Cooperative emission from a disordered system: A classical model

T. V. Shahbazyan,[a] M. E. Raikh,[b] and Z. V. Vardeny[b]

[a]Department of Physics, Ohio University, Athens, OH 45701 USA

[b]Department of Physics, University of Utah, Salt Lake City, Utah 84112 USA

ABSTRACT

The effect of disorder on the collective emission from a system of classical oscillators is studied theoretically. Three types of disorder are considered: random orientation of dipole moments, finite spread in frequencies of the individual oscillators (diagonal disorder), and dipole-dipole interaction (off-diagonal disorder). We found that for sufficiently high concentration of oscillators the diagonal disorder does not completely destroy the collective character of the emission. We show that the eigenmodes of a cooperative system with disorder comprise a large number of oscillators and, due to cooperative character of the emission, their lifetime is much longer than the lifetime of an individual oscillator. Consequently, the cooperative emission spectrum of the system is not simply broadened by the disorder, but represents a superposition of relatively narrow peaks.

Keywords: disorder, cooperative emission, oscillator, dipole-dipole interaction

1. INTRODUCTION

Recent advances in the field of luminescent conducting polymers have demonstrated that these materials show not only strong photoluminescence (PL) in the visible spectral range, but also high optical gain.[1–7] The latter property, in principle, should eventually lead to "plastic lasers" in the visible spectral range, made out of conducting polymers as the active layers. The large optical gain in conducting polymers is based on the large excitonic optical cross-section, which is of order[5] 10^{-15}cm^2. This large optical emission cross-section opens up the possibility that the photoexcited excitons, if close enough, would interact among themselves in thin polymer films via their emissive electric field, and may lead to the phenomenon of the cooperative emission, or superfluorescence (SE).[4–6] Indeed stimulated emission has been observed in various thin films of conducting polymers.[1–7] It has been found that SE has the following characteristic properties: drastic emission spectrum narrowing for excitation intensities, I_0, higher than a characteristic threshold excitation intensity I_{th}, quadratic dependence of the emission intensity, I, on I_0 for $I_0 \sim I_{th}$, and a drastic radiative lifeime shortening for $I_0 > I_{th}$. This phenomena have been independently attributed to mirrorless lasing,[4] amplified spontaneous emission,[3] exciton condensation[7] and cooperative emission.[5,6]

On the other hand, it is well known that conducting polymers are strongly disordered.[8] They contain impurities and defects which break the polymer chains into segments of relatively short conjugation length, with the distribution depending on the film quality.[9] This has a direct effect on the exciton energy, $\hbar\omega$, since it has been found that $\hbar\omega$ depends on the chain conjugation length.[10] Then the question of whether this and other disorder will simply destroy the anticipated phenomenon of cooperative emission in conducting polymer films is in order. This question is addressed in the present paper.

To study the effect of disorder we adopt the simplest classical description of cooperative emission. This description captures the basic features of the phenomenon, such as drastic shortening of the radiation time, but does not account for the initial stage of the process (the so called delay time statistics).

2. THEORY

We consider a system of N oscillators located at random points $\mathbf{R}_n$, with frequencies ω_n distributed within an interval $(\omega_0 - \Omega, \omega_0 + \Omega)$. Each oscillator is driven by the radiation field $\mathbf{E}(\mathbf{R},t)$ produced by the same oscillator and by all other oscillators. The equation of motion for the displacement r_i of a given oscillator i, reads

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

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 motion of oscillators, can be written as

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

The current $\mathbf{J}$ plays the role of a source, which generates the field $\mathbf{E}(\mathbf{R},t)$. This field generation is described by the equation

$$\Delta_{\mathbf{R}}\mathbf{E} - \frac{1}{c^2}\ddot{\mathbf{E}} = \frac{4\pi}{c^2}\dot{\mathbf{J}}, \tag{3}$$

where c is the velocity of light.

Within the classical approach the radiation spectrum of the system should be calculated as follows. We assume that at the initial moment ($t = 0$) all oscillators are excited and have the same energy of oscillations, but have different phases ϕ_i. The radiation field at the initial moment is zero: $\mathbf{E}(\mathbf{R},0) = 0$. The evolution of $\mathbf{E}$ with time can be obtained by solving Eqs. (1–3). Taking the limit $R \to \infty$ and expanding the field into harmonics, we obtain the spectral intensity $I(\omega) = |\mathbf{E}(\infty,\omega)|^2$.

To carry out this program it is convenient to employ the Laplace transformation. Then the system of equations for the Laplace–transformed functions $\bar{r}_i(p)$ and $\bar{\mathbf{E}}(\mathbf{R},p)$ takes the form

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

$$\Delta_{\mathbf{R}}\bar{\mathbf{E}}(\mathbf{R},p) - \frac{p^2}{c^2}\bar{\mathbf{E}}(\mathbf{R},p) = \frac{4\pi e}{c^2}\sum_i \mathbf{n}_i \left[p^2\bar{r}_i(p) - a(p\cos\phi_i - \omega_i\sin\phi_i)\right]\delta(\mathbf{R}-\mathbf{R}_i), \tag{5}$$

where $a\sin\phi_i$ and $\omega_i a\cos\phi_i$ are the displacement and the velocity of the oscillator i at $t = 0$. The solution of Eq.(5) for $\bar{\mathbf{E}}(\mathbf{R},p)$ can be presented as a linear combination of eigenmodes $\mathbf{E}_\nu(\mathbf{R})$ of electromagnetic field which satisfy the wave equation

$$\Delta_{\mathbf{R}}\mathbf{E}_\nu(\mathbf{R}) + \frac{\omega_\nu^2}{c^2}\mathbf{E}_\nu(\mathbf{R}) = 0, \tag{6}$$

with ω_ν being the eigenfrequency of the mode ν. Assuming that the modes are normalized ($\int d\mathbf{R}\mathbf{E}_\nu^2(\mathbf{R}) = 1$), we obtain the following expression for $\bar{\mathbf{E}}(\mathbf{R},p)$

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

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

$$(\omega_i^2+p^2)\bar{r}_i(p) - a(p\cos\phi_i - \omega_i\sin\phi_i) = -\frac{4\pi e^2}{m}\sum_{j,\nu}\left[p^2\bar{r}_j(p) - a(p\cos\phi_j - \omega_j\sin\phi_j)\right]\frac{[\mathbf{n}_j\cdot\mathbf{E}_\nu(\mathbf{R}_j)]\,[\mathbf{n}_i\cdot\mathbf{E}_\nu(\mathbf{R}_i)]}{\omega_\nu^2+p^2}. \tag{8}$$

It is convenient to rewrite Eq. (8) introducing the new variables

$$v_i(p) = \frac{p^2\bar{r}_i(p)}{a} - (p\cos\phi_i - \omega_i\sin\phi_i). \tag{9}$$

Then Eq. (8) takes the form

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

where the constants S_{ij}, which describe the coupling between oscillators i and j via the radiation field, are defined as

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

Before analyzing Eq. (10) let us express the intensity, $I(\omega)$, through the values $v_i(p)$. The expression for $\bar{\mathbf{E}}(\mathbf{R}, p)$ follows from Eqs. (7) and (9)

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

The Fourier transform of the electric field is found by replacing p by $i\omega$ in Eq. (12). In the limit $R \to \infty$ only the pole $\omega_\nu = \omega$ contributes to the sum over ν and we have

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

The terms $\propto [\mathbf{E}_\nu(\mathbf{R}) \cdot \mathbf{E}_\mu(\mathbf{R})]$, 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 $[\mathbf{n}_j \cdot \mathbf{E}_\nu(\mathbf{R}_j)]\,[\mathbf{n}_i \cdot \mathbf{E}_\nu(\mathbf{R}_i)]$. Note that the same products enter into the coupling coefficients S_{ij} (Eq. (11)). This allows us to present the final expression for the spectral intensity in a much more compact form

$$I(\omega) \propto \sum_{ij} v_i(i\omega) v_j^*(i\omega) \mathrm{Im} S_{ij}. \tag{14}$$

We assume that the spread, 2Ω, of the oscillators frequencies due to the disorder is much smaller than the central frequency ω_0 ($\Omega \ll \omega_0$). This means that the frequency dependence of the coupling constants is weak, so that S_{ij} can be evaluated at $p = i\omega_0$. The real part of S_{ii}, which comes from the principle value of the sum over modes in Eq. (11), diverges for $i = j$. This divergency is the manifestation of the Lamb shift, which is well-known in quantum electrodynamics. The diverging term can be absorbed into ω_i^2. The imaginary part of S_{ii} in Eq. (11) results from the pole $\omega_\nu = \omega_0$ and is finite. We denote $\mathrm{Im} S_{ii}(i\omega_0)$ as $2\omega_0/\tau_0$, where τ_0 is the radiative lifetime of the oscillator. In the case of one oscillator in the vacuum, the modes $\mathbf{E}_\nu$ are plane waves, and the summation over modes recovers the textbook result

$$\tau_0 = \frac{3mc^3}{e^2\omega_0^2}. \tag{15}$$

We note, however, that the expression for τ_0 changes if an oscillator resides inside a thin film. In this case the modes $\mathbf{E}_\nu$ are the waveguided modes (both propagating and leaking).

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 a typical wavelength of the radiation in vacuum. For $R_{ij} \gg \lambda_0$ both real and imaginary parts of S_{ij} oscillate with R_{ij}, and the effect of coupling for a large ensemble of oscillators is negligibly small. For $R_{ij} \ll \lambda_0$ the real part of S_{ij}, $\mathrm{Re} S_{ij}$, represents the dipole–dipole interaction of the two oscillators. It is more convenient to rewrite $\mathrm{Re} S_{ij}$ in the form

$$\mathrm{Re} S_{ij} = \frac{2\omega_0}{\tau_0} \beta_{ij}, \tag{16}$$

where β_{ij} is a dimensionless constant defined as

$$\beta_{ij} = \left(\frac{\lambda_0}{2\pi R_{ij}}\right)^3 \left[(\mathbf{n}_i \cdot \mathbf{n}_j) - \frac{3(\mathbf{n}_i \cdot \mathbf{R}_{ij})(\mathbf{n}_j \cdot \mathbf{R}_{ij})}{R_{ij}^2}\right]. \tag{17}$$

In a similar way we may introduce the constant α_{ij} to parametrize the imaginary part, $\mathrm{Im} S_{ij}$, of S_{ij}.

$$ImS_{ij} = \frac{2\omega_0}{\tau_0}\alpha_{ij}, \quad \alpha_{ij} = \mathbf{n}_i \cdot \mathbf{n}_j - \frac{1}{10}\left(\frac{2\pi R_{ij}}{\lambda}\right)^2 \left[2(\mathbf{n}_i \cdot \mathbf{n}_j) - \frac{(\mathbf{n}_i \cdot \mathbf{R}_{ij})(\mathbf{n}_j \cdot \mathbf{R}_{ij})}{R_{ij}^2}\right]. \tag{18}$$

Because the distribution of oscillator frequencies is relatively narrow ($\Omega \ll \omega_0$) we can make some simplifications in Eq. (10). Namely, for $p = i\omega$, the factor $(\omega_i^2 + p^2)$ in the left–hand side can be replaced by $2\omega_0(\omega_i - \omega)$, and the right–hand side can be written as $-i\omega_0 e^{-i\phi_i}$. Then Eq. (10) takes the form

$$v_i(\omega_i - \omega) + \frac{1}{\tau_0}\sum_j (i\alpha_{ij} + \beta_{ij})v_j = -\frac{i}{2}e^{-i\phi_i}. \tag{19}$$

Eq. (19) together with Eq. (14) allows us to calculate the spectral intensity $I(\omega)$ for an arbitrary set, $\{\phi_i\}$, of initial phases of oscillators. In the experiment, the measured spectrum $I(\omega)$ represents the result of averaging over many excitation pulses. 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 these phases.

We start our discussion with the two simplest cases of a single oscillator ($N = 1$) and a pair of oscillators ($N = 2$).

i) $N = 1$.

The system of Eqs. (19) reduces in this case to a single equation

$$v_1\left(\omega_1 - \omega + \frac{i}{\tau_0}\right) = -\frac{i}{2}e^{-i\phi_1}. \tag{20}$$

Substituting this solution into (14) we get a standard Lorentzian emisssion spectrum

$$I(\omega) \propto \frac{1}{1 + (\omega_1 - \omega)^2\tau_0^2}, \tag{21}$$

with full width at half maximum (FWHM) $\Gamma = 2/\tau_0$.

ii) $N = 2$

Without loss of generality it can be assumed that the frequencies of the two oscillators in this case are symmetric with respect to ω_0, namely, $\omega_1 = \omega_0 + \delta\omega$, $\omega_2 = \omega_0 - \delta\omega$. Then the system of Eqs. (19) takes the form

$$\begin{aligned} v_1\left(\omega_0 - \omega + \delta\omega + \frac{i}{\tau_0}\right) + \frac{v_2}{\tau_0}(i\alpha_{12} + \beta_{12}) &= -\frac{i}{2}e^{-i\phi_1}, \\ v_2\left(\omega_0 - \omega - \delta\omega + \frac{i}{\tau_0}\right) + \frac{v_1}{\tau_0}(i\alpha_{12} + \beta_{12}) &= -\frac{i}{2}e^{-i\phi_2}. \end{aligned} \tag{22}$$

After a simple algebra one gets the following expression for the spectral intensity (Eq. (14))

$$I(\omega) \propto \frac{(\omega_0 - \omega)^2\tau_0^2 + \delta\omega^2\tau_0^2 - 2\alpha_{12}\beta_{12}(\omega_0 - \omega)\tau_0 + 1 - \alpha_{12}^2 + \beta_{12}^2}{\left[(\omega_0 - \omega)^2\tau_0^2 - \delta\omega^2\tau_0^2 - 1 - \beta_{12}^2 + \alpha_{12}^2\right]^2 + 4\left[(\omega_0 - \omega)\tau_0 - \alpha_{12}\beta_{12}\right]^2}. \tag{23}$$

Let us analyze the result obtained. As it was mentioned in the Introduction, there are three types of disorder that affect the shape of the spectrum in Eq. (23). Diagonal disorder is desribed by $\delta\omega$, off-diagonal disorder is described by β_{12}, and disorder in orientations of dipoles enters though the product $(\mathbf{n}_1\mathbf{n}_2)$ in α_{12}. We will now discuss the role of each type of disorder separately.

Assuming first that both diagonal and off-diagonal disorders are absent, i.e. $\delta\omega = 0$, $\beta_{12} = 0$, the spectrum Eq. (23) can be rewritten in the form

$$I(\omega) \propto \left[\frac{1-\alpha_{12}}{(\omega_0-\omega)^2\tau_0^2 + (1-\alpha_{12})^2} + \frac{1+\alpha_{12}}{(\omega_0-\omega)^2\tau_0^2 + (1+\alpha_{12})^2} \right]. \tag{24}$$

We see that the spectrum represents two Lorentzians with FWHM $\Gamma_{as} = 2(1-\alpha_{12})/\tau_0$ and $\Gamma_s = 2(1+\alpha_{12})/\tau_0$ respectively. Such a form is a clear signature of the cooperative emission. It shows that the true modes of a pair of oscillators, coupled through the radiation field, are a symmetric mode (with a short lifetime $\tau_s = \tau_0/(1+\alpha_{12})$) and an antisymmetric mode (with a long lifetime $\tau_{as} = \tau_0/(1-\alpha_{12})$). The difference $\Delta\tau$ between the lifetimes is large when α_{12} is close to 1. This is the case when the dipole moments are parallel. It is seen from Eq. (18) that for $\mathbf{n}_1 \parallel \mathbf{n}_2$ we have $(1-\alpha_{12}) \sim (R_{12}/\lambda_0)^2 \ll 1$. Eq. (24) then indicates that the orientational disorder suppresses the cooperative nature of the emission. As the angle between $\mathbf{n}_1$ and $\mathbf{n}_2$ increases, α_{12} becomes smaller and the difference $\Delta\tau$ between the symmetric and antisymmetric lifetimes vanishes.

Now let us assume that $\delta\omega \neq 0$, but off–diagonal disorder is still absent ($\beta_{12} = 0$). In this case $I(\omega)$ modifies to

$$I(\omega) \propto \frac{1-\sqrt{\alpha_{12}^2 - (\delta\omega\tau_0)^2}}{(\omega_0-\omega)^2\tau_0^2 + [1-\sqrt{\alpha_{12}^2-(\delta\omega\tau_0)^2}]^2} + \frac{1+\sqrt{\alpha_{12}^2 - (\delta\omega\tau_0)^2}}{(\omega_0-\omega)^2\tau_0^2 + [1+\sqrt{\alpha_{12}^2-(\delta\omega\tau_0)^2}]^2}. \tag{25}$$

For $\delta\omega\tau_0 \leq \alpha_{12}$ the spectrum in Eq. (25) is still the sum of two Lorentzians centered at $\omega = \omega_0$. Eq. (25) illustrates how the cooperative emission is suppressed by the diagonal disorder. Even in the case of an "ideal" coupling, $\alpha_{12} = 1$, finite $\delta\omega$ broadens the narrow Lorentzian and narrows the broad Lorentzian. At $\delta\omega\tau_0 = \alpha_{12}$ both Lorentzians have the same width.

Finally we assume that the coupling is ideal and only off–diagonal disorder is present ($\delta\omega = 0$, $\beta_{12} \neq 0$). From Eq. (23) we get

$$I(\omega) \propto \frac{1}{[(\omega_0-\omega)\tau_0 + \beta_{12}]^2 + 4}. \tag{26}$$

We see that the narrow Lorentzian has disappeared completly. Although the FWHM of the broad Lorentzian is still $4/\tau_0$, the frequency of the maximum is shifted due to the dipole–dipole interactions by β_{12}/τ_0.

Summarizing the discussion for the case of two oscillators, we conclude that all three types of disorder tend to suppress the cooperative emission.

We now turn to the case of a large number of oscillators ($N \gg 1$). We will adopt the same strategy as above and study separately the effect of each type of disorder on the emission spectrum.

i) Ideal case ($\mathbf{n}_i \parallel \mathbf{n}_j$, $\omega_i = \omega_0$, $\beta_{ij} = 0$ for all i, j)

As we mentioned above, when all dipole moments are parallel to each other the coefficients α_{ij} in Eq. (18) are close to 1. Strong simplification can be then achieved if we assume all α_{ij} to be equal; i.e. $\alpha_{ij} = \alpha$, where α is the average α_{ij} over the distances R_{ij} between the dipoles. Eq. (19) can be then rewritten as

$$v_i \left[\omega_0 - \omega + \frac{i}{\tau_0}(1-\alpha) \right] + \frac{i}{\tau_0}\alpha\sigma = -\frac{i}{2}e^{-i\phi_i}, \tag{27}$$

where

$$\sigma = \sum_{i=1}^{N} v_i. \tag{28}$$

It is straightforward to solve Eqs. (27). Summing up all equations, we get a closed expression for σ

$$\sigma = -\frac{i}{2\left[\omega_0 - \omega + \frac{i}{\tau_0}(1-\alpha+\alpha N)\right]} \sum_i e^{-i\phi_i}. \tag{29}$$

Using this expression we find all the v_i and then substitute them into Eq. (14) for the spectral intensity. After averaging over the phases we get

$$I(\omega) \propto \left[\frac{(1-\alpha)(N-1)}{(\omega_0-\omega)^2\tau_0^2 + (1-\alpha)^2} + \frac{1+\alpha(N-1)}{(\omega_0-\omega)^2\tau_0^2 + (1-\alpha+\alpha N)^2}\right]. \tag{30}$$

In accordance to the classical result,[11] we find that the FWHM of the broad Lorentzian is $\Gamma_s \approx 2N/\tau_0$, which corresponds to the short lifetime $\tau_s = \tau_0/N$. The FWHM of the narrow Lorentzian and its related lifetime remain, however, the same, $\Gamma_{as} = 2(1-\alpha)/\tau_0$, as for the the case of a pair of oscillators.

ii) Orientational disorder

In this case the coupling constants α_{ij} can be presented as $\alpha \mathbf{n}_i \cdot \mathbf{n}_j$. The system (19) takes the form

$$v_i\left[\omega_0 - \omega + \frac{i}{\tau_0}(1-\alpha)\right] + \frac{i}{\tau_0}\alpha \mathbf{n}_i \cdot \mathbf{s} = -\frac{i}{2}e^{-i\phi_i}, \tag{31}$$

with vector $\mathbf{s}$ defined as

$$\mathbf{s} = \sum_{i=1}^{N} v_i \mathbf{n}_i. \tag{32}$$

We proceed by multiplying (31) by $\mathbf{n}_i$ and taking the sum over i. The resulting equation for $\mathbf{s}$ reads

$$\mathbf{s}\left[\omega_0 - \omega + \frac{i}{\tau_0}(1-\alpha)\right] + \frac{i\alpha}{\tau_0}\sum_i \mathbf{n}_i(\mathbf{n}_i \cdot \mathbf{s}) = -\frac{i}{2}\sum_i \mathbf{n}_i e^{-i\phi_i}. \tag{33}$$

The second term depends on the orientations of the dipole moments. If all dipoles are parallel we return to Eq. (29). If, however, the dipoles orientations are random, the sum over oscillators in (33) yields $N\mathbf{s}/3$. This leads to a spectrum $I(\omega)$ in the form similar to that of Eq. (30) with the width of the broad Lorentzian $\Gamma_s \approx N/3\tau_0$, i.e. three times smaller than Γ_s in the ideal case. Thus we conclude that for large N, contrary to the case $N = 2$, orientational disorder only weakens, but does not destroy the cooperative emission.

iii) Diagonal disorder

When the frequencies ω_i of the oscillators are different, the system of Eqs. (19) can still be solved in a closed form by following the above procedure for the ideal case. Instead of Eq. (29) we get the following equation for σ (defined in Eq. (28)).

$$\sigma\left[1 + \frac{i\alpha}{\tau_0}\sum_k \frac{1}{\omega_k - \omega + \frac{i(1-\alpha)}{\tau_0}}\right] = -\frac{i}{2}\sum_k \frac{e^{-i\phi_k}}{\omega_k - \omega + \frac{i(1-\alpha)}{\tau_0}}. \tag{34}$$

The following steps include expressing v_i through σ, substituting them into Eq. (14) and averaging over phases. We present here only the final result:

$$I(\omega) = \frac{\alpha F_2 - (1-\alpha)[2\alpha(1-\alpha)F_4 + \alpha^2\{2F_1F_3 + 2(1-\alpha)^2F_2F_4 - F_2^2\}]}{\alpha^2F_1^2 + [1+\alpha(1-\alpha)F_2]^2} + (1-\alpha)F_2, \tag{35}$$

where the dimensionless functions $F_1(\omega), F_2(\omega), F_3(\omega)$, and $F_4(\omega)$ are defined as

$$F_1(\omega) = \sum_k \frac{(\omega_k-\omega)\tau_0}{(\omega_k-\omega)^2\tau_0^2 + (1-\alpha)^2}, \quad F_2(\omega) = \sum_k \frac{1}{(\omega_k-\omega)^2\tau_0^2 + (1-\alpha)^2},$$
$$F_3 = \sum_k \frac{(\omega_k-\omega)\tau_0}{[(\omega_k-\omega)^2\tau_0^2 + (1-\alpha)^2]^2}, \quad F_4(\omega) = \sum_k \frac{1}{[(\omega_k-\omega)^2\tau_0^2 + (1-\alpha)^2]^2}. \tag{36}$$

To analyze the spectrum expressed in Eq. (35) we have plotted $I(\omega)$ for various sets of frequencies $\{\omega_k\}$. The random sets $\{\omega_k\}$ were computer generated within the interval $(-5/\tau_0, 5/\tau_0)$. We have gradually increased the number N of oscillators in the set. Some representative examples for $\alpha = 0.83$ are shown in Fig. 1. We see that for $N = 4$ the spectrum shows some structure with a characteristic period $\sim 1/\tau_0$. With increasing N the number of maxima increases and *they become narrower*. For $N = 16$ and $N = 24$ the corresponding spectrum has the form of a plateau with a fine structure superimposed on it. As N is increased further the number of maxima goes down due to their overlapping. The amplitude of the fine structure gradually decreases as it is illustrated in Fig. 1 for $N = 64$.

Obviously, the spectral narrowing of the maxima with increasing N is the consequence of the cooperative behavior of the system of oscillators. For noninteracting oscillators with widths $1/\tau_0$ the fine structure in the spectrum would be completely washed out for $N = 16$ and $N = 24$. Formation of the fine structure within a broad range of N is a generic feature of the emission spectrum of a system of oscillators coupled through their radiation field. This is our main result. The physical origin of the fine structure is studied in the next section, where we also discuss the role of the off-diagonal disorder.

3. DISCUSSION

Deeper understanding of the origin of the fine structure in the cooperative emission spectrum can be gained from the analysis of the eigenmodes in the system of oscillators with diagonal disorder. The eigenmodes correspond to the solutions of the system of Eqs. (19) in which the right-hand side is set to zero. As a result, Eq. (34) turns into

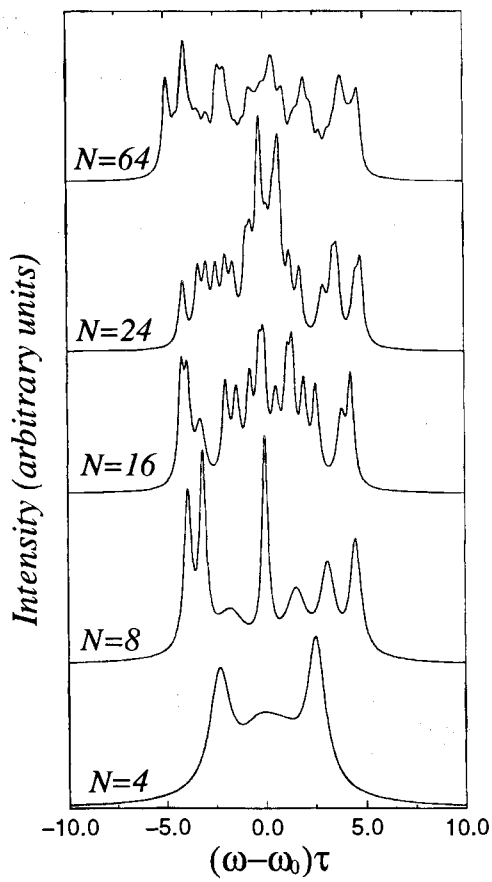

Figure 1. Evolution of the spectral intensity with increasing N is shown for $\alpha = 0.83$ and $\Omega\tau_0 = 5.0$

the following equation for the complex eigenfrequencies of the system

$$1+\frac{i\alpha}{\tau_0}\sum_k \frac{1}{\omega_k-\omega+\frac{i(1-\alpha)}{\tau_0}}=0. \tag{37}$$

Let us extract one term with k equal to some l from the sum over k in Eq. (37) and denote the remaining sum as Σ_0:

$$\Sigma_0=\sum_{k\neq l}\frac{1}{\omega_k-\omega+\frac{i(1-\alpha)}{\tau_0}}. \tag{38}$$

If we formally assume Σ_0 to be ω-independent, the solution of Eq. (37) can be easily obtained

$$\omega=\omega'+i\omega''=\omega_l+\frac{i}{\tau_0}(1-\alpha)+\frac{i\alpha}{\tau_0}\left[\frac{1}{1+\frac{i\alpha}{\tau_0}\Sigma_0}\right]. \tag{39}$$

The idea behind rewriting Eq. (37) in the form of Eq. (39) is that we anticipate the real parts of the eigenfrequencies to be close to the bare oscillators frequencies. Let us estimate Σ_0 when the real part of ω equals to ω_l. Let us assume for this estimate that the values ω_k are evenly spaced; i.e. $\omega_k=\omega_0-\Omega(1-2k/N)$. Then the real and imaginary parts of Σ_0 can be presented as follows:

$$Re\Sigma_0=\Sigma_0'=\frac{N}{2\Omega}\ln\left[\frac{N-l}{l}\right],\quad Im\Sigma_0=\Sigma_0''=-\left(\frac{1-\alpha}{\tau_0}-\omega''\right)\frac{N^2}{4\Omega^2}\sum_{k\neq l}\frac{1}{(k-l)^2+[\frac{1-\alpha}{\tau_0}-\omega'']^2\frac{N^2}{4\Omega^2}}, \tag{40}$$

where the sum over k in $Re\Sigma_0$ was evaluted by replacing it with an integral. We see from Eq. (40) that the real $\Sigma_0'\sim N/\Omega$, while σ_0'' does not exceed N/Ω. This leads us to the conclusion that if N is much smaller than $\Omega\tau_0$, one can neglect Σ_0 in Eq. (39) and obtain $\omega''=1/\tau_0$. In fact, this conclusion could be anticipated. The mean spacing $\Delta\omega$ between the bare frequencies ω_k is Ω/N; when $\Delta\omega$ is much bigger than the inverse lifetime of an individual oscillator ($\Delta\omega\gg 1/\tau_0$), the oscillators do not "feel" each other. The opposite case, when $N\gg\Omega\tau_0$, is less trivial. In this case we have $\frac{1}{\tau_0}\Sigma_0'\gg 1$, which means that the second term in the right-hand side of Eq. (39) is strongly suppressed. This is the reason for the narrowing of the spectral maxima with increasing N in Fig. 1. It can be readily seen from Eqs. (39), (40) that if $N\ll\Omega\tau_0/(1-\alpha)$ (but at the same time $N\gg\Omega\tau_0$, since α is close to 1) then the imaginary part of the eigenfrequency is $\omega''\sim\Omega/N\ll 1/\tau_0$, i.e. ω'' falls off as N^{-1}. Then, as N becomes much larger than $\Omega\tau_0/(1-\alpha)$, Eq. (39) yields $\omega''=(1-\alpha)/\tau_0$ independently of N.

At this point it is important to note that N and α are not the right experimental parameters. In the experiment the excitation intensity I_0 determines the concentration n of the oscillators. To connect the previous consideration to the experiment we argue in the following way. Consider some volume L^3 within a large system of oscillators. Suppose that the average number of oscillators, enclosed inside this volume, $N_L=nL^3$ is big ($N_L\gg 1$). Since the typical distance between two oscillators within the volume is $\sim L$, we can estimate from Eq. (18) the typical value of the parameter α, which determines the oscillators coupling, $(1-\alpha_L)\sim(L/\lambda_0)^2$. Using the values N_L and α_L, we can then calculate the frequencies of the N_L eigenmodes by solving Eq. (37). The crucial question now is whether these eigenmodes, calculated for the isolated system, are the real eigenmodes of the larger system? In other words, whether or not the coupling of oscillators inside the volume L^3 with the oscillators surrounding this volume would perturb significantly the calculated frequencies of the eigenmodes. The answer certainly depends on the magnitude of L. Suppose that L increases gradually. It follows from the above analysis that the magnitude of the imaginary parts of the eigenfrequencies depends on the relation between N_L and $\Omega\tau_0/(1-\alpha_L)$. The crossover between the two regimes corresponds to $N_L\sim\Omega\tau_0/(1-\alpha_L)$. The latter condition determines the characteristic size

$$L=L_0=\left(\frac{\Omega\tau_0\lambda_0^2}{n}\right)^{1/5}. \tag{41}$$

This size is relevant if it is much smaller than the wavelength λ_0. The corresponding condition $L_0<\lambda_0$ can be rewritten as $\Omega\tau_0<n\lambda_0^3$. If L is smaller than L_0, we have $\omega''\sim\Omega/N_L$; i.e. the imaginary parts of the eigenfrequencies are of the order of the mean spacing between the real parts. The fact that α_L differs from 1 is not important in

this regime. This, in turn, implies that the eigenmodes of two neighboring volumes L^3 will be strongly coupled to each other. Thus, the eigenmodes of the volume $L^3 < L_0^3$ are *not* the eigenmodes of the larger system. In the opposite case $L > L_0$ our analysis yielded $\omega'' = (1 - \alpha_L)/\tau_0 > \Omega/N_{L_0}$. This means that the frequency widths of the eigenmodes are bigger than the mean spacing between them; i.e. such eigenmodes are not well defined. The reason for such an inconsistency is that in a large enough system the assumption that the parameter α is the same for each pair of oscillators is not justified. Therefore, we arrive to the conclusion that the *real* eigenmodes of the large system occupy the volume $\sim L_0^3$ and comprise $\sim N_{L_0}^3$ oscillators. The typical value of ω'' for these eigenmodes is given by

$$\omega'' \sim \frac{\Omega}{N_{L_0}} \sim \frac{1}{\tau_0}\left(\frac{\Omega\tau_0}{n\lambda_0^3}\right)^{2/5}. \tag{42}$$

Recall now, that the concentration n of the oscillators is proportional to the excitation intensity I_0. As a result, the characteristic volume L_0^3 and correspondigly the characteristic number of oscillators N_{L_0} appear to depend on I_0. It then follows from Eq. (42) that the width ω'' decreases with I_0. Consequently, the peaks in the emission spectrum $I(\omega)$ become more pronounced with increasing the excitation intensity. To study quantitatively the evolution of the spectrum with I_0 one should use Eq. (35) with $N = N_{L_0}$ and $\alpha = \alpha_{L_0}$. An example of such a calculation is presented in Fig. 2.

In conclusion of this section let us discuss the role of the off-diagonal disorder. This type of disorder influences the cooperative emission spectrum in two ways. Firstly, even for an ideal system, the dipole-dipole interaction causes a certain spread in the frequencies of the eigenmodes due to the randomness in the inter oscillators distances. Secondly,

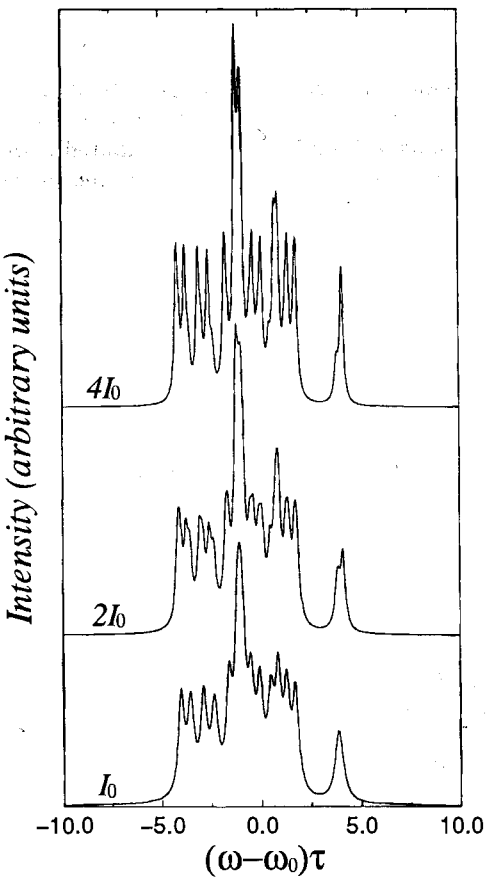

Figure 2. Evolution of the spectral intensity with increasing excitation is shown for $\Omega\tau_0 = 5.0$. The spectra are calculated for 16 random frequencies with the excitation intensities I_0 ($\alpha = 0.83$), $2I_0$ ($\alpha = 0.87$), and $4I_0$ ($\alpha = 0.90$).

the dipole-dipole interaction leads to the coupling between different oscillators, which is additional to the coupling through the radiation field. In other words, if we set all α_{ij} to zero, the eigenmodes of the system of oscillators would still comprise of many individual oscillators. Assuming $\alpha = 0$, let us denote with $\mathbf{w}_\nu$ the normalized N-component vector, corresponding to the eigenmode ν in the presence of the dipole-dipole interaction. Let the corresponding (real) eigenfrequency be ω_ν. Now let us take into account that α is finite. Then the equation for the complex eigenfrequencies of the system takes the form

$$1 + \frac{i\alpha}{\tau_0} \sum_\nu \frac{(\mathbf{1} \cdot \mathbf{w}_\nu)^2}{\omega_\nu - \omega + \frac{i(1-\alpha)}{\tau_0}} = 0, \tag{43}$$

where $\mathbf{1}$ is the vector with all N components equal to unity. The latter equation generalizes Eq. (37) to the case of a finite dipole-dipole interaction. Comparing Eq. (43) to Eq. (37), we conclude that the applicability of our previous analysis relies on two conditions: (1) the spread in frequencies, caused by the dipole-dipole interaction is smaller than the diagonal disorder; (2) the eigenvectors $\mathbf{w}_\nu$ are completely random. The latter condition implies that for $N \gg 1$ the scalar product $(\mathbf{1} \cdot \mathbf{w}_\nu)^2$ in Eq. (43) can be (on average) replaced by unity.

4. CONCLUSION

In the present paper we have studied the effect of disorder on the cooperative emission from a system of classical oscillators. The main conclusion is that the emission from a disordered system retains its cooperative character if the concentration of oscillators is sufficiently high. More precisely, the necessary condition for the cooperative emission is that the spacing between the neighboring oscillators frequencies within the volume λ_0^3 is smaller than the individual spectral width.

The most interesting qualitative result of our study can be formulated as follows. In an ideal (homogeneous) system the spectrum of cooperative emission is composed of a superposition of a broad and a narrow peak. In the presence of disorder (if the spread in the frequencies 2Ω satisfies the above criterion), the single narrow peak splits into a group of *narrow* peaks. The positions of these peaks are distributed within an interval $(\omega_0 - \Omega, \omega_0 + \Omega)$. This means that the eigenmodes of the system comprise many oscillators even in the presence of disorder. According to Eq. (42) the width of an individual narrow peak decreases with the excitation intensity I_0 as $I_0^{-2/5}$. These narrow peaks may be observed as a fine structure in the emission spectrum. Remarkably, with increasing the size D of the system (determined by the area of the illuminated spot) the fine structure does not disappear abruptly: the characteristic period of the modualtion remains the same, while the r.m.s. amplitude falls off as $D^{-3/2}$.

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