The exact solution for the fluorescence of low density Frenkel excitons in
double and triple lattice-layers

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In low density regime, the fluorescence of Frenkel excitons in crystal slab can be studied
without the aid of rotating wave and Markoffian approximation. The equations for the
case of double and triple lattice-layers are now solved exactly to give the eigen decay rates,
frequency shifts and the statistical properties of the fields.

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I. INTRODUCTION

The fluorescence of excitons in a quantum well or crystal slab is of charater of collective radiation, since
exciton, as exited state of the whole quantum well (crystal slab) has a collective transition dipole moment.
However not all these collective radiation are superradiance. Actually, the exciton emission has many eigen
modes, some of them are superradiant modes and some of them are subradiant modes.

Exciton have important application in photonic devices because exciton device may have small size,
low power dissipation, high speed and high efficiency. All of these are needed by integrated photo-electric
circuits.

It is well known that the exciton in bulk crystal does not radiate, but forms polariton instead\cite{1,2}. This
shows that a general treatment of exciton radiation should take the reabsorption effect into account.

There has been quite a lot of theoretical studies on fluorescence of excitons\cite{3–10}. In the case of Frenkel
excitons of low density, Knoester studied\cite{5} the crossover from superradiant excitons to bulk polaritons
when the number of the lattice-layers in the crystal increases without bound. He gives the correct form of
the eigen equation for the frequency shift and decay rate in the single lattice layer case. We have pointed
out\cite{11} that it should count in the two-photon coupling term properly in order to get this correct form
of eigen equation. Knoester proposed\cite{5} that $F_{kk'}$ which describe the coupling between the excitons of
wave vector $k$ and $k'$ by exchange of photons is strongly peaked around $k = k'$ and one may keep only
the diagonal elements to a good approximation. There are some ambiguities in this proposition, since the
set of the values for $k$ exists different selections. Besides, when one derives the whole set of eigen decay
rates, some of them are small (subradiant modes), the contribution from off diagonal elements could be
important.

We have studied the single lattice layer case in some detail\cite{11}. However in the case of Frenkel exciton
fluorescence, neither single layer case nor the very thick case is important in practice. In this paper we
will study the fluorescence of Frenkel exciton in thin crystal film of double and triple lattice-layers. The
Heisenberg equations without rotating wave approximation are solved in the low density regime without
Mackoffian approximation. The two-photon coupling term in the interaction Hamiltonian is included
properly. All eigen decay rates, frequency shifts as well as the evolution of fields in terms of their initial
values are obtained consequently. We note the non-diagonal elements of $F_{kk'}$ is essential to derivation of
these results. Our approach can be readily generalized to the case of more lattice layers.
The exciton-phonon interaction is not taken into account in this investigation. We shall give a brief review of general formulation in section 2. Section 3 and 4 are devoted to the cases of double and triple lattice-layers respectively. A brief conclusion is given in section 5.

II. BRIEF REVIEW OF GENERAL FORMULATION

We first write down the general formulation for the crystal slab of $N$ lattice-layers. The crystal is assumed to have simple cubic structure. When the two-photon coupling term $\frac{\hbar^2}{mc}A^2$ is taken into account, the interaction Hamiltonian between exciton and photon for the low density excitons are described as\[11\]

$$
\hat{H}_{\text{int}} = \hbar \sum_{q,k} G(q)O(k + q)[\hat{B}_k(t) + \hat{B}_{-k}^+(t)][\hat{a}_q(t) + \hat{a}_{-q}^+(t)] + 
$$

$$
\hbar \sum_{q,q',k} \frac{1}{\Omega} G(q)G(q')O(q' - k)O(k + q)[\hat{a}_q(t) + \hat{a}_{-q}^+(t)][\hat{a}_{q'}(t) + \hat{a}_{-q'}^+(t)]
$$

where $q$ and $k$ are wave vectors for photon and exciton respectively, they are in the $z$-direction, perpendicular to the crystal slab, $\hat{B}_k(t)$ and $\hat{a}_q(t)$ are exciton and photon annihilation operators respectively, $\hat{B}_k(t)$ and $\hat{B}_{-k}^+(t)$ are assumed to satisfy the boson commutation relations

$$
[\hat{B}_k(t), \hat{B}_{k'}^+(t)] = \delta_{kk'},
$$

and $\Omega$ is the frequency of the isolated lattice atom, $G(q)$ is the coupling constant,

$$
G^2(q) = \frac{2\pi N_T \Omega^2}{V h |q| c} d^2
$$

where $V = AL$ is the normalization volume for the photon, $N_T$ is the total number of lattice sites in the crystal slab ($N_T = N N_L$, $N_L$ is the number of lattice sites in each layer). $d$ denotes the transition dipole moments of a single lattice atom supposed for simplicity perpendicular to the $z$-axis.

In Ref.\[5\] it is said that $k$ takes the discrete value

$$
k = \frac{2\pi m}{N a}
$$

with $m = 0, 1, \cdots N - 1$. But in the eq(1) of Ref.\[5\] as well as in our $\hat{H}_{\text{int}}$, $k$'s are assumed symmetric with respect to the zero, hence we should take $m = -\frac{1}{2}(N - 1), \cdots , \frac{1}{2}(N - 1)$ instead\[11\]. The values of $q$ is as usual:

$$
q = \frac{2\pi j}{L}, \quad j = 0, \pm 1, \pm 2, \cdots , \pm \infty.
$$

$O(k - q)$ is the wave-vector matching factor, now takes the form\[11\]

$$
O(k - q) = \frac{1}{N} \sum_{l = -\frac{1}{2}(N - 1)}^{\frac{1}{2}(N - 1)} e^{i(k - q)l a} = \frac{1}{N} \sum_{l = -\frac{1}{2}(N - 1)}^{\frac{1}{2}(N - 1)} \sin \frac{\pi}{2}(k - q)Na
$$

$l$ is the index for the layers. In case $k = q$, $O(k - q)$ will equal to one, when $k \neq q$ and $N$ is sufficient large, $O(k - q)$ will be small.

We notice that no rotating wave approximation is made in eq.(1), and the two terms on the right-hand side correspond to single-photon coupling and two-photon coupling respectively.

From $\hat{H}_{\text{int}}$ and the commutation relation, one may immediately write down the Heisenberg equations for $\hat{B}_k(t)$, $\hat{a}_q(t)$ and their hermitian conjugates. These equations are linear equations so that they can be solved exactly. Carrying out the half-side Fourier transformation
\[ \hat{B}_k(\omega) = \int_0^\infty \hat{B}_k(t)e^{i\omega t}dt, \quad \hat{B}^+_k(\omega) = \int_0^\infty \hat{B}^-_k(t)e^{i\omega t}dt, \quad \text{etc.} \] (6)

as did in Ref. [5] and then eliminating the photon operators, we get

\[
\sum_{k'} \left[ (\omega^2 - \Omega^2)\delta_{kk'} - \frac{2\omega^2}{\Omega} F_{kk'}(\omega) \right] [\hat{B}_k(\omega) - \hat{B}^+_k(\omega)]
= i[(\omega + \Omega)\hat{B}_k(0) - (\omega - \Omega)\hat{B}^-_k(0)] - 2i\omega \sum_{k'} F_{kk'}(\omega) [\hat{B}_{k'}(0) - \hat{B}^+_k(0)]
+ 2i \sum_q \omega G(q)O(k - q) \left[ \hat{a}_q(0) + \frac{\hat{a}^+_q(0)}{\omega - |q|c} - \frac{\hat{a}^-_q(0)}{\omega + |q|c} \right]
\] (7)

where \( \hat{B}_{k'}(0) \) means \( \hat{B}_{k'}(t = 0) \), etc. and

\[
F_{kk'}(\omega) = \sum_q \frac{2|q|c}{\omega^2 - q^2c^2} G^2(q)O(k - q)O(k' - q)
= -\frac{Na\Omega f^2}{4\pi c^2} \int dq \frac{O(k - q)O(q - k')}{q^2 - \omega^2/c^2},
\] (8)

in which

\[ f^2 = \frac{8\pi\Omega a^2}{\hbar c^3}. \] (9)

Note that our \( F_{kk'}(\omega) \) and \( O(k - q) \) are somewhat different from those defined in Ref. [5]. For simplicity we have neglected the term of static dipole-dipole interactions.

We should mention that even disregarding the terms proportional to \( \hat{a}_q(0) \) and \( \hat{a}^-_q(0) \), eq. (7) is still different from the result of Ref. [5] as mentioned in Ref. [11].

### III. THE CASE OF DOUBLE LATTICE-LAYERS

Now we consider the special case \( N = 2 \). The wave-vector matching factor for \( N = 2 \) becomes

\[ O(k - q) = \cos \frac{1}{2}(k - q)a \] (10)

where \( k \) takes the values \( \pm \frac{\pi}{2a} \). We shall use \( F_{++}(\omega), F_{--}(\omega) \) to represent \( F_{\pm \pm}(\omega), F_{\mp \mp}(\omega) \) etc. and evaluate the integrals in eq. (8) by contour integration. The results are

\[
F_{++}(\omega) = F_{--}(\omega) = -\frac{\eta\Omega}{\omega}, \quad F_{+-}(\omega) = F_{-+}(\omega) = -\frac{\eta\Omega}{\omega} e^{i\omega c}
\] (11)

where

\[ \eta = \frac{af^2}{2c}. \] (12)

We note that the nondiagonal elements (\( F_{++} \) and \( F_{--} \)) is of the same orderly as the diagonal elements (\( F_{++} \) and \( F_{--} \)).

The coupled equation (7) now becomes

\[
(\omega^2 + i\omega\eta - \Omega^2)[\hat{B}_+(\omega) - \hat{B}^+_+(\omega)] + i\omega\eta e^{i\omega\eta c}[\hat{B}_-(\omega) - \hat{B}^+_-(\omega)] = \hat{A}_0(\omega),
\]

\[
i\omega\eta e^{i\omega\eta c}[\hat{B}_+(\omega) - \hat{B}^+_+(\omega)] + (\omega^2 + i\omega\eta - \Omega^2)[\hat{B}_-(\omega) - \hat{B}^+_-(\omega)] = -\hat{A}^+_0(-\omega)
\] (13a, 13b)

where
\[ \hat{A}_0(\omega) = i[(\omega + \Omega + i\eta)\hat{B}_+(0) - (\omega - \Omega + i\eta)\hat{B}_-(0)] - \eta e^{\frac{\pi i}{2}}[\hat{B}_-(0) - \hat{B}_+^+(0)] \\
+ 2\sqrt{2}\omega i \sum_q G(q) \cos(\frac{\pi}{4} - \frac{qa}{2})[\hat{a}_q(0) - \hat{a}_{-q}(0)] \]

\[ \hat{A}_0^+(-\omega) = i[(\omega + \Omega + i\eta)\hat{B}_+^+(0) - (\omega + \Omega + i\eta)\hat{B}_-(0)] - \eta e^{\frac{\pi i}{2}}[\hat{B}_+(0) - \hat{B}_+^+(0)] \\
- 2\sqrt{2}\omega i \sum_q G(q) \cos(\frac{\pi}{4} + \frac{qa}{2})[\hat{a}_q(0) - \hat{a}_{-q}(0)] \]

Eqs.(13) are easily solved to get:

\[ \hat{B}_+(\omega) - \hat{B}_+^+(\omega) = \frac{(\omega^2 + i\omega\eta - \Omega^2)\hat{B}_0(\omega) + i\omega\eta e^{\frac{\pi i}{2}}\hat{A}_0^+(-\omega)}{(\omega^2 + i\omega\eta - \Omega^2 + i\omega\eta e^{\frac{\pi i}{2}})(\omega^2 + i\omega\eta - \Omega^2 - i\omega\eta e^{\frac{\pi i}{2}})} \]  

\[ \hat{B}_-(\omega) - \hat{B}_+^+(\omega) = -\frac{i\omega\eta e^{\frac{\pi i}{2}}\hat{A}_0(\omega) + (\omega^2 + i\omega\eta - \Omega^2)\hat{A}_0^+(-\omega)}{(\omega^2 + i\omega\eta - \Omega^2 + i\omega\eta e^{\frac{\pi i}{2}})(\omega^2 + i\omega\eta - \Omega^2 - i\omega\eta e^{\frac{\pi i}{2}})} \]

The roots of

\[ \omega^2 + i\omega\eta - \Omega^2 = 0 \]  

and

\[ \omega^2 + i\omega\eta - \Omega^2 = 0 \]

will determine the eigen decay rates and corresponding frequency shifts. These roots cannot have positive imaginary part, namely the poles of \( \hat{B}_+(\omega) - \hat{B}_+^+(\omega) \) will not be localized in the upper half \( \omega \)-plane, since two necessary conditions can be deduced for eq.(15) to have root of positive imaginary part:

\[ \eta > 2\Omega \quad \text{and} \quad \frac{\eta a}{c} > 2\pi \]

and both of these conditions are untenable. Similarly eq.(16) also cannot have root of positive imaginary part. These results mean the basic physics laws will not be violated as in the case of monolayer\[11\].

We have derived four physical roots of eqs.(15) and (16) as:

\[ \omega_1 = \Omega_1 - i\Gamma_1, \quad \omega_2 = -\Omega_1 - i\Gamma_1 \]

\[ \omega_3 = \Omega_2 - i\Gamma_2, \quad \omega_4 = -\Omega_2 - i\Gamma_2 \]

in which

\[ \Omega_1 \cong \Omega(1 - \frac{\eta^2}{2c^2}, \quad \Gamma_1 \cong \eta, \]

\[ \Omega_2 \cong \Omega(1 - \frac{\eta^2}{2c^2}), \quad \Gamma_2 \cong \frac{1}{4}\frac{\Omega^2q^2}{c^2} \]

In the following we will omit the terms proportional to \( \hat{a}_q(0), \hat{a}_{-q}(0) \) as did in Ref[5], since here we just study the fluorescence of excitons. Then the electric field is derived as follows\[11\]

\[ \hat{E}(z,t) = \frac{1}{2\pi} \int_{\infty+i\epsilon}^{\infty-i\epsilon} d\omega \hat{E}(z,\omega) e^{-i\omega t}, \]

\[ \hat{E}(z,\omega) = i\sum_q \sqrt{\frac{2\pi|q|c}{V}} [\hat{a}_q(\omega) - \hat{a}_{-q}(\omega)] e^{iqz}, \]

\[ \hat{a}_q(\omega) - \hat{a}_{-q}(\omega) = \frac{2\omega \sqrt{N}}{\Omega(\omega^2 - q^2c^2)} G(q) \sum_k O(k - q) \]

\[ \times [\omega(\hat{B}_k(\omega) - \hat{B}_k^+(\omega)) - i(\hat{B}_k(0) - \hat{B}_k^+(0))]. \]
The summation of $q$ in eq.(19b) can be tranformed to integration and carried out by contour integration. In the positive $z$ region outside the crystal slab, we get for the double lattice-layer case

$$
\hat{E}(z, t) = \sum_{l=\pm, k=\pm} e^{ik\lambda l} \hat{B}_k(t) = \frac{1}{\sqrt{2}} [\hat{B}_+(t) + \hat{B}_-(t)],
$$

where $A$ is the area of each layer, it is also the cross area of the normalization volume for the photon as mentioned above.

The electric field $\hat{E}(z, t)$ in this region is calculated by eq.(19a), with the results

$$
\hat{E}(z, t) = 0, \quad \text{for} \quad z - ct > 0 \quad \text{(21a)}
$$

$$
\hat{E}(z, t) = \hat{E}(z, t) + h.c., \quad \text{for} \quad z - ct < 0 \quad \text{(21b)}
$$

where

$$
\hat{E}(z, t) = \frac{f}{c} \sqrt{\frac{\pi \hbar \Omega a}{8A}} (1 + \frac{\Omega}{\Omega_1} - i \frac{\Gamma}{\Omega_1}) \cos \frac{(\Omega_1 - i \Gamma) a}{2c} \left[ (\hat{B}_+(0) + \hat{B}_-(0)) + \frac{\Omega_1 - \Omega - i \Gamma}{\Omega_1 + \Omega - i \Gamma} (\hat{B}_+(0) + \hat{B}_-(0)) \right] e^{-\frac{\Delta_1 (t - \frac{h}{\Omega}) - \Gamma_1 (t - \frac{h}{\Omega})}{2c}}
$$

$$
\hat{E}(z, t) = \frac{f}{c} \sqrt{\frac{\pi \hbar \Omega a}{8A}} (1 + \frac{\Omega}{\Omega_2} - i \frac{\Gamma}{\Omega_2}) \sin \frac{(\Omega_2 - i \Gamma) a}{2c} \left[ (\hat{B}_+(0) - \hat{B}_-(0)) - \frac{\Omega_2 - \Omega - i \Gamma}{\Omega_2 + \Omega - i \Gamma} (\hat{B}_+(0) - \hat{B}_-(0)) \right] e^{-\frac{\Delta_2 (t - \frac{h}{\Omega}) - \Gamma_2 (t - \frac{h}{\Omega})}{2c}}.
$$

We note that this solution is free from Markoffian approximation and also free from antirotating wave interaction.

The electric field in the $z < 0$ region can be derived similarly, with the resultant waves propagating in backward $z$ direction as expected.

There are two eigen decay rates appeared in the $\hat{E}(z, t)$: $\Gamma_1$ and $\Gamma_2$. The corresponding eigen modes are linear combination of the two modes of $m = \frac{1}{2}$ and $m = -\frac{1}{2}$. As can be seen from eq.(20), these two eigen modes, which will be called as superradiant mode and subradiant mode, correspond to the operators $\frac{1}{\sqrt{2}} (\hat{B}_+(0) + \hat{B}_-(0))$ and $\frac{1}{\sqrt{2}} (\hat{B}_+(0) - \hat{B}_-(0))$ respectively. Hence, They correspond to modes of $k = 0$ and $k = 1$ with the operators

$$
\hat{B}_0(t) = \frac{1}{2} \sum_{l=\pm, k=\pm} e^{ik\lambda l} \hat{B}_k(t) = \frac{1}{\sqrt{2}} [\hat{B}_+(t) + \hat{B}_-(t)],
$$

$$
\hat{B}_1(t) = \frac{1}{2} \sum_{l=\pm, k=\pm} e^{ik\lambda l} \hat{B}_k(t) = \frac{1}{\sqrt{2}} [\hat{B}_+(t) - \hat{B}_-(t)].
$$

Evidently, the dipoles of the two layers have the same phase for the former and have opposite phase for the latter. We note that the decay rate $\Gamma_2$ of subradiant mode here is still as large as $\frac{3}{2}$ times the decay rate of a single atom(molecular), because the atoms in each layer are still cooperated. The decay rate of $k = 0$ mode is twice of that of monolayer, which is just the character of superfluorescence. As can be seen from eq.(22) that even for the superradiant mode in which the emission is totally collective, the emitted light still may have different statistics and coherent properties according to the initial exciton state (also see the discussion in Ref[11]). For example, the coherent part of the electric field $< \hat{E}(z, t) >$ will be nonzero when the initial state of the exciton is a coherent state. But when the initial density matrix of the exciton is diagonal in Fock representation (including number state, chaotic state), the coherent part of $< \hat{E}(z, t) >$ will be zero.

Up to the first order of $\frac{\hbar a}{\Omega}$ and $\frac{\Omega}{\gamma}$, the $\hat{E}(z, t)$ is expressed by the superradiant mode operator $\hat{B}_0(0)$ and the subradiant mode operator $\hat{B}_1(0)$ as follows:
\[ \hat{E}(z, t) = \sqrt{\frac{2 \pi \eta \Omega}{cA}} [\hat{B}_0(0) - \frac{i\eta}{2\Omega} \hat{B}_0^+(0)] e^{-i\Omega x (t - \frac{z}{c}) - \Gamma_1(t - \frac{z}{c})} + \sqrt{\frac{2 \pi \eta \Omega}{cA}} \hat{B}_1(0) e^{-i\Omega x (t - \frac{z}{c}) - \Gamma_2(t - \frac{z}{c})} + h.c. \] (24)

for \( z > 0 \) and \( t - \frac{z}{c} > 0 \). Similar results for \( z < 0 \), \( t + \frac{z}{c} > 0 \). \( \hat{E}(z, t) \) is equal to zero if \( (z > 0, t - \frac{z}{c} < 0) \) or \( (z < 0, t + \frac{z}{c} < 0) \).

Since we have seen irregular behavior in the usual intensity operator for the solution of single layer case\[11\], here only the energy flux operator is given instead. The energy flux is usually defined by

\[ \hat{S}(z, t) = \frac{c}{4\pi} : \hat{E}(z, t) \times \hat{B}(z, t) :. \] (25)

It is readily to show that \( \hat{S} \) is always directed outward from the crystal film. So we rewrite \( \hat{S} \) as \( \vec{n} \hat{S} \) which \( \vec{n} \) is unit vector directing outer space from lattice-layers. Namely, it is in positive \( z \) direction in the \( z > 0 \) region and in negative \( z \) direction in the \( z < 0 \) region as required.

So we may obtain \( S(z, t) \) from eq.(24). After neglecting oscillating terms and higher order terms of \( \frac{c}{\Omega} \) and \( \frac{2s}{c} \) (only keep first order terms), we have:

\[ \hat{S}(z, t) = \frac{\eta \Omega}{A} [\hat{B}_0^+(0) \hat{B}_0(0) + \frac{i\eta}{2\Omega} \hat{B}_0^2(0) - \frac{i\eta}{2\Omega} \hat{B}_0^{+2}(0)] e^{2\eta(x - t)} + \eta \frac{\Omega}{A} \hat{B}_1^+(0) \hat{B}_1(0) e^{2\eta(x - t)} + \frac{\sqrt{\eta \Omega}}{A} [\hat{B}_0^+(0) \hat{B}_1(0) + \hat{B}_0(0) \hat{B}_1(0)] + \frac{i\eta}{2\Omega} (\hat{B}_0^+(0) \hat{B}_1(0) - \hat{B}_1(0) \hat{B}_0^+(0))] e^{(\eta + \eta')(x - t)}, \] (26a)

with

\[ \eta' = \frac{\Omega^2 a^2}{4c^2}. \] (26b)

We see from eq.(26) that the energy flux decays in three different rate. The first term which is contributed by the exciton of the short lifetime plays an important part at the beginning time. The second term contributed by the exciton of the long lifetime becomes dominant at late time. The third term will exhibit itself in the intermediate stage.

**IV. THE CASE OF TRIPLE LATTICE-LAYERS**

The cases of odd \( N \) and even \( N \) have a qualitative difference in the \( m \)-value series \(-\frac{1}{2}(N-1), \cdots, \frac{1}{2}(N-1)\) for eq.(4). In the former case, \( m \) contains zero, while in the latter, not. \( N = 3 \) is the simplest case of odd \( N \), apart from the trivial case \( N = 1 \), which has no nondiagonal terms \( F_{mm'} \) (here and in the following we use \( F_{mm'} \) to denote \( F_{kk'} \) according to the relation \( k = \frac{2\pi m}{N\pi} \)). Thus we will study it as an example. For \( N = 3 \)

\[ O(k - q) = \frac{1}{3} [2 \cos(k - q)a + 1] \] (27)

leading to the matrix \( F \) (with elements \( F_{mm'} \), \( m, m' = 1, 0, -1 \)) as

\[ F(\omega) = -\frac{ia^2 \Omega}{12 \omega c} \begin{pmatrix} -x^2 - 2x + 3 & -x^2 + x & 2x^2 - 2x \\ -x^2 + x & 2x^2 + 4x + 3 & -x^2 + x \\ 2x^2 - 2x & -x^2 + x & -x^2 - 2x + 3 \end{pmatrix} = -\frac{\eta \Omega}{6\omega} \eta D(\omega), \] (28)

where \( x = e^{i\delta} \equiv e^{i\delta} \). To the second order of \( \delta \),
We see that the nondiagonal elements are of the same order of magnitude as they should be. The eigen decay rates and corresponding frequency shifts are determined by the roots of the following equation:

\[
(\omega^2 - \Omega^2)[\hat{B}_m(\omega) - \hat{B}_m^+(\omega)] + \frac{1}{3} \eta \omega (B + 2A) (\omega^2 - \Omega^2)^2 + \frac{1}{9} \eta^2 \omega^2 (2AB + A^2 - E + E - 2C^2)(\omega^2 - \Omega^2) \\
+ \frac{1}{27} \eta^3 \omega^3 (A^2 - E^2)B + 2C^2 (E - A) = 0,
\]

where \(A, B, C\) and \(E\) are matrix elements of \(D(\omega)\), defined as follows:

\[
D(\omega) = \begin{pmatrix}
A(\omega) & C(\omega) & D(\omega) \\
C(\omega) & B(\omega) & C(\omega) \\
D(\omega) & C(\omega) & A(\omega)
\end{pmatrix}.
\]

We get the six roots of eq.(31) as follows:

\[
\omega_1 = \Omega_1 - i\Gamma_1, \quad \omega_1' = -\Omega_1 - i\Gamma_1,
\]
\[
\omega_0 = \Omega - i\Gamma, \quad \omega_0' = -\Omega_0 - i\Gamma_0,
\]
\[
\omega_{-1} = \Omega_{-1} - i\Gamma_{-1}, \quad \omega_{-1}' = -\Omega_{-1} - i\Gamma_{-1}.
\]

with

\[
\Omega_1 \approx \Omega(1 - \frac{\eta a}{3c}), \quad \Gamma_1 = \frac{1}{27} \eta \Omega a^2 c^2, \tag{34a}
\]
\[
\Omega_0 \approx \Omega(1 - \frac{9\eta a}{8\Omega^2} + \frac{4\eta a}{3c}), \quad \Gamma_0 = \frac{3}{2} \eta \Omega \tag{34b}
\]
\[
\Omega_{-1} \approx \Omega(1 - \frac{\eta a}{c}), \quad \Gamma_{-1} = \eta \Omega^2 a^2 c^2. \tag{34c}
\]

All the roots are in the lower half plane of complex \(\omega\) as they should be.

The direct way to solve for \(\hat{B}_m(\omega) - \hat{B}_m^+(\omega)\) from eq.(30) is to diagonalize the matrix \(D(\omega)\) defined by eq.(28). Up to second order of \(\delta\), we get the transformation matrix \(T(\omega)\) which satisfies

\[
T(\omega)D(\omega)\overline{T}(\omega) = \begin{pmatrix}
D_1(\omega) & D_0(\omega) & D_{-1}(\omega)
\end{pmatrix}
\]

as

\[
T(\omega) = \begin{pmatrix}
\frac{M}{\sqrt{2}} & -\sqrt{2}CM & M \\
\frac{M}{B - A - E} & M & -\frac{M}{\sqrt{2}} \\
0 & B - A - E & -\frac{M}{\sqrt{2}}
\end{pmatrix}.
\]
where
\[
M = \frac{1}{\sqrt{1 + \frac{2c^2}{(B-A-E)^2}}}. \tag{35c}
\]
The result for \( \hat{B}_m(\omega) - \hat{B}^+_m(\omega) \) is expressed then by
\[
\hat{B}_m(\omega) - \hat{B}^+_m(\omega) = \sum_{m'} T_{m', m} \omega^2 - \Omega^2 + \frac{\eta \omega D_{m'}}{3} \left[ (\omega + \Omega + \frac{\eta D_{m'}}{3}) \hat{\beta}^{(1)}_{m'}(\omega, 0) - (\omega - \Omega + \frac{\eta D_{m'}}{3}) \hat{\beta}^{(2)}_{m'}(\omega, 0) \right] \tag{36a}
\]
in which
\[
\hat{\beta}^{(1)}_{m}(\omega, 0) = \frac{1}{6} \sum_{m'} T_{mm'}(\omega) \hat{B}_{m'}(0), \quad \hat{\beta}^{(2)}_{m}(\omega, 0) = \frac{1}{6} \sum_{m'} T_{mm'}(\omega) \hat{B}^+_{m'}(0) \tag{36b}
\]
Substituting eqs.(36) into eqs.(19) and carrying out the integrations, it finally yields
\[
\hat{a}_q(\omega) - \hat{a}^+_q(\omega) = \frac{4\omega}{\omega^2 - q^2 c^2} G(q) \sum_{m,m'} \left[ 2 \cos\left( \frac{2\pi m}{3} - qa \right) + 1 \right] \times \frac{i T_{m'm}}{\omega^2 - \Omega^2 + \frac{\eta \omega D_{m'}}{3}} \left[ (\omega + \Omega) \hat{\beta}^{(1)}_{m'}(\omega, 0) - (\omega - \Omega) \hat{\beta}^{(2)}_{m'}(\omega, 0) \right], \tag{37a}
\]
and
\[
\hat{E}(z, \omega) = \frac{f}{c} \sqrt{\frac{6 \pi \hbar \Omega}{A}} \sum_{m,m'} \left[ 2 \cos\left( \frac{2\pi m}{3} - \frac{\omega a}{c} \right) + 1 \right] \times \frac{i T_{m'm}}{\omega^2 - \Omega^2 + \frac{\eta \omega D_{m'}}{3}} \left[ (\omega + \Omega) \hat{\beta}^{(1)}_{m'}(\omega, 0) - (\omega - \Omega) \hat{\beta}^{(2)}_{m'}(\omega, 0) \right] \tag{37b}
\]
and
\[
\hat{E}(z, t) = \sqrt{\frac{3 \pi \hbar \Omega}{c A}} \theta(t - \frac{z}{c}) \sum_m \hat{a}_m e^{-i\Omega_m (t - \frac{z}{c}) - \Gamma_m (t - \frac{z}{c})} + h.c., \tag{37c}
\]
for \( z > 0, t - \frac{z}{c} > 0 \), where
\[
\hat{a}_m = \frac{1}{\Omega_m} \sum_{m'} \left[ 2 \cos\left( \frac{2\pi m'}{3} - \frac{\omega_m a}{c} \right) + 1 \right] T_{m'm}(\omega_m) \left[ (\Omega + \omega_m) \hat{\beta}^{(1)}_{m}(\omega_m, 0) - (\Omega - \omega_m) \hat{\beta}^{(2)}_{m}(\omega_m, 0) \right]. \tag{37d}
\]
To the leading term, \( \hat{a}_m \) are given by
\[
\hat{a}_1 \cong \frac{\Omega a}{9c} [\hat{B}_1(0) + \hat{B}^-_1(0)],
\hat{a}_0 \cong \hat{B}_0(0),
\hat{a}_{-1} \cong \frac{\Omega a}{\sqrt{3}c} [\hat{B}_1(0) - \hat{B}^-_1(0)], \tag{38}
\]
We see that \( m = \pm 1 \) modes are not of eigen decay rates. On the contrary, the eigen modes are nearly maximum mix of these two modes.

In terms of the creation operator for an excitation in the \( \ell \)th layer\(^{[3,11]} \), we have

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\[
\hat{B}_k(t) = \frac{1}{\sqrt{N}} \sum_{l=-\frac{N}{2}}^{\frac{N-1}{2}} e^{-ikla} \hat{B}_l(t)
\]  

(39)

Thus we have, denoting \( \hat{B}_k \) by \( \hat{B}_m \) as before, the three eigen modes. They are approximated as following

\[
\frac{1}{\sqrt{2}} [\hat{B}_{m=1}(0) + \hat{B}_{m=-1}(0)] = \frac{1}{\sqrt{6}} [\hat{B}_{l=1}(0) + 2\hat{B}_{l=0}(0) - \hat{B}_{l=-1}]
\]

(40a)

\[
\hat{B}_{m=0} = \frac{1}{\sqrt{3}} [\hat{B}_{l=1}(0) + \hat{B}_{l=0}(0) + \hat{B}_{l=-1}(0)],
\]

(40b)

\[
\frac{1}{\sqrt{2}} [\hat{B}_{m=1}(0) - \hat{B}_{m=-1}(0)] = \frac{i}{\sqrt{2}} [\hat{B}_{l=1}(0) - \hat{B}_{l=-1}(0)],
\]

(40c)

while

\[
\hat{B}_{m=\pm 1}(0) = \frac{1}{\sqrt{3}} [\left(-\frac{1}{2} \mp \frac{\sqrt{3}}{2} i\right) \hat{B}_{l=1}(0) + \hat{B}_{l=0}(0) + \left(-\frac{1}{2} \pm \frac{\sqrt{3}}{2} i\right) \hat{B}_{l=-1}(0)].
\]

(40d)

We see the superradiant mode \((m = 0)\) has a decay rate \(2\Gamma_0 = 3\eta\) which is triplet of that for monolayer, showing the emission is totally cooperative. However, as in the double layer case, the statistical properties of the light of this mode still may have different varieties which depend on the initial state of the excitons. For the \( z > a \) region, we may also give the energy flux of the case of triple lattice-layers according to eq.(25) and eq.(37) as following:

\[
\langle \hat{S}(z, t) \rangle = \langle \hat{S}_1(z, t) \rangle + \langle \hat{S}_2(z, t) \rangle,
\]

(41)

where \( \langle \hat{S}_1(z, t) \rangle \) is the main part, it is expressed by

\[
\langle \hat{S}_1(z, t) \rangle = \frac{\hbar \Omega}{6\bar{A}} \{ 9(\hat{B}_0^+(0)\hat{B}_0(0)) e^{-3\eta(t-\tau)} \\
+ \frac{2\Omega^2a^2}{9\epsilon^2}(\hat{B}_1^+(0)\hat{B}_+)(0)) e^{-\frac{2\eta}{3}(t-\tau)} \\
+ \frac{6\Omega^2a^2}{\epsilon^2}((\hat{B}_1^+(0)\hat{B}_-(0)) e^{-3\eta(t-\tau)} \}
\]

(42)

with \( \hat{B}_\pm(0) = \frac{1}{\sqrt{2}}(\hat{B}_1(0) \pm \hat{B}_{-1}(0)) \) and \( \langle \hat{S}_2(z, t) \rangle \) may be approximated by

\[
\langle \hat{S}_2(z, t) \rangle = -i \frac{\hbar \Omega}{3\sqrt{3}A} \sqrt{\eta'} \delta [\hat{B}_1^+(0)\hat{B}_0(0) - \hat{B}_0^+(0)\hat{B}_+](0)] + \\
i3\sqrt{3}(\hat{B}_1^+(0)\hat{B}_0(0) - \hat{B}_0^+(0)\hat{B}_+)(0) ) e^{-\frac{4\eta}{3}(t-\tau)}.
\]

(43)

It only appears when the initial exciton density matrix in Fock representation has non-diagonal elements.

V. BRIEF SUMMARY

1. Knoester\textsuperscript{(4)} claimed that \( F_{kk'}(\omega) \) is diagonal to a good approximation. But we show explicitly that this is not generally true.

2. In low-density case, even the emission is superradiant in nature, the light still may have different coherent statistical properties, depending on the initial state of exciton.

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