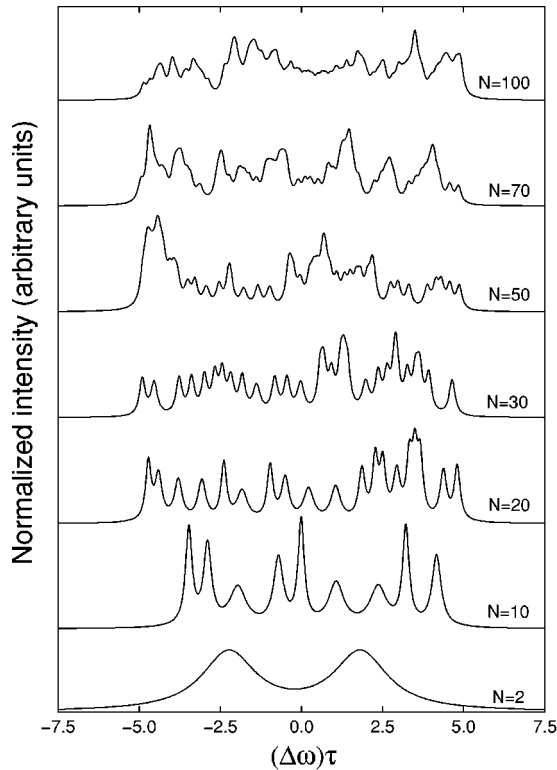
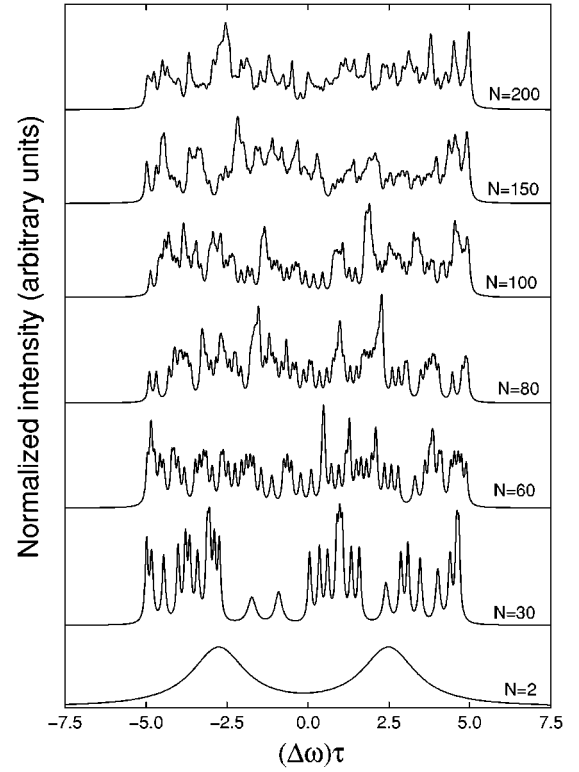
E. Numerical results

In Fig. 2 we plot the normalized spectral intensity in the absence of coupling, i.e., $\alpha = 0$, with increasing number of oscillators. Each spectrum is calculated with a computer generated set of N random frequencies ω_i , which we have chosen, for simplicity, to be uniformly distributed in the interval $(\omega_0 - \Omega, \omega_0 + \Omega)$. For convenience, the spectra corresponding to different N are normalized and shifted in the vertical direction. It can be seen that the peaks are resolved in the spectrum as long as the disorder Ω is larger than N/τ . We also see that for sufficiently large N , the intensity peaks are washed out from the spectrum.

In Figs. 3–6 we present the results for $I(\omega)$ calculated using Eq. (30) for several values of α close to 1. The striking feature of the emission spectrum is its *mesoscopic* character. In the presence of disorder, the narrow subradiant peak of Eq. (29) (see Fig. 1) is not smeared out due to a large spread in ω_i , as in the case of uncoupled oscillators (see Fig. 2), but rather splits into a *multitude of narrow peaks* corresponding to the eigenmodes of the disordered system. Furthermore, although the curves are calculated with different random sets of frequencies, the overall pattern of the emission spectrum exhibits certain universal features. In particular, it can be seen by comparing Figs. 3–6 that with increasing N , the

FIG. 4. Same as in Fig. 3, but for $\alpha = 0.85$.

random structure of the spectrum undergoes several transformations, and that the characteristic N , at which the changes in the pattern occur, is sensitive to the proximity of α to 1. This indicates a rather nontrivial structure of the eigenmodes, which we address in the next section.

FIG. 5. Same as in Fig. 3, but for $\alpha = 0.9$.FIG. 6. Same as in Fig. 3, but for $\alpha = 0.95$.

IV. STRUCTURE OF EIGENMODES

The eigenmodes of a system of N oscillators coupled through their radiation field are determined by the homogeneous part of Eq. (19) (we set $\beta_{ij} = 0$ in this section),

$$(\omega_i - \omega)v_i + \frac{i}{\tau} \sum_j \alpha_{ij} v_j = 0. \quad (35)$$

Since the typical values of $(1 - \alpha_{ij}) \sim r_{ij}^2 / \lambda_0^2$ are small, we split the second term in Eq. (35) into a sum of the main contribution, with $\alpha_{ij} = 1$, and a correction proportional to $\alpha_{ij} - 1$. Analogously to the consideration in the previous section, we rewrite Eq. (35) as

$$(\omega_i - \omega)v_i + \frac{i}{\tau} s(1 + \sigma_i) = 0, \quad (36)$$

with

$$s = \sum_j v_j, \quad \sigma_i = \frac{1}{s} \sum_j (\alpha_{ij} - 1)v_j. \quad (37)$$

Expressing v_i from Eq. (36) and taking the sum over i , we obtain

$$1 + \frac{i}{\tau} \sum_j \frac{1 + \sigma_j}{\omega_j - \omega} = 0. \quad (38)$$

The equation for σ_i follows from substituting of v_j , found from Eq. (36), into the definition of σ_i , Eq. (37),

$$\sigma_i + \frac{i}{\tau} \sum_j \frac{\alpha_{ij} - 1}{\omega_j - \omega} (1 + \sigma_j) = 0. \quad (39)$$

The solutions of Eqs. (38) and (39) determine the complex frequencies of the eigenmodes, $\tilde{\omega}_k \equiv \tilde{\omega}'_k + i\tilde{\omega}''_k$.

A. “Point” sample

Let us first analyze the effect of disorder on a system with all $\alpha_{ij} = 1$, corresponding to the limit of a “point” sample, i.e., $(L/\lambda_0)^2 \ll 1$. With $\sigma_i = 0$, the real and imaginary parts of Eq. (38) read

$$\frac{1}{\tau} \sum_j \frac{\omega_j - \omega'}{(\omega_j - \omega')^2 + \omega''^2} = 0, \quad (40)$$

$$\frac{1}{\tau} \sum_j \frac{\omega''}{(\omega_j - \omega')^2 + \omega''^2} = 1. \quad (41)$$

This system of equations has two different solutions with a crossover between them governed by the parameter $\Omega\tau/N$. For large disorder $\Omega \gg N/\tau$, it can be readily seen that only one term in the sum in each of Eqs. (40) and (41) contributes. In this case, the solutions are simply $\omega = \omega_j + i/\tau$, as if the oscillators were uncoupled. In fact, this conclusion could be anticipated. The above parameter represents the ratio of the mean frequency spacing (MFS) of oscillators, Ω/N , and the inverse lifetime of an individual oscillator, $1/\tau$; when the former is much larger than the latter, $\Omega/N \gg 1/\tau$, the oscillators do not “feel” each other.

In the opposite case of large N (or weak disorder), $N \gg \Omega\tau$, the analysis of Eqs. (40) and (41) is carried out as follows. First note that in Eq. (40), which determines the real parts of the eigenfrequencies, $\tilde{\omega}'_k$, all the terms in the sum contribute now. Let us drop ω''^2 in the denominator of Eq. (40) (this step will be justified below). Then we find that the solutions $\tilde{\omega}'_k$ are given by the extrema of the polynomial $P(\omega) = \prod_j (\omega_j - \omega)$. These determine the frequencies of the $N-1$ subradiant modes. At the same time, in Eq. (41), which determines the imaginary parts of the eigenfrequencies, $\tilde{\omega}''_k$, all the terms in the sum are positive, so that one should keep only the term with ω_j closest to $\tilde{\omega}'_k$. Since $\tilde{\omega}'_k - \omega_j \sim \Omega/N$ for this term, we obtain the following estimate for the width of the subradiant mode: $\tilde{\omega}''_k \approx \gamma$, where

$$\gamma \sim \tau \Omega^2 / N^2. \quad (42)$$

It can be seen that γ is much smaller than the MFS (by the factor $\Omega\tau/N \ll 1$). This justifies neglecting ω''^2 in the denominators of Eqs. (40) and (41).

The superradiant solution of Eqs. (40) and (41) corresponds to the case $\omega'' \gg \Omega$. Then we readily obtain $\tilde{\omega}' = N^{-1} \sum_j \omega_j$ and $\tilde{\omega}'' = \Gamma \sim N/\tau$. We see that, indeed, $\Gamma/\Omega \sim N/\Omega\tau \gg 1$. Therefore, *the superradiant band in the spectral intensity is not affected by the disorder.*

We therefore conclude that cooperative emission is *not* destroyed by disorder. The spectrum of the system consists of a single superradiant and $N-1$ subradiant eigenmodes. For large $N/\Omega\tau$, the subradiant modes are well defined, since their spectral widths are much smaller than the MFS.

B. Limit of weak disorder

Let us address a nontrivial question about the fate of cooperative eigenmodes when the disorder in frequencies vanishes. In this limit, $\Omega\tau/N \rightarrow 0$, all oscillator frequencies become equal, i.e., $\omega_i \rightarrow \omega_0$. In the absence of cooperative coupling, $\alpha_{ij} = 0$ ($i \neq j$), the eigenfrequencies of the system are those of individual oscillators with the energy width much larger than the MFS, $1/\tau \gg \Omega/N$, so that the spectrum of the system is degenerate.

However, the situation is more complicated in the presence of cooperative coupling, $\alpha_{ij} \neq 0$. Consider the case of a “point sample,” $\alpha_{ij} = 1$. In this case, the width of subradiant modes is given by Eq. (42). Although the MFS diminishes with decreasing Ω , the width γ decreases even faster: $\gamma/(\Omega/N) \sim \Omega\tau/N \rightarrow 0$. In other words, in the presence of even a very weak disorder, the narrow subradiant peaks do *not* overlap. Therefore, the cooperative modes remain *distinct* even though the “bare” oscillator modes were already degenerate. In the case of general coupling, the width γ of subradiant modes for small values of $\Omega\tau/N$ will be determined by the *fluctuations* of α_{ij} , as we will see below.

C. Fluctuations of α_{ij}

Let us turn to the case with realistic coupling α_{ij} . The eigenfrequencies $\tilde{\omega}_k$ should now be determined from Eq. (35), which in component form reads

$$\frac{1}{\tau} \sum_j \frac{(\omega_j - \omega')(1 + \sigma'_j) - \omega'' \sigma''_j}{(\omega_j - \omega')^2 + \omega''^2} = 0, \quad (43)$$

$$\frac{1}{\tau} \sum_j \frac{\omega''(1 + \sigma'_j) + (\omega_j - \omega') \sigma''_j}{(\omega_j - \omega')^2 + \omega''^2} = 1, \quad (44)$$

with $\sigma_i(\omega) = \sigma'_i(\omega) + i\sigma''_i(\omega)$ satisfying Eq. (39), or in component form

$$\sigma''_i + \frac{1}{\tau} \sum_j (\alpha_{ij} - 1) \frac{(\omega_j - \omega')(1 + \sigma'_j) - \omega'' \sigma''_j}{(\omega_j - \omega')^2 + \omega''^2} = 0, \quad (45)$$

$$\sigma'_i - \frac{1}{\tau} \sum_j (\alpha_{ij} - 1) \frac{\omega''(1 + \sigma'_j) + (\omega_j - \omega') \sigma''_j}{(\omega_j - \omega')^2 + \omega''^2} = 0. \quad (46)$$

For $\omega'' \ll \Omega/N$, the system (43)–(46) can be approximately solved in the same way as for a “point” sample. The corresponding condition will be derived in Sec. V.

When evaluating the contribution to the lhs of Eq. (44) coming from the first term in the numerator, one should keep only one term in the sum with ω_j closest to $\tilde{\omega}'_k$: $(\omega_j - \tilde{\omega}'_k) \sim \Omega/N$. Then we obtain

$$\tilde{\omega}''_k \sim \frac{\tau \Omega^2}{N^2} \left(1 - \frac{1}{\tau} \sum_j \frac{\sigma''_j}{\omega_j - \tilde{\omega}'_k} \right), \quad (47)$$

where we again dropped ω''^2 in the denominator. Since $\sigma'_i \ll \sigma''_i \ll 1$ (see Sec. V), the frequencies $\tilde{\omega}'_k$ in Eq. (47) are the

same as those for the case $\alpha_{ij}=1$. Finding σ_i'' in the first order from Eq. (46), and substituting the result into Eq. (47), we obtain

$$\tilde{\omega}_k'' \sim \frac{\tau\Omega^2}{N^2} \left[1 + \frac{1}{\tau^2} \sum_{ij} \frac{\alpha_{ij}-1}{(\omega_i-\tilde{\omega}_k')(\omega_j-\tilde{\omega}_k')} \right]. \quad (48)$$

The second term is the desired correction to the width of the subradiant modes. Remarkably, this term turns to *zero* if the matrix elements α_{ij} are replaced by their average $\bar{\alpha}$. Indeed, in this case the double sum in Eq. (48) would factorize into a product of two sums, each vanishing due to the fact that $\tilde{\omega}_k'$ are the solutions of Eq. (40) (corresponding to $\alpha_{ij}=1$). Therefore, the widths of the subradiant modes are determined by the *fluctuations*, $\delta\alpha_{ij}$, of the coupling parameters α_{ij} rather than the deviation of their average, $\bar{\alpha}$, from unity. It should be noted that this property is general: one can easily see by comparing Eqs. (43) and (44) to Eqs. (46) and (46) that for $\alpha_{ij}=\text{const}$, we have $\sigma_i''=0$ and $\sigma_i' \ll 1$, so that the eigenfrequencies $\tilde{\omega}_k$ are unaffected.

D. Discussion of the numerical results

We are now in the position to explain the spectra shown in Figs. 3–6. For the model coupling $\alpha_{ij}=\alpha$, $\alpha_{ii}=1$, fluctuations only in the diagonal elements are finite: $\delta\alpha_{ij}=(1-\alpha)\delta_{ij}$. Substituting this $\delta\alpha_{ij}$ into Eq. (48) [instead of $(\alpha_{ij}-1)$] and keeping only the term with $(\tilde{\omega}_k'-\omega_j) \sim \Omega/N$ in the remaining sum, we obtain

$$\gamma \sim \left[\frac{\tau\Omega^2}{N^2} + \frac{1}{\tau}(1-\alpha) \right]. \quad (49)$$

The above expression indicates that after the cooperative modes have been formed (at $N \sim \Omega\tau$), the system can be found in two different regimes characterized by the relative magnitude of the first and second terms in the rhs. For an intermediate number of oscillators, $\Omega\tau \lesssim N \lesssim \Omega\tau(1-\alpha)^{-1/2}$, the width decreases with increasing N , as can be seen by comparing the bottom second and third curves in each of Figs. 3–6 (note that the lowest curves with $N=2$ show no sign of cooperative emission). In this regime, the system behaves in the same way as a ‘‘point’’ sample. With increasing number of oscillators, the dependence on N saturates, and the width is dominated by the fluctuations of α_{ij} . Correspondingly, the change in the pattern of the peaks in Figs. 3–6, calculated for different values of $1-\alpha$, occurs at different N , as can be seen by comparing the next two curves in each figure. Note, however, that with further increase in N , the curves exhibit yet another change in pattern. Namely, the peaks get smeared out (the top two curves in each figure). This occurs when the value of $(1-\alpha)/\tau$ exceeds the MFS, Ω/N , which is inconsistent with the above analysis. The reason for such a discrepancy is that for large N , the model with coupling α_{ij} independent of separation between oscillators becomes inadequate, as we mentioned above. For the correct description of the peaks smearing at large N , the spatial dependence of α_{ij} is crucial; this question is addressed in Sec.

VI. Nevertheless, for $N \lesssim \Omega\tau/(1-\alpha)$, this model describes accurately the mesoscopic features of the spectral intensity, as shown in the next section.

V. STRONG MESOSCOPIC REGIME

Let us now estimate the typical width of the radiation eigenmodes due to the fluctuations in α_{ij} . Since the configurational average of the second term in Eq. (48) (with $\delta\alpha_{ij}$ instead of $\alpha_{ij}-1$) vanishes, we need to evaluate $\langle (\tilde{\omega}_k'')^2 \rangle$. Using the fact that only diagonal terms in the average $\langle \delta\alpha_{ij}\delta\alpha_{i'j'} \rangle$ survive and omitting the first term in Eq. (48), we write

$$\langle (\tilde{\omega}_k'')^2 \rangle \sim \left(\frac{\Omega^2}{\tau N^2} \right)^2 \left\langle \sum_{ij} \frac{(\delta\alpha_{ij})^2}{(\omega_i-\tilde{\omega}_k')^2(\omega_j-\tilde{\omega}_k')^2} \right\rangle. \quad (50)$$

The sum is dominated by the terms with $(\omega_i-\tilde{\omega}_k') \sim (\omega_j-\tilde{\omega}_k') \sim \Omega/N$. Since the typical spatial separation between two oscillators with close frequencies is $\sim L$, the separation fluctuations are of the same order. Thus, the typical fluctuation of α_{ij} is $\delta\alpha \equiv \sqrt{\langle (\delta\alpha_{ij})^2 \rangle} \sim (L/\lambda_0)^2$, and we finally obtain the typical width of a subradiant mode, $\gamma \equiv \sqrt{\langle (\tilde{\omega}_k'')^2 \rangle}$, as

$$\gamma \sim \frac{\delta\alpha}{\tau} \sim \frac{1}{\tau} \left(\frac{L}{\lambda_0} \right)^2. \quad (51)$$

Comparing Eq. (51) to Eq. (48), we see that fluctuations in α_{ij} dominate the width γ for $N \gtrsim \Omega\tau(\lambda_0/L)$.

In order to characterize the fine structure in the emission spectrum, it is convenient to introduce the dimensionless parameter

$$\kappa = \frac{\Omega\tau}{N} \left(\frac{\lambda_0}{L} \right)^2. \quad (52)$$

It represents the product of a small, $\Omega\tau/N$, and a large, $(\lambda_0/L)^2$, factor, which characterize the disorder and the system size, respectively. In terms of κ , the condition for the formation of the cooperative modes, $N/\Omega\tau \gg 1$, can be presented as $\kappa \ll (\lambda_0/L)^2$.

Using Eq. (52), the width (51) can be expressed in terms of the MFS as

$$\gamma \sim \frac{1}{\kappa} \left(\frac{\Omega}{N} \right). \quad (53)$$

This result applies when $\kappa \lesssim \lambda_0/L$. On the other hand, it was implicit in the above derivation (Sec. IV) that typical σ_i' and σ_i'' are smaller than unity. The latter parameters can be estimated in a similar way from Eqs. (45) and (46) with the result: $\sigma'' \sim \kappa^{-1}$ and $\sigma' \sim \sigma''^2 \sim \kappa^{-2}$. Thus, the lower boundary for κ , for which Eq. (53) applies, is $\kappa \gtrsim 1$. For $\kappa \sim 1$, all terms in Eqs. (43)–(46) become of the same order of magnitude, and for smaller κ this system has no *subradiant* solutions, as discussed above.

Since the MFS exceeds the width γ within the entire domain $1 \lesssim \kappa \lesssim \lambda_0/L$, the fine structure in the spectral intensity $I(\omega)$ is well pronounced. In other words, this domain corresponds to the strong mesoscopic regime. The opposite case $\kappa \lesssim 1$ is considered in the next section.

VI. WEAK MESOSCOPIC REGIME

In the domain $\kappa \ll 1$, the system cannot sustain eigenmodes that involve all N oscillators. As a result, the eigenmodes become localized, in the sense that each eigenmode would comprise some $N_c \ll N$ oscillators and occupy the volume with characteristic size $L_c \ll L$. The magnitude of L_c and N_c can be estimated from the following argument. Let us divide the system of oscillators into subsystems of increasingly smaller size. When the size of the subsystem becomes $\sim L_c$, the system of equations (43) and (44), applied to a subsystem, first acquires a solution. This happens when the width $\gamma_c \sim \tau^{-1}(L_c/\lambda_0)^2$, determined from Eq. (51) for a subsystem, becomes of the order of MFS within a subsystem, i.e.,

$$\frac{1}{\tau} \left(\frac{L_c}{\lambda_0} \right)^2 \sim \frac{\Omega}{N_c}. \quad (54)$$

Taking into account that $N_c = N(L_c/L)^3$, we find

$$L_c \sim \kappa^{1/5} L, \quad N_c \sim \kappa^{3/5} N. \quad (55)$$

Substituting these results back into Eq. (54), we find for the eigenmodes width

$$\gamma = \gamma_c \sim \frac{1}{\kappa^{3/5}} \left(\frac{\Omega}{N} \right) \gg \frac{\Omega}{N}. \quad (56)$$

From Eq. (55), we can also estimate how the relative amplitude of mesoscopic fluctuations in the spectral intensity $I(\omega)$ falls off with decreasing κ :

$$\frac{\delta I}{I} \sim \left(\frac{N_c}{N} \right)^{1/2} = \kappa^{3/10}. \quad (57)$$

Thus, smearing of the fine structure in the cooperative emission spectrum with decreasing κ occurs rather slowly.

VII. DIPOLE-DIPOLE INTERACTIONS

In this section we study the effect of dipole-dipole interactions on the cooperative emission from a disordered system. Note that for a ‘‘point’’ sample with $L \ll \lambda_0$, the typical magnitude of the dipole-dipole interaction between two oscillators is much larger than their superradiant coupling, $\beta_{ij}/\alpha_{ij} \sim (\lambda_0/L)^3 \gg 1$. The structure of the eigenmodes in the absence of superradiant coupling, given by Eq. (19) with $\alpha_{ij} = 0$, was considered in several papers.^{22–27} Renormalization-group arguments of Ref. 22 (see also Ref. 24) suggest that all eigenmodes are delocalized. Numerical studies^{25–27} indicate a wide range of spatial scales in eigenmodes and thus seem to support this conclusion. In Ref. 23, the role of general random-matrix perturbation in the spectrum of multilevel system was studied analytically; the ensemble-averaged renormalization of the spectrum of the system was derived, which does not capture, however, the mesoscopic effects. Below we argue that finite disorder in combination with superradiant coupling leads to a certain ‘‘resistance’’ of the system to large, but zero on average, dipole-dipole terms because of the formation of cooperative modes.

In the absence of superradiant coupling ($\alpha_{ij} = 0$) the dipole-dipole interactions lead to shifts in the frequencies of individual oscillators. The resulting additional spread in ω_i is, in general, much larger than the ‘‘bare’’ spread Ω . This can be readily seen from the lowest-order correction to the frequency, $\delta\omega_i$, which has the form

$$\delta\omega_i = \frac{1}{\tau^2} \sum_{j \neq i} \frac{\beta_{ij}^2}{\omega_i - \omega_j} \quad (58)$$

(since $\beta_{ii} = 0$, the lowest-order correction to ω_i is quadratic). The main contribution to the sum comes from pairs of oscillators located closely in space, with $r_{ij} \sim LN^{-1/3}$ (nearest-neighbor interaction), so that

$$\beta_{ij} \sim N \left(\frac{\lambda_0}{L} \right)^3. \quad (59)$$

Since the typical frequency difference for such pairs is $\sim \Omega$, we obtain

$$\delta\omega_i \sim \frac{N^2}{\Omega \tau^2} \left(\frac{\lambda_0}{L} \right)^6. \quad (60)$$

On the other hand, the results obtained in the previous sections apply only if the additional disorder, caused by dipole-dipole interactions, does not affect the MFS. This requires the condition $\delta\omega_i \ll \Omega$ to be met. Using Eq. (60), this condition could be rewritten as $\lambda_0/L \ll (\Omega \tau/N)^{1/3}$. Since the formation of cooperative modes occurs only if $\Omega \tau/N \ll 1$, one could draw the conclusion that neglecting the dipole-dipole interactions would be inconsistent with our basic assumption $L \ll \lambda_0$.

The resolution of this apparent contradiction lies in the observation that, in the presence of superradiant coupling, i.e., $\alpha_{ij} \neq 0$, the true eigenmodes of the system are cooperative modes comprised of a large number of oscillators. Therefore, the relevant condition should involve the shifts, $\delta\tilde{\omega}'_k$, of the eigenmodes frequencies, rather than $\delta\omega_i$. In the first order, $\delta\tilde{\omega}'_k$ is given by an expression similar to the second term in the rhs of Eq. (48) (with β_{ij} instead of $\alpha_{ij} - 1$). Since this term vanishes on average, as discussed above, the typical shift, $\delta\tilde{\omega}' \equiv \sqrt{\langle (\delta\tilde{\omega}'_k)^2 \rangle}$, can be estimated from [compare with Eq. (50)]

$$\langle (\delta\tilde{\omega}'_k)^2 \rangle \sim \left(\frac{\Omega^2}{\tau N^2} \right)^2 \left\langle \sum_{ij} \frac{(\beta_{ij})^2}{(\omega_i - \tilde{\omega}'_k)^2 (\omega_j - \tilde{\omega}'_k)^2} \right\rangle. \quad (61)$$

There are two main contributions to the sum in the rhs. The first comes from the nearest-neighbor interaction with β_{ij} given by Eq. (59). The second contribution originates from the pairs (ij) that are close in frequency; for such pairs, $\beta_{ij} \sim (\lambda_0/L)^3$. Both contributions turn out to be of the same order of magnitude, resulting in

$$\delta\tilde{\omega}' \sim \frac{1}{\tau} \left(\frac{\lambda_0}{L} \right)^3. \quad (62)$$

This result is smaller than $\delta\omega_i$ in Eq. (60) by the factor $N(N/\Omega \tau)(\lambda_0/L)^3 \gg 1$. Such a dramatic difference illustrates the ‘‘resistance’’ of a coupled system of oscillators with dis-

order in frequencies to dipole-dipole interactions, as mentioned above. This property can also be qualitatively explained as follows. The dipole-dipole interaction between two subradiant modes can be viewed as an interaction between a mode and the electric field $\vec{E}(\mathbf{r})$ created by the dipole moments of oscillators making up the other mode. Since the number of oscillators in a mode is large, their electric fields effectively cancel each other, so that the resulting net field $\vec{E}(\mathbf{r})$ varies in space *much slower* than those of the individual oscillators. Note now that a slowly varying electric field couples only weakly to a *subradiant* mode. In fact, the suppression of the dipole-dipole interaction between subradiant modes has the same physical origin as their decoupling from the radiation field: had the electric field \vec{E} been uniform, the cooperative modes would not interact at all with each other. This is the reason why the corrections to $\tilde{\omega}'_k$ and $\tilde{\omega}''_k$ vanish *on average*, and consequently the typical $\delta\tilde{\omega}'$ and γ are determined by the *fluctuations* of S_{ij} . In contrast, the frequency shifts of individual oscillators are due to their interactions with the nearest neighbors, so that no cancellations occur.

Thus we arrive at the condition $\lambda_0/L \ll (\Omega\tau)^{1/3}$, or, in terms of the parameter κ ,

$$\kappa \gg \frac{1}{N} \left(\frac{\lambda_0}{L} \right)^5. \quad (63)$$

This condition should be consistent with the condition for the formation of the cooperative modes, $\kappa \ll (\lambda_0/L)^2$. We see that both conditions are satisfied for sufficiently large N , i.e., $N \gg (\lambda_0/L)^3$. To account for different mesoscopic regimes, it is convenient to present Eq. (63) in the form

$$N \gg \left(\frac{\lambda_0}{L} \right)^n. \quad (64)$$

Then $n=3, 4$, and 5 correspond to the ‘‘point’’ sample, strong mesoscopic, and weak mesoscopic regimes, respectively.

VIII. CONCLUSIONS

The main result of the present paper is that disorder in oscillator frequencies does not destroy the cooperative character of the emission from a ‘‘point’’ sample, as long as the MFS, Ω/N , is smaller than the linewidth of an individual oscillator, τ^{-1} . In the opposite case, when $\Omega/N \gg \tau^{-1}$, the spectrum represents a system of nonoverlapping Lorentzians with width τ^{-1} .

It is convenient to characterize the disorder in terms of the

dimensionless parameter $\kappa = \Omega\tau\lambda_0^2/NL^2$. Below we summarize our results for the characteristic width γ of the subradiant peaks (in units of Ω/N) for different domains of κ :

$$\gamma = \frac{\Omega}{N} \Phi(\kappa, L/\lambda_0), \quad (65)$$

where the dimensionless function Φ has the following asymptotes:

$$\Phi = \kappa \left(\frac{L}{\lambda_0} \right)^2 \quad \text{for} \quad \frac{\lambda_0}{L} \leq \kappa \leq \left(\frac{\lambda_0}{L} \right)^2, \quad \text{‘‘point’’ sample,}$$

$$\Phi = \kappa^{-1}, \quad \text{for} \quad 1 \leq \kappa \leq \frac{\lambda_0}{L}, \quad \text{strong mesoscopic regime,}$$

$$\Phi = \kappa^{-3/5}, \quad \text{for} \quad \kappa \leq 1, \quad \text{weak mesoscopic regime.} \quad (66)$$

For $\kappa \geq (\lambda_0/L)^2$, the spectrum corresponds to uncoupled oscillators.

In Sec. III, we presented an exact solution of a model with simplified (separable) coupling, Eq. (20). This model describes accurately the first two (‘‘point’’ sample and strong mesoscopic) regimes in Eq. (66). It becomes, however, inadequate in the third (weak mesoscopic) regime, giving κ^{-1} instead of the correct $\kappa^{-3/5}$ dependence for the period of mesoscopic structure in the cooperative emission spectrum.

Throughout the paper we have considered a three-dimensional system of oscillators. When the oscillators are confined to a plane, only the results for $\kappa \leq 1$ should be modified. In this case, repeating the consideration of Sec. VI, we obtain $\Phi = \kappa^{-1/2}$. Also for the relative magnitude of mesoscopic fluctuations $\delta I/I$ instead of Eq. 57 we obtain $\delta I/I \sim \kappa^{-1/4}$.

Note finally that in experiments, such as photoexcited excitons in polymer films, the number of oscillators N is governed by the excitation intensity.¹² Thus, for a given disorder, the crossover from the strong mesoscopic regime ($\kappa > 1$) to the weak mesoscopic regime ($\kappa < 1$) can be simply achieved by increasing the excitation intensity level.²⁸

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