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Superradiance and subradiance in systems of excited nuclei

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SUPERRADIANCE AND SUBRADIANCE
IN SYSTEMS OF EXCITED NUCLEI

by

TIAN-XIANG SHEN

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Abstract

This thesis studies superradiance and subradiance in the normal modes of excitation of systems of resonant nuclei. The nuclei are treated as classical electric dipole oscillators interacting through their radiation fields. This coupling radically alters the radiative decay of the system relative to the decay of an isolated nucleus, and leads to both superradiant and subradiant states with strongly enhanced or suppressed radiative decay rates, and with normal mode frequencies which can be strongly shifted from the natural resonance frequency. This issue is of considerable current interest because it is now possible to create spatially coherent single exciton nuclear states by illuminating crystals containing resonant Mössbauer nuclei with synchrotron radiation pulses. The purpose of this thesis is to solve for the radiative eigenmodes in a system of nuclei and to investigate superradiance and subradiance both in the eigenmode solutions and in the synchrotron radiation produced nuclear exciton state.
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I. Introduction

This thesis studies superradiance and subradiance in the normal modes of excitation of systems of resonant nuclei. The nuclei are treated as classical electric dipole oscillators interacting through their radiation fields.

That radiative coupling can radically alter the radiative decay rate of an excited system relative to that of an isolated atom or nuclei is well known.\textsuperscript{1, 2} Following Trammell \textsuperscript{2}, consider two identical nuclei with non-degenerate ground states a\textsubscript{1} and a\textsubscript{2} and excited states b\textsubscript{1} and b\textsubscript{2}. Because of the radiative coupling between the nuclei, the exponentially decaying states (normal modes) are not b\textsubscript{1}a\textsubscript{2} or a\textsubscript{1}b\textsubscript{2}, but rather the symmetric and anti-symmetric combinations \( \frac{1}{2}(b_1a_2 \pm a_1b_2) \). If \( \Gamma_\gamma \) is the partial width for radiative decay from either nucleus alone, the radiative decay rates of these two modes are:

\[
\Gamma_\pm = \frac{1}{2} \Gamma_\gamma \text{Im}[1\pm \exp(\text{i}k\cdot R)]^2 = \Gamma_\gamma (1\pm \frac{\sin kR}{kR}),
\]

where \( k \) is the wave vector of the emitted radiation, and \( R \) is the relative displacement between the two nuclei. For \( kR<<1 \), then \( \Gamma_+ = 2\Gamma_\gamma \) and \( \Gamma_- = 0 \). In that limit, the symmetric mode has an enhanced decay rate while the antisymmetric mode becomes non-radiative. We call the phenomena that a collective state of a system has a radiative decay rate larger than that of single nucleus "superradiance". If the decay rate is smaller than that of single nucleus, the mode is "subradiant".

This behavior is easy to understand classically. The symmetric mode corresponds to two oscillators oscillating together in-phase, which, for \( kR<<1 \), doubles the dipole moment. The radiation fields are doubled, and the radiation power is increased to four times that of a single oscillator. Since the total mechanical energy \( \mathcal{E}_\tau \) is also doubled, the decay rate for the system is \( -\frac{d\mathcal{E}_\tau}{dt}/\mathcal{E}_\tau = 2\Gamma_\gamma \). In contrast, in the anti-symmetric mode, the oscillators move 180\textdegree out of phase, and the radiation fields from the two oscillators interfere destructively, giving a very weak (electric quadruple) radiative decay.

In addition to the altered decay rates, the normal modes also exhibit shifted resonance frequencies. For the two nuclei system, the resonance shifts are (averaged over the direction \( \hat{R} \))
\[ \Delta \omega \approx \pm \Gamma \gamma \frac{\cos(kR)}{kR}, \]  

(1-2)

which will give large resonance shifts for \( kR \ll 1 \), with the symmetric mode oscillating at the lower frequency.

For a crystal of resonant nuclei, the effects can be much more pronounced. As a first approximation, the wave functions for the exponentially decaying excited states (normal modes) in a crystal might be expected to be of the form of Bloch waves,\(^ \text{2,4} \)

\[ |\Psi_e(q)\rangle = \frac{1}{\sqrt{N}} \sum_{l=1}^{N} e^{iq \cdot R_l} |e_l\rangle |G_0(l)\rangle, \]  

(1-3)

where \( |e_l\rangle |G_0(l)\rangle \) represents the state in which the \( l \)th nucleus is excited while all other nuclei remain in their ground states, \( R_l \) is the position of the \( l \)th nucleus and \( q \) is the Bloch vector. The partial width for the radiative decay of this "\( q \)th mode" is (in the Golden Rule approximation)

\[ \Gamma_e(q) = \frac{\Gamma \gamma}{4\pi N} \int d\Omega(n) |\sum_{l=1}^{N} \exp[-i(k-q) \cdot R_l]|^2, \]  

(1-4)

where \( \omega_0 \) is the wave vector of the outgoing radiation (\( \omega_0 \) is the resonance frequency of the nuclei). Most states \( |\Psi_e(q)\rangle \) are subradiant, with \( \Gamma_e(q) \) on the order of \( \frac{\Gamma \gamma}{N} \). However, if for some specific emission direction \( n \), the condition \( k-q=\tau \) holds, where \( \tau \) is a reciprocal vector of the crystal lattice, then the mode \( |\Psi_e(q)\rangle \) will have highly directional coherent decay about the \( q+\tau \) direction at an enhanced decay rate

\[ \Gamma_e(q) = \Gamma \gamma \pi n \lambda^2 L_{||}(q+\tau), \]  

(1-5)

where \( L_{||}(q+\tau) \) is the thickness of the crystal in the direction of \( (q+\tau) \) and \( n \) is the number of nuclei per unit volume. We will give a more detailed discussion of this result in Chapter II. For \( \lambda=1 \text{A}, n=0.1 \text{A}^{-3} \), we get a decay rate \( \Gamma_e=\Gamma \gamma \) for a crystal with a thickness of only 100A. Thus, in such a superradiant mode, if the crystal is sufficiently thick, the coherent decay rate will dominate the incoherent decay rate \( \Gamma \gamma \) and the decay rate due to internal conversion \( \Gamma_\alpha \).
Now in fact the Bloch waves $|\Psi_e(q)\rangle$ are generally not the true radiative normal modes in a crystal. As is shown in Chapter V, the proper forms of the eigenmodes for a parallel sided crystal have a sine or cosine modulation (coming in from the crystal faces) with a complex wave vector. Generally the Bloch state $|\Psi_e(q)\rangle$ is a superposition of these radiative eigenmodes, which have a spread of eigenfrequencies and decay rates. The important exception is the case in which $|q| = \frac{\omega_0}{c}$ and $q$ satisfies a Bragg condition $q \cdot \tau = \frac{1}{2} \tau^2$ where $\tau$ is a reciprocal vector of the crystal which is perpendicular to the parallel faces. In this case, $|\Psi_e(q)\rangle$ is a superradiant eigenmode, radiating at its natural resonance frequency $\omega_0$, and decaying at an enhanced decay rate including the contributions from coherent decay into both the $q$ and $q+\tau$ directions,

$$\Gamma_e(q) = \Gamma_c \tau r n^2 L(q) + L(q+\tau).$$  \hspace{1cm} (1-6)$$

An important general result that we will find is that the eigenmodes are not Hermitian orthogonal, and as a consequence, the decay of a general superposition state depends upon the spread of mode eigenfrequencies as well as the individual eigenmode decay rates.

This issue is of considerable current interest because it is now possible to create spatially coherent single exciton nuclear states $|\Psi_e(k_0)\rangle$ of the Bloch form by illuminating crystals containing resonant Mössbauer nuclei (e.g. Fe$^{57}$) with synchrotron radiation pulses.$^{3-12}$ The resulting state has $k_0 = \frac{\omega_0}{c} n_0$, where $\omega_0$ is the resonance frequency and $n_0$ is the incidence direction of the synchrotron pulse. By proper choice of $n_0$, $k_0$ will satisfy a Bragg condition and the state $|\Psi_e(k_0)\rangle$ becomes a superradiance eigenmode decaying into the $k_0$ and $k_0+\tau$ directions. For $k_0$ off Bragg, $|\Psi_e(k_0)\rangle$ is a superposition of various normal modes of different eigenfrequencies and decay rates. Nevertheless, as we'll see in Chapter V, such a state still has an initial superradiant decay into the "forward" $k_0$ direction, with $\Gamma_e$ given by Eq.(1-5) with $\tau=0$.

The purpose of this thesis is to solve for the radiative eigenmodes in a system of nuclei and to investigate superradiance and subradiance both in the eigenmode solutions and superposition states.

In Chapter II, because of its importance and general interest for our considerations, we show how spatially coherent superradiant single exciton nuclear states $|\Psi_e(k_0)\rangle$ can be created using synchrotron radiation pulses, and discuss the superradiance of such states in
the Born approximation. This chapter is a summery of the development given by Trammell and Hannon\textsuperscript{3-5}

In Chapter III, we set up the coupled equations of the radiative normal modes for a system of N classical electric dipole oscillators interacting through their radiation fields, and examine certain general sum rules for the eigenfrequencies and widths, and the nature of the orthogonality of the normal modes.

In chapter IV, we take the simplest system, two identical oscillators, and solve for the eigenmodes and complex eigenfrequencies. We examine the limits of \( kR \ll 1 \), where the coupling is in the "near field" region, and of \( kR \gg 1 \) where the coupling is in the radiation zone. We also give two alternative derivations of \( \Gamma_\pm \) by using the Poyting flux and by direct calculation of the rate at which work is done by the radiation fields. In this section we also solve for the radiative eigenmodes for two nonidentical oscillators and discuss the quantum beats which occur for a general excitation of the system.

In Chapter V, we solve for the radiative eigenmodes in a crystal of oscillators. We first solve for a 2-dimensional array. The normal mode excitations are shown to be the two dimensional Bloch waves with a phasing factor \( e^{i\mathbf{q} \cdot \mathbf{R}_j} \), and the radiation is emitted into a finite number of plane waves forming symmetric pairs of channels "above" and "below" the plane. The concept of opened and closed channels is discussed, and we discuss the strong enhancement that occurs if a mode \( \mathbf{q} \) has a channel which is "just opened".

With the 2-dimensional array as our base, we move on to systems of 2 and 3 layers, and set up the coupled equations between the different plane layers. The eigenmodes are obtained using the direct determinant approach.

A more general approach is used to solve for systems of \( M \) plane layers. Here the general coupled equations are first simplified to a 2-wave approximation, consisting of the "transmission" and "reflection" channels which make an angle \( \phi \) relative to the crystal planes. These equations give \( M \) radiative normal modes which have either a sine or cosine modulation coming in from the crystal faces, with a complex wave vector \( k' \). As illustrative examples, we solve for \( M=3 \) and \( M=10 \). The mode frequency shifts \( \Delta \omega^{(n)} \) and radiative decay widths \( \Gamma^{(n)} \) are plotted as a function of \( \phi \) for \( \omega_0 \) radiation. For \( \phi \) satisfying a Bragg angle, only one mode is superradiative with width

\[
\Gamma^{(n)} = \Gamma_c = \Gamma_c \gamma 2\pi n^2 L_\parallel(\phi), \tag{1-7}
\]
and the frequency shift $\Delta \omega^{(n)}=0$. The remaining $M-1$ modes are non-radiative. When $\phi$ is off-Bragg, all $M$ modes are radiative although most of them are subradiant with $\Gamma^{(n)} < \Gamma_Y$. The frequencies of all modes are shifted either above or below $\omega_0$, obeying the sum rule
\[
\frac{1}{M} \sum_n \Delta \omega^{(n)} = 0,
\]
while the widths obey the sum rule
\[
\frac{1}{M} \sum_n \Gamma^{(n)} = \Gamma_0.
\]

We then return to consider the single exciton states $|\Psi_e(k_0)\rangle$ created by synchrotron radiation pulses. We expand $|\Psi_e(k_0)\rangle$ as a superposition of normal mode states, solve for the subsequent time dependence of the state of system and for the emitted radiation fields, and examine the nature of the superradiant decay. We will show that if the phasing vector $k_0$ is on-Bragg with respect to the crystal planes, then the system is excited into a single superradiant eigenmode. The subsequent emission goes symmetrically into the forward and the "Bragg reflection" directions, with enhanced decay rates $\Gamma_c$ (Bragg) given by Eq. (1-7). For $k_0$ off-Bragg, more than one eigenmode are excited, and the subsequent development is superradiant nonexponential decay into the forward $k_0$-direction, with an initial decay rate $\Gamma_c$ (off-Bragg) = $\frac{1}{2} \Gamma_c$ (Bragg), consistent with the Born approximation calculation. An interesting point here is that for large $M$, $\Gamma_c$ (off-Bragg) greatly exceeds any of the eigenmode decay rates $\Gamma^{(n)}$, so that it is the nonorthogonality of the eigenmodes and the spread of eigenfrequencies which determine the superradiant decay of $|\Psi_e(k_0)\rangle$ off-Bragg.
II. The Coherent Excitation of Nuclei by Synchrotron Radiation Pulses

§2-1 Introduction.

Recently, several groups have succeeded in producing well-defined nuclear exciton states in crystals using synchrotron radiation pulses and studied some of the remarkable after effects\(^6-\)\(^{12}\). Synchrotron pulses will excite low lying nuclear levels, and in a crystal they will create nuclear exciton states which are spatially coherent superpositions of various excited state hyperfine levels of all the nuclei in the crystal. The subsequent decay is radically affected by the coherence, exhibiting both a speed-up due to the "coherent enhancement", and a quantum beat modulation of the decay rate which gives a periodic speed-up/slow-down of the rate for radiative decay into the coherent channels. This section summarizes the main ideas, following the development of Trammell and Hannon.\(^3-\)\(^5\)

§2-2 Nuclear Exciton States Induced By Synchrotron Radiation Pulses.

Synchrotron radiation from a source operating in the single-bunch mode consists of sharp pulses of about \(10^{-10}\) sec duration and about \(10^{-6}\) sec separation between pulses. Excited nuclear states of energy less than 100 keV commonly have life times in the range \(\Gamma^{-1} = 10^{-6} - 10^{-10}\) sec. If the pulse, monochromized to about 1 eV bandwidth at the nuclear transition, impinges on a small crystal containing resonant nuclei, then the electronically scattered X-rays, photo-electrons, etc., will emerge promptly during the \(10^{-10}\) sec pulse, while those processes involving nuclear excitation will be delayed a mean time \(\Gamma^{-1}\). Therefore by using a timed detector which can recover from the prompt pulse in a time short compared to \(\Gamma^{-1}\), the resonant and non-resonant events can be separated temporally. The pulse's duration and transit time to go across the crystal are short compared to \(\Gamma^{-1}\), and as a consequence, creates a collective nuclear excited state which is of the form\(^3-\)\(^5\),

\[
|\Psi_e> = \sum_{l=1}^{N} e^{i k_0 \cdot R} \sum_{l\prime} c_{n_l} |e_{l\prime} n_l> |G_0(l)> , \tag{2-1}
\]

where \(|G_0(l)\rangle\) indicates that all other nuclei \((l' \neq l)\) are in their initial ground states \(|g_{l'} m_{l'}\rangle\) and \(k_0 = k n_0 = \frac{\omega_0}{c} n_0\) where \(\omega_0\) is the natural resonance frequency of the nuclei and \(n_0\) is the
incidence direction of the pulse, $c_{nm}$ is the excitation amplitude for exciting the nucleus at $\mathbf{R}_j$ from the ground state $|g_j m_j>$ to excited state $|e_j n_j>$,

$$c_{nm} = \sqrt{\frac{ho}{N}} e^{i\mathbf{n} \cdot \mathbf{k}_0} F_{nm},$$

(2-2)

where $\rho$ is the Mössbauer probability for recoilless absorption, $\vec{\mu}$ is the dipole moment operator of the nucleus ($\vec{\mu} = d$, the electric dipole moment, for E1 and $\vec{\mu} = m$, the magnetic dipole moment, for M1 transitions) and $F$ is the Fourier component of the incident pulse field ($F = E$ for E1 and $F = B$ for M1 transitions).

Eq. (2-1) is of the Bloch form similar to Equation (1-3). In each contributing term, one nucleus is excited into one of its excited state hyperfine levels, and all the remaining nuclei stay in their initial ground states. The relative spatial phase factor is $e^{i\mathbf{n} \cdot \mathbf{k}_0}$. Hence $|\Psi_e>$ is a single exciton collective state, with one excitation distributed coherently over the entire system, with a spatial phasing wave vector $\mathbf{k}_0$.

Following the excitation, the system undergoes the process of de-excitation or decay. There are two kinds of channels which contribute to the system decay. One is the incoherent decay which comes from the processes of spatially incoherent radiative decay and of internal conversion. We will not discuss the incoherent decay in this thesis. The other channel is the coherent radiative decay in which the system decays from $|\Psi_e>$ back to the initial ground state (before excitation by the synchrotron pulse),

$$|\Psi_0> = \Pi |g_j m_j>.$$  

(2-3)

Because of "coherence enhancement", the coherent decay will dominate the decay process for sufficiently thick crystal, as we discuss below.

§2-3 Coherent Enhancement and Coherent Speedup.

The total decay width for a system of nuclei excited to $|\Psi_e>$ in the form of Eq. (2-1) is

$$\Gamma = \Gamma_c + \Gamma_r' + \Gamma_x,$$

(2-4)

where $\Gamma_c$ is the partial width for spatially coherent decay, $\Gamma_r'$ is the spatially incoherent radiative decay and $\Gamma_x$ is the partial width for internal conversion. If we assume that there
is no hyperfine splitting of the nuclei, then we can write $\Gamma_c$ as (in the Fermi Golden Rule approximation)\(^{2-5}\)

$$\Gamma_c = \Gamma_{coh}(k_0) = \Gamma_{coh} N^{-1}(4\pi)^{-1} \int d\Omega(n) \left| \sum_{l=1}^{N} e^{i(k - k_0) \cdot R_l} \right|^2$$ \hspace{1cm} (2-5)

where $k = kn = \frac{\omega_0}{c} n$ and the coherent decay width for a single nucleus $\Gamma_{coh} = \frac{2j_i + 1}{4j_0 + 2} fC\Gamma^*$ where $f$ is the Mössbauer factor, $C$ is the abundance of the nuclei isotope and $j_0$ and $j_i$ are the spins of the nuclear ground and excited state*.

Taking a look at the angular distribution of the integrand in the bracket ( ), we see that in the small angle about $n_0$ for which $|\langle n - n_0 \rangle \cdot (R_i - R_j) | << \lambda$ for all internuclear distances, we can approximately take it as $N^2$. Thus the contribution to $\Gamma_c(k_0)$ of this "forward direction" coherent radiation is $\Gamma_c(k_0) = N^2\Delta\Omega/4\pi$, where $\Delta\Omega$ is the solid angle around $n_0$ for which $|\langle n - n_0 \rangle \cdot (R_i - R_j) | << \lambda$ for all internuclear distances. If the nuclei are uniformly distributed in a prism of dimensions $L_\perp$ and $L_\parallel$ relative to the direction $n_0$, and if $L_\parallel \leq \lambda^2/\lambda$ (which is the usual situation), then we have $\Delta\Omega = (\lambda/L_\perp)^2$ and consequently,

$$\Gamma_c(k_0) = \Gamma_{coh} n\lambda^2 L_\parallel(k_0),$$ \hspace{1cm} (2-6)

where $n$ is the number of radiators per unit volume.

Eq. (2-6) shows that there is a coherent enhancement in the radiative decay rate proportional to the thickness of the crystal in the direction of $k_0$, $L_\parallel(k_0)$. The enhancement is generally very large. For $\lambda = 1\,\text{Å}$ and $n = 0.1\,\text{Å}^{-3}$, a thickness of only $\sim 300\,\text{Å}$ will double the radiative decay rate. For 14.4keV resonance in $^{57}\text{Fe}$, the coherent length $L_\parallel$ of $2\times10^{-3}\,\text{cm}$ will give us the enhancement up to $10^3 \Gamma^*$.

---

* Refer to reference (3) for a detailed derivation of $\Gamma_{coh}$. For 14.4keV resonance in $^{57}\text{Fe}$, $f=0.8$, $C=1$ (for pure $^{57}\text{Fe}$), $j_0=1/2$, $j_i=3/2$, we have $\Gamma_{coh} = 0.8 \Gamma_\gamma$.

** For $\lambda = 1\,\text{Å}$, $n = 0.1\,\text{Å}^{-3}$, $\Gamma_{coh} = \Gamma_\gamma$ and $L_\parallel(k_0) = 300\,\text{Å}$, $\Gamma_c = \Gamma_{coh} n\lambda^2 L_\parallel(k_0) = 2.3\Gamma_\gamma$. For $^{57}\text{Fe}$ 14.4keV resonance, $\lambda = 0.8\,\text{Å}$, $\Gamma_{coh} = 0.8\Gamma_\gamma$, $n = 0.1\,\text{Å}^{-3}$, $L_\parallel(k_0) = 2\times10^{-3}\,\text{cm}$, we get $\Gamma_c = 0.8\times10^3 \Gamma_\gamma$. 
In addition to the enhancement from coherent decay in the "forward direction", there will be further enhancement if \( k_0 \) satisfies a Bragg condition with one set of crystalline planes associated with the reciprocal lattice vector \( \tau \). Then constructive interference also occurs for those \( k \) lying in the small solid angle about \( k_0 + \tau \). In this case,

\[
\Gamma_c = \Gamma_c(k_0) + \Gamma_c(k_0 + \tau) = \Gamma_{coh} \pi n \lambda^2 [L_{\parallel}(k_0) + L_{\parallel}(k_0 + \tau)].
\]  

(2-7)

For a symmetric Bragg reflection from a parallel sided crystal, then \( L_{\parallel}(k_0) = L_{\parallel}(k_0 + \tau) = M d / \sin(\phi_0) \), where \( d \) is the inter-planar separation, \( M \) is the number of layers for the thin crystal sample, \( \phi_0 \) is the incident angle with respect to the planes. The coherent radiative width is then doubled relative to the off-Bragg situation, with

\[
\Gamma_c = \Gamma_{coh} 2 \pi n \lambda^2 [L_{\parallel}(k_0)] = \frac{2 \pi n \lambda^2 d}{\sin(\phi_0)} M \Gamma_{coh}.
\]  

(2-8)

With equal probability, the photon will either be emitted in the "forward" direction, or in the "Bragg reflected" direction.

It is important to note that the state \( |\Psi> \) that the synchrotron pulse excite generally is not radiative normal mode, as will be shown in Chapter V. Instead, it will be the superposition of various radiative normal modes which have different eigenfrequencies and decay rates. Most of the modes will be subradiant, some will be superradiant. Nevertheless, we will still see an initial enhanced decay rate given by Eq. (2-6). The only exceptions are when the incident pulse makes a Bragg angle with one of the crystalline planes. There, we not only get one more channel for superradiant decay, we excite only one superradiant normal mode as well. In these cases, there will be a true exponential decay with an enhanced rate as expressed in Eq. (2-8).

\section*{§2-4 Quantum Beats.}

Due to (1) the hyperfine splitting of the sublevels or (2) the energy shifts between nuclei located at different chemical or magnetic sites, there will be difference in transition frequencies for different nuclei. The frequency difference is small comparing to the resonance frequency \( \omega_0 \). So the whole effect will be a quantum-beats modulation imposed upon the radiation and decay predicted without those splitting in section §2-3.

The beats will be determined by:
\[ \Omega_B(n, m; \rho; n', m'; \rho') = \omega_{n,m}(\rho) - \omega_{n',m'}(\rho'), \]  

(2-9)

where \( \omega_{n,m}(\rho) \)'s are all the allowed nuclear hyperfine transition frequencies from all the different sites \( \rho \). The total radiation field is therefore the superposition of damped waves of different resonance frequency \( \omega_{n,m}(\rho) \), which result periodically between constructive interference and destructive interference. Consequently, the radiative decay rate will no longer be a constant as described in Eq. (2-5). Instead, the "quantum beats" modulation will turn certain channels periodically from strongly enhanced superradiant into strongly suppressed subradiant. By analyzing the beats frequencies, we will be able to study the hyperfine energy structures of the nuclei or the energy shifts for nuclei residing at different sites.

We shall not discuss much about quantum beats in this thesis. However, quantum beats are of much interest and importance, and in Chapter II, we include a section which discusses the normal modes and beats for a system of two nuclei with different natural frequencies. Readers should turn to references 3, 4, 5, 7 for more discussion on this issue.
III. System of Coupled Nuclei

Consider a system of N nuclei interacting through their radiation fields. The problem will be treated classically with each nucleus taken as a one dimensional harmonic oscillator with resonance frequency $\omega_0$ coupled to the other nuclei via the radiation fields. N such identical dipole resonators constitute a coupled system which has radiation and decay properties quite different from that of a single oscillator.

§3-1 Radiative Coupling.

(A) Dipole Radiation fields.

For an oscillating dipole,*

$$d = d_0 f(t) = d_0 \exp(-i\omega t),$$  \hspace{1cm} (3-1-1)

where $d_0=qX(0)$ is the amplitude of the oscillating dipole moment, q is the electric charge, the Hertz Vector (the Hertz Polarization Potential) is,\textsuperscript{15}

$$\Pi \equiv \frac{1}{r} d_0 f(t - \frac{r}{c}) = \frac{1}{r} d_0 \exp[i(kr - \omega t)].$$  \hspace{1cm} (3-1-2)

The dipole radiation field is,

\[
\frac{1}{2} m \ddot{x}_1 = -\frac{1}{4} m \omega_0^2 (x_1 - x_2) - \frac{1}{2} m \Gamma \dot{x}_1 + \frac{1}{2} qE,
\]

and since $x_1 = -x_2$, the resonator equation of motion is,

$$m \ddot{x}_1 = -m \omega_0^2 x_1 - m \Gamma \dot{x}_1 + qE.$$

The resonator dipole moment is, $d = \sum_i q_i x_i = q x_1 = -q x_2$.

* The idealized dipole resonator is a system of two charges, $q_1 = -q_2 = -q/2$, with equal masses $m_1 = m_2 = (1/2)m$ and with a linear restoring force $-\frac{1}{4} m \omega_0^2 (x_1 - x_2)$ coupling the charges. Under an external field $E$ which is uniform across the dipole system, the charges will move with antisymmetric motion $x_2 = -x_1$. The force equation for $x_1$ is,
\[ E = \nabla x (\nabla x \Pi) = \nabla x \{ \nabla x \frac{1}{r} d_0 \text{exp}[i(\omega t - \omega t)] \}, \quad (3-1-3) \]

where \( k = \frac{\omega}{c} \) is the wave number of the radiation, \( r \) is the distance between the source and observation point, and \( \omega \) is the frequency of the oscillation. Eq. (3-1-3) gives (see Appendix I),

\[ E = -k^2 [n x (n x d_0)] \frac{1}{r} e^{i(\omega t - \omega t)} + [3n (n \cdot d_0) - d_0] \frac{1}{r^2} \left( \frac{ik}{r^2} \right) e^{i(\omega t - \omega t)}, \quad (3-1-4) \]

where \( n = \frac{r}{r} \) is the direction from the oscillator to the observation point.

Eqs. (3-1-3) and (3-1-4) are valid under the "small source approximation" which requires that the dimension of the oscillator \( |X| \) be much smaller than the distance \( r \) and the wavelength of the radiation \( \lambda \). So long as this is satisfied, the results can be applied to the static \( (r \ll \lambda) \), intermediate and the radiation \( (r \gg \lambda) \) zones. Our particular interest is in "Mössbauer radiators", for which \( \lambda \sim 1 \text{Å} \) and \( |X| \sim 10^{-4} \text{Å} \), so the condition \( |X| \ll \lambda \) is well satisfied. The internuclear separations are also \( \geq 1 \text{Å} \), but Eqs. (3-1-3) and (3-1-4) would be good approximation even for \( r \sim 10^{-3} \text{Å} \).

An isolated oscillating dipole moment will lose energy by emission of electromagnetic radiation resulting in an exponential damping imposed upon its harmonic oscillation amplitude. In addition to the radiative damping (damping rate \( \Gamma = \frac{2q^2 \omega^2}{3mc^3} \)), there will also be non-radiative friction forces such as internal conversion (damping rate \( \Gamma_{\alpha} \)), giving a total damping rate \( \Gamma = \Gamma_{\gamma} + \Gamma_{\alpha} \), and the oscillation will decay as \( X(t) = X(t_0) e^{-i(\omega' - i\Gamma/2)t} \), where \( \omega' \) is a purely real quantity and \( I(t) = 1 \) for \( t \geq 0 \) and \( I(t) = 0 \) for \( t < 0 \). So long as the damping rate is much less than the oscillation frequency, i.e., if \( \Gamma \ll \omega' \), then to good approximation (to terms of order \( \Gamma/\omega' \)), the emitted radiation field is still given by Eqs. (3-1-3) and (3-1-4) with \( e^{-i\omega' t'} \rightarrow I(t') e^{-i(\omega' - i\Gamma/2)t'} \), where \( t' = t - r/c \) is the retarded time.* For

* The damped oscillator motion

\[ X(t) = X I(t) e^{-i(\omega' - i\Gamma/2)t} \]

can be Fourier analyzed as,
Mössbauer nuclei, we typically have $\Gamma/\omega' \sim 10^{-12}$, so this will be a very good approximation.

(B) Radiative Normal Modes.

For a system of $N$ coupled oscillators, the system will not be in a true exponentially decaying state unless it is in one of the radiative normal modes. In these normal modes, all the oscillators undergo oscillations with a unique frequency and damping rate, which we can put together as a complex frequency, $\omega$. The general motion of the system will be a superposition of these normal modes. In the following discussion and the rest of the chapters, we will try to determine these normal modes.

The system we will consider will be a set of identical one-dimensional oscillators undergoing harmonic oscillation along the $\hat{x}$ direction. Assume the system is in one of its radiative normal modes. By definition of "radiative normal modes", all oscillators are undergoing oscillation with one single complex frequency $\omega$. The initial oscillation amplitudes are $X_i$ ($i = 1, 2, ..., N$), so the amplitude of the $i$th oscillating dipole moment is $d_i^{(0)} = q X_i \hat{x}$. The problem will be to determine the complex normal mode frequency $\omega^{(k)}$ and the configuration $X_i^{(k)}$ of the various normal modes.

In order to do that, we must first examine the radiative interaction (coupling) between the two oscillators and set up the coupled equation of motion.

\[
X(t) = X_0 \int_{-\infty}^{+\infty} d\omega f(\omega) e^{-i\omega t},
\]

where

\[
f(\omega) = \frac{i}{2\pi} \left( \omega - \omega' + i\frac{\Gamma}{2} \right)^{-1}.
\]

For each harmonic component $\omega$, the radiation field is given by Eq. (3-1-3), so the total $E$ is the Fourier transform of Eq. (3-1-3), with weighting amplitude $f(\omega)$, giving,

\[
E = \nabla_x \left\{ \nabla_t \int \frac{1}{t} d_0 e^{-i(\omega - i\Gamma/2)(t-r/c)} I(t-r/c) \right\}.
\]

Expanding and dropping terms of order $(\Gamma/\omega')$ which do not appear as exponential phase factors, $E$ is then given by Eq. (3-1-4) with,

\[
e^{-i\omega(t-r/c)} \rightarrow I(t') e^{-i(\omega' - i\Gamma/2)(t-r/c)}.
\]
(C) Interaction Between Two Oscillators.

Let's first look at the interaction between two oscillators which are undergoing radiative normal mode motion. At the position $\mathbf{R}_j$ of oscillator $j$, the field due to oscillator $i$ (at $\mathbf{R}_i$) is,

$$ E_{ji} = q \nabla_j \times \left\{ \nabla_j \times \frac{\exp[-i(k|\mathbf{R}_j - \mathbf{R}_i| - \omega t)]}{|\mathbf{R}_j - \mathbf{R}_i|} \hat{x} \right\} $$

$$ = (-k^2 [n_{ji} x(n_{ji} x \hat{x})] \frac{1}{r_{ji}} + [3n_{ji}(n_{ji} \hat{x} - \hat{x})] (\frac{1}{r_{ji}^3} - \frac{ik}{r_{ji}^2}) qX_i \exp[i(\mathbf{k} \cdot \mathbf{r}_{ji} - \omega t)]) (3-1-5) $$

where $n_{ji} = \frac{\mathbf{R}_j - \mathbf{R}_i}{|\mathbf{R}_j - \mathbf{R}_i|}, r_{ji} = |\mathbf{R}_j - \mathbf{R}_i|$. Similarly, the field from oscillator $j$ to $i$ can be written by interchanging $i$ and $j$ in (3-1-5). Since all oscillators are constrained to move in the $\hat{x}$ direction, only the $\hat{x}$ component of (3-1-5) will affect the motion of oscillator $j$. Thus the force acting upon oscillator $j$ due to the radiation field from oscillator $i$ is

$$ F_{ji} = q \hat{x} \cdot E_{ji} $$

$$ = \left( [1 - (n_{ji} \hat{x})^2] + [3(n_{ji} \hat{x})^2 - 1] \left[ \frac{1}{(kr_{ji})^2} - \frac{i}{kr_{ji}} \right] \right) q^2 \frac{k^2}{r_{ji}} \exp[i(\mathbf{k} \cdot \mathbf{r}_{ji} - \omega t)] X_i $$

$$ = \left( [1 - (n_{ji} \hat{x})^2] + [3(n_{ji} \hat{x})^2 - 1] \left[ \frac{1}{(kr_{ji})^2} - \frac{i}{kr_{ji}} \right] \right) X_i \exp[i(\mathbf{k} \cdot \mathbf{r}_{ji} - \omega t)] \frac{3m_{\omega}}{2kr_{ji}} \Gamma \gamma (3-1-6) $$

We call,

$$ g_{ji} = \frac{F_{ji}}{X_i e^{-i\omega t}} $$

$$ = q^2 \hat{x} \cdot \left\{ \nabla_j \times \frac{\exp(-ik|\mathbf{R}_j - \mathbf{R}_i|)}{|\mathbf{R}_j - \mathbf{R}_i|} \hat{x} \right\} $$

$$ = \left( [1 - (n_{ji} \hat{x})^2] + [3(n_{ji} \hat{x})^2 - 1] \left[ \frac{1}{(kr_{ji})^2} - \frac{i}{kr_{ji}} \right] \right) \exp(ikr_{ji}) \frac{3m_{\omega}}{2kr_{ji}} \Gamma \gamma (3-1-7) $$

as our interaction coefficient. Notice $n_{ji} = -n_{ij}$ and $r_{ji} = r_{ij}$, so $g_{ji}$ bears the nice property of symmetry: $g_{ji} = g_{ij}$. We now can write the interaction from oscillator $i$ to $j$ as:

$$ F_{ji} = g_{ji} X_i \exp(-i\omega t) = g_{ji} X_i(t), (3-1-8) $$
where \( X_j(t) = X_j \exp(-i \omega t) \) is the oscillator's instantaneous complex displacement.

(D) Normal Mode Equation of Motion.

Now consider a system of \( N \) oscillators undergoing radiative normal mode motion. Oscillator \( j \) is influenced by all other oscillators in the form of Eq. (3-1-8), as well as by its own linear restoring force, \(-m \omega_0^2 X_j(t)\), and the radiative and nonradiative damping force, \(-m \Gamma X_j(t)\), where \( m \) is the mass of the oscillator, \( \omega_0 \) is the natural resonance frequency of the oscillator, and \( \Gamma = \gamma + \alpha \). The equation of motion for the \( j \)th oscillator is then,

\[
mX_j(t) + m\omega_0^2 X_j(t) + m\Gamma X_j(t) = \sum_{i \neq j} g_{ji} X_i(t) \quad (j=1, 2, \ldots, N),
\]

(3-1-9)

Since all the oscillators are undergoing oscillations with a unique complex frequency \( \omega \) (as the definition of "normal mode" requires), we can rewrite Eq. (3-1-9) as,

\[
m(\omega_0^2 - \omega^2 - i \omega \Gamma) X_j = \sum_{i \neq j} g_{ji} X_i \quad (j=1, 2, \ldots, N)
\]

(3-1-10)

(E) Linearized Normal Mode Equation of Motion.

The self-decay rate \( \Gamma \) is very small compared to the natural resonance frequency \( \omega_0 \). Excited nuclear states of energy \( \leq 100 \) keV commonly have life times in the range \( \Gamma^{-1} = 10^{-6} - 10^{-10} \) sec which makes \( \Gamma / \omega_0 = 10^{-13} - 10^{-9} \). On the other hand, \( g_{ji} \) on the right hand side is on the order of \( \left(1 + \frac{1}{(kr_{ji})^2} + \frac{1}{kr_{ji}}\right) \frac{3m\omega}{2kr_{ji}} \Gamma \gamma \), which in our case (\( \lambda \sim 10^{-1} - 10^{-2} \) A and \( r_{ji} \geq 1 \) A) equals to approximately \( 10^{-1} m\omega \Gamma \gamma \). Thus from Equation (3-1-10), we expect the solution to frequency \( \omega \) as \( \omega = \omega_0 + \Delta \), with \( \Delta \sim \Gamma \ll \omega_0 \). If we neglect terms of order \( \frac{\Gamma}{\omega_0} \) and higher compared to unity, the left hand side of Eq. (3-1-10) can be written as,

\[
LHS = m(\omega_0^2 - \omega^2 - i \omega \Gamma) X_j
\]

\[
= -2i \omega_0 [\Delta(1 + \frac{\Delta}{2\omega_0}) + i \frac{\Gamma}{2\omega_0} (1 + \frac{\Delta}{\omega_0})] X_j
\]
\[ (\omega - \omega i) X_j = \frac{q^2}{2m\omega_0} \sum_{i \neq j} \left\{ \nabla_j \times \nabla_j \times \frac{\exp(-ik_0|\mathbf{r}_j - \mathbf{R}_j|)}{|\mathbf{r}_j - \mathbf{R}_j|} \right\} X_i \quad (3-1-11) \]

\[ = \sum_{i \neq j} \left\{ [1 - (n_{j i} \Delta \lambda)^2] + [3(n_{j i} \Delta \lambda)^2 - 1] \left( \frac{1}{(k_0 r_{j i})^2} - \frac{i}{k_0 r_{j i}} \right) \right\} X_i \exp(ik_0 r_{j i}) \left( \frac{3}{4k_0^2 r_{j i}^2} \right) \Gamma \quad (3-1-12) \]

For the sake of simplicity and clarity, we rewrite Eq. (3-1-12) in the form,

\[ (\omega - \omega i) X_j = \frac{\Gamma}{2} \sum_{i \neq j} \kappa_{j i} \left( \frac{e^{ik_0 r_{j i}}}{k_0 r_{j i}} \right) X_i \quad (3-1-13) \]

where

\[ \kappa_{j i} \left( \frac{e^{ik_0 r_{j i}}}{k_0 r_{j i}} \right) = \frac{3}{2k_0} \left\{ \nabla_j \times \nabla_j \times \frac{\exp(ik_0|\mathbf{r}_j - \mathbf{r}_i|)}{|\mathbf{r}_j - \mathbf{r}_i|} \right\} \quad (3-1-14) \]

\[ = \frac{3}{2} \left\{ [1 - (n_{j i} \Delta \lambda)^2] + [3(n_{j i} \Delta \lambda)^2 - 1] \left( \frac{1}{(k_0 r_{j i})^2} - \frac{i}{k_0 r_{j i}} \right) \right\} \left( \frac{e^{ik_0 r_{j i}}}{k_0 r_{j i}} \right). \quad (3-1-15) \]

If the distance between the nearest nuclei \( r_{j i} \gg \lambda \), we have,

* Replacing \( k \) with \( k_0 \) in the exponential \( \exp(ikr) \) will bring in phase errors, \( \delta \theta = kr - k_0 r_i = \frac{\Delta}{c} r_i < \frac{\Delta}{c} L \), where \( L \) is the dimension of the crystal sample. These phase errors can be ignored if \( (\Delta/c)L << \pi \). As an example, even for a large normal mode shift \( \Delta = 100 \Gamma \) with \( \Gamma = 5 \times 10^{-9} \text{eV} \) (appropriate for the 14.4keV Fe\textsuperscript{57} transition), then \( (\Delta/c)L << \pi \) only requires that the sample sizes be restricted to \( L << 10^2 \text{cm} \).
\[ \kappa_{ji} = \frac{3}{2} \left[ 1 - (n_{ji} \hat{X})^2 \right]. \] (3-1-16)

Or if \( r_{ji} \ll \lambda \), we have,

\[ \kappa_{ji} = \frac{3}{2} \left[ 3(n_{ji} \hat{X})^2 - 1 \right] \left[ \frac{1}{(k_0 r_{ji})^2} - \frac{i}{k_0 r_{ji}} \right]. \] (3-1-17)

§3-2 Solving for the Normal Modes:

(A) Normal Modes and Eigenfrequencies

We can see from (3-1-13) that we have \( N \) linear equations for \( X_j \), \( (j=1, 2, 3, ..., N) \). Putting the equation group into matrix form, we have,

\[ \vec{H} \vec{X} = \omega \vec{X}, \] (3-2-1)

where \( \vec{H} \) is an \( N \times N \) matrix with the elements \( h_{ii} = \omega_0 - i \frac{\Gamma}{2} \), \( h_{ij} (i \neq j) = -\frac{\Gamma}{2} \frac{\kappa_{ij} e^{ik_j x_j}}{k_0 r_{ij}} \), and

\[ \vec{X} = \begin{pmatrix} X_1 \\ X_2 \\ \vdots \\ X_N \end{pmatrix} \]

is a \( N \) component vector with jth component being the oscillation amplitude of the jth oscillator.

We note that Eq. (3-2-1) is an "effective Hamiltonian" equation for the eigenfrequencies and configurations of the radiative semi-stationary states. It will be shown in subsequent work that a QED treatment leads to essentially the same equation for the semi-stationary states of a system of two level nuclei interacting via the radiation field, in which case
\[
\mathbf{X} \rightarrow |\psi> = \sum_{l=1}^{N} C_l |e_l> |G_0(t)>
\]

In order to get the normal modes and the corresponding eigenfrequencies, we let,

\[
\text{determinant} [\tilde{\mathbf{H}} - \omega 1] = 0, \tag{3-2-2}
\]

where $1$ is the unity matrix. From Eq. (3-2-2), we can solve for the $\omega$'s for various normal modes, $\omega^{(k)} = \omega^{(k)} - i \Gamma^{(k)}/2$, $k=1,2,\ldots,N$. $\Delta\omega^{(k)} = \omega^{(k)} - \omega_0$ gives us the frequency shift of the $k$th normal mode from the resonance frequency of a single oscillator while $\Gamma^{(k)}$ gives the mode's decay rate which will generally be quite different from the decay rate $\Gamma$ of a single oscillator.

(B) Orthogonality and Sum Rules.

We see that the matrix $\tilde{\mathbf{H}}$ is transpose symmetric instead of being Hermitian ($h_{ij} = h_{ji}$). In turn, the normal modes that we get from (3-2-1) will be transpose orthogonal instead of Hermitian orthogonal,*

* This fact can be proved as following.

Suppose $\mathbf{X}^{(k)}$ and $\mathbf{X}^{(i)}$ are two eigenvectors of $\tilde{\mathbf{H}}$ corresponding to eigenvalues $\omega^{(k)}$ and $\omega^{(i)}$ respectively,

\[
\tilde{\mathbf{H}} \mathbf{X}^{(k)} = \omega^{(k)} \mathbf{X}^{(k)} \tag{1}
\]

\[
\tilde{\mathbf{H}} \mathbf{X}^{(i)} = \omega^{(i)} \mathbf{X}^{(i)} \tag{2}
\]

From (2), we have

\[
\mathbf{X}^{(i)^T} \tilde{\mathbf{H}} = \omega^{(i)} \mathbf{X}^{(i)^T},
\]

or because we have $\tilde{\mathbf{H}}^T = \tilde{\mathbf{H}}$,

\[
\mathbf{X}^{(i)^T} \tilde{\mathbf{H}} = \omega^{(i)} \mathbf{X}^{(i)^T}. \tag{3}
\]

Taking $\mathbf{X}^{(i)^T} - (3) \mathbf{X}^{(k)}$ gives us

\[
[X^{(i)^T}\tilde{\mathbf{H}} \mathbf{X}^{(k)}] - [X^{(i)^T}\tilde{\mathbf{H}} \mathbf{X}^{(k)}] = (\omega^{(k)} - \omega^{(i)}) \mathbf{X}^{(i)^T} \mathbf{X}^{(k)}. \tag{4}
\]

The left hand side of (4) is zero, so we have
\[(X^{(k)})^T(X^{(l)}) = \delta_{kl}\]  

(3-2-3)

Eq. (3-2-2) is an Nth order equation in \(\omega\), from which we get N eigenvalues \(\omega^{(k)} (k = 1,2,\ldots,N)\). It means that \(\tilde{\mathbf{H}}\) can be diagonalized through a similarity transformation** into \([\omega^{(k)} \delta_{kl}]\). Since similarity transformations do not alter the "trace", we have

\[\text{Trace}\{[\omega^{(k)} \delta_{kl}]\} = \text{Trace}\{\tilde{\mathbf{H}}\},\]

which gives

\[\sum_{k=1}^{N} \omega^{(k)} = N(\omega_0^{-1} \Gamma).\]  

(3-2-4)

Taking the real and imaginary parts of Eq. (3-2-4), gives the "Sum Rules" for the eigenfrequency shifts and normal mode decay rates:

\[\sum_{k} (\omega^{(k)} - \omega_0) = 0\]  

(3-2-5)

\[X^{(l)}X^{(k)} = 0 \quad \text{(for } \omega^{(k)} \neq \omega^{(l)}),\]

which proves that the eigenvectors of a transpose symmetric matrix \(\tilde{\mathbf{H}}\) with different eigenvalues are transpose orthogonal to each other.

** The transformation matrix \(\mathbf{U}\) is obtained from the eigenvector \(X^{(k)}\) in the usual manner,

\[\mathbf{U} = \begin{pmatrix}
X^{(1)}^T \\
\vdots \\
X^{(N)}^T
\end{pmatrix} = \begin{pmatrix}
X^{(1)}_1 & \cdots & X^{(1)}_N \\
\vdots & \ddots & \vdots \\
X^{(N)}_1 & \cdots & X^{(N)}_N
\end{pmatrix} \]

Because of the orthogonality condition Eq. (3-2-3), \(U^{-1} = U^T\). Since \(\tilde{\mathbf{H}} X^{(k)} = \omega^{(k)} X^{(k)}\), it immediately follows that

\[[U \tilde{\mathbf{H}} U^T]_{ij} = \omega^{(i)} \delta_{ij}.\]
and

\[
\frac{1}{N} \sum_k \Gamma^{(k)} = \Gamma.
\]

(3-2-6)

We see from (3-2-5) that for some modes the frequencies increase, while for others the frequencies decrease, such that the frequency shifts average to zero.

Eq. (3-2-6) shows that the total decay rates of the various normal modes average to the decay rate \( \Gamma = \Gamma_{\gamma} + \Gamma_{\alpha} \) of a single oscillator, where \( \Gamma_{\gamma} \) is the radiative decay rate for a single oscillator and \( \Gamma_{\alpha} \) is the rate for incoherent and non-radiative decay. The total decay rate for the \( k \)th mode can be expressed as \( \Gamma^{(k)} = \Gamma^{(k)}_{c} + \Gamma^{(k)}_{\alpha} \) where \( \Gamma^{(k)}_{c} \) is the rate for coherent radiative decay and \( \Gamma^{(k)}_{\alpha} \) is the rate for nonradiative decay. Because there is no spatial coherence involved in the incoherent non-radiative decay, \( \Gamma^{(k)}_{\alpha} = \Gamma_{\alpha} \) for all modes.

Thus Eq. (3-2-6) can be re-written as

\[
\frac{1}{N} \sum_k \Gamma^{(k)}_{c} = \Gamma_{\gamma}
\]

(3-2-7)

* The radiative decay rate of a set of classical phased oscillators located at fixed positions is necessarily spatially coherent. The emitted radiation field, which carries off the system energy, is the superposition of the fields emitted by all the various oscillators.

However, in the quantum case, there will generally be spatially incoherent radiative decay, \( \Gamma_{\gamma}' \), in addition to the spatially coherent decay, \( \Gamma^{(k)}_{c} \). For example, referring to the discussion in Chapter II, if a phased excited state \( |\psi_{c}(k,0)\rangle \) decays to a final state \( |\psi_f\rangle \) which is different from \( |\psi_{0}\rangle \) by having a "spin flip" at the \( l \)th nucleus, i.e., \( m_{l}(I) \neq m_{0}(I) \), then the scattering (and decay) definitely occurred at the \( l \)th site. Because the \( l \)th site is "tagged", there is no spatial coherence in the emission. As a consequence, for the quantum case, the sum rule should be

\[
\frac{1}{N} \sum_k \Gamma^{(k)}_{c} = \Gamma_{\gamma}' - \Gamma_{\gamma} = \Gamma_{\text{coh}}'
\]
Some modes will have $\Gamma_{c}^{(k)} < \Gamma_{\gamma}$ while for some $\Gamma_{c}^{(k)} > \Gamma_{\gamma}$, such that the coherent radiative decay rate averages to $\Gamma_{\gamma}$. As we will see in Chapter V, it is possible to have "superradiant" modes, for which $\Gamma_{c}^{(k)} \gg \Gamma_{\gamma}$, and subradiant modes, for which $\Gamma_{c}^{(k)} \ll \Gamma_{\gamma}$.

§3-3 Motion after an Arbitrary Initial Excitation.

Let us suppose we have an initial excitation that gives such an initial condition $X(0)$ for the system,

$$X(0) = \begin{pmatrix} X_1(0) \\ X_2(0) \\ \vdots \\ \vdots \\ X_N(0) \end{pmatrix} \quad (3-3-1)$$

where $X_j(0)$ represents the initial complex displacement for the jth oscillator.* The excitation is a superposition of the N normal modes of the system,

$$X(0) = \sum_{k=1}^{N} A_k X^{(k)}, \quad (3-3-2)$$

where $X^{(k)}$ is the kth normal mode obtained from Eq. (3-2-1) and Eq. (3-2-2), and $A_k$ is the complex expansion coefficient. According to the orthogonality condition Eq. (3-2-3), we have,

$$A_k = [X^{(k)}]^T \cdot X(0). \quad (3-3-3)$$

The general motion at any $t > 0$ is then,

---

* $X_j(0)$ specifies both the initial displacement and the initial velocity of the jth oscillator. The initial displacement is $\text{Re}[X_j(0)]$. The initial velocity is, to very good approximation, $\omega_0 \text{Im}[X_j(0)]$, since all relevant frequencies differ from $\omega_0$ by only terms of order $\Gamma/\omega_0 \ll 1$. 

\[ X(t) = \sum_{k=1}^{N} A_k X_j^{(k)} \exp[-i\omega^{(k)}t], \quad (3-3-4) \]

where, directly from Eq. (3-2-2),

\[ \omega^{(k)} = \omega^{(k)} - i \frac{1}{2} (\Gamma_\alpha^{(k)} + \Gamma_c^{(k)}) \quad (3-3-5) \]

is the complex eigenfrequency of the kth normal mode.

The complex displacement for the jth oscillator is simply the jth component of \( X(t) \). We therefore have,

\[ X_j(t) = \sum_{k=1}^{N} A_k X_j^{(k)} \exp[-i\omega^{(k)}t] \quad (3-3-6) \]

representing the oscillation of the jth oscillator. We use the notation \( A_k X_j^{(k)} = |A_k X_j^{(k)}| \exp(i\alpha_j^{(k)}) \) so that we can put Eq. (3-3-6) in the form of,

\[ X_j(t) = \sum_{k=1}^{N} |A_k X_j^{(k)}| \exp[-i(\omega^{(k)}t + \alpha_j^{(k)})], \quad (3-3-7) \]

where \( \alpha_j^{(k)} \) is the phase angle of \( A_k X_j^{(k)} \).

§ 3-4. Radiation Fields from the System

The radiation field from a single one-dimensional oscillator moving with frequency \( \omega \) is given by Eqs. (3-1-3) and (3-1-4). Replacing \( q \) by \( q X_j^{(k)} \), \( \omega \) by \( \omega^{(k)} = [\omega^{(k)} - i \frac{1}{2} (\Gamma_\alpha^{(k)} + \Gamma_c^{(k)})] \) and \( r \) by \( |R-R_j| \) we get the radiation field from the jth oscillator (located at \( R_j \)) in the system undergoing the kth normal mode motion,

\[ E^{(k)}_j = q \nabla \chi \left\{ \nabla \chi \frac{1}{|R-R_j|} X_j^{(k)} \exp[-i\omega^{(k)}(t - \frac{|R-R_j|}{c})] \right\}. \quad (3-4-1) \]
From Eq. (3-4-1), we get the radiation field from the system as the superposition of the fields from all the oscillators in the system,

\[
E^{(k)} = q \sum_{j=1}^{N} \nabla \times \left( \nabla \times \frac{1}{|\mathbf{R} - \mathbf{R}_j|} X_j^{(k)} \right) \exp[-i \omega^{(k)}(t - \frac{|\mathbf{R} - \mathbf{R}_j|}{c})].
\]  

(3-4-2)

Similarly, we get the magnetic field,

\[
B^{(k)} = -i \frac{q \omega^{(k)}}{c} \sum_{j=1}^{N} \nabla \times \left( \frac{X_j^{(k)}}{|\mathbf{R} - \mathbf{R}_j|} \right) \exp[-i \omega^{(k)}(t - \frac{|\mathbf{R} - \mathbf{R}_j|}{c})].
\]  

(3-4-3)

Eqs. (3-4-2) and (3-4-3) in turn can be expanded into the form of Eq. (3-1-4). Generally, observation points are far enough from each oscillator so that the "radiation zone" approximation can be used. Furthermore, if we assume that the dimension of the system is much smaller than the distance between the system and the observation point, we have,

\[
E^{(k)} = q k_0^2 \frac{[\hat{\mathbf{X}} - (\mathbf{n} \cdot \hat{\mathbf{X}}) \mathbf{n}]}{r} \exp[-i \omega^{(k)}(t - \frac{r}{c})] \sum_{j=1}^{N} X_j^{(k)} \exp[-i \mathbf{k} \cdot \mathbf{R}_j],
\]  

(3-4-4)

and,

\[
B^{(k)} = q k_0^2 \frac{\mathbf{n} \times \hat{\mathbf{X}}}{r} \exp[-i \omega^{(k)}(t - \frac{r}{c})] \sum_{j=1}^{N} X_j^{(k)} \exp[-i \mathbf{k} \cdot \mathbf{R}_j],
\]  

(3-4-5)

where \( \mathbf{k} = k_0 \mathbf{n} \). We have used \( k_0 \) in place of \( k \) outside of the exponential functions, and we have assumed \( \exp[-i(\omega^{(k)} - \omega_0) \mathbf{n} \cdot \mathbf{R}_j] \approx 1 \) for the reason discussed in the footnote on Page 15.

The general motion \( \mathbf{X}(t) \) [given by Eq. (3-3-4)] is the superposition of various normal modes. Consequently the radiation field emitted by a system of \( N \) oscillators will be the superposition of the fields \( E^{(k)} \) from \( N \) normal modes,
\[ E = \sum_{k=1}^{N} A_k E^{(k)}, \quad (3-4-6) \]

and,

\[ B = \sum_{k=1}^{N} A_k B^{(k)}, \quad (3-4-7) \]

where \( A_k \) is given by Eq. (3-3-3) and \( E^{(k)}, B^{(k)} \) are given by Eqs. (3-4-2), (3-4-3), or for far fields, by Eqs (3-4-4), (3-4-5).

§ 3-5. Mechanical Energy

(A) Kinetic Energy of the jth Oscillator

For the jth oscillator, the kinetic energy is,

\[ K_j(t) = \frac{1}{2} m \left| \text{Re} \dot{X}_j(t) \right|^2, \quad (3-5-1) \]

where

\[ \dot{X}_j(t) = -i \sum_{k=1}^{N} \omega^{(k)} |A_k X^{(k)}_j| \exp[-i(\omega^{(k)} t - \alpha^{(k)}_j)]. \quad (3-5-2) \]

Now, as we will see later in specific examples, the maximum shift in normal mode resonance frequencies, \( \Delta \omega^{(k)} = |\omega^{(k)} - \omega_0| \), and the maximum coherent decay rate, \( \Gamma^{(k)}_c \), are typically no more than \((10^2-10^3)\Gamma_f=(10-10^2)\Gamma\). Taking Fe\(^{57}\) as an example, we have \( \Gamma^{-1} \) on the order of \(10^2\)ns and \( \omega_0^{-1} \) on the order of \(10^{-10}\)ns, which makes \( \frac{\Delta \omega^{(k)}}{\omega_0} \) and \( \frac{\Gamma^{(k)}_c}{\omega_0} \) on the order of \(10^{-11}-10^{-10}<<1\). Therefore we can replace the complex \( \omega^{(k)} \) with \( \omega_0 \) in Eq. (3-5-2) as long as \( \omega^{(k)} \) does not appear in the exponential function \( \exp[-i(\omega^{(k)} t)] \). That gives us,
\[ \dot{X}_j(t) = -i \omega_0 \sum_{k=1}^{N} |A_k X_j^{(k)}| \exp[-i(\omega^{(k)} t - \alpha_j^{(k)})] = -i \omega_0 X_j(t). \]  \hspace{1cm} (3-5-3)

Now Eq. (3-5-1) becomes,

\[ K_j(t) = \frac{1}{2} m \omega_0^2 [\text{Im} X_j(t)]^2 \]

\[ = \frac{1}{2} m \omega_0^2 \left\{ \sum_{k=1}^{N} |A_k X_j^{(k)}|^2 \exp\left[-\frac{1}{2} \left( \Gamma_{c}^{(k)} + \Gamma_{\alpha} \right) t \right] \sin(\omega^{(k)} t - \alpha_j^{(k)}) \right\}^2 \]

\[ = \frac{1}{2} m \omega_0^2 \left\{ \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}||A_l X_j^{(l)}| \exp\left[-\frac{1}{2} \left( \Gamma_{c}^{(k)} + \Gamma_{c}^{(l)} + 2\Gamma_{\alpha} \right) t \right] \sin(\omega^{(k)} t - \alpha_j^{(k)}) \sin(\omega^{(l)} t - \alpha_j^{(l)}) \right\} \]

\[ = \frac{1}{4} m \omega_0^2 \left\{ \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}||A_l X_j^{(l)}| \exp\left[-\frac{1}{2} \left( \Gamma_{c}^{(k)} + \Gamma_{c}^{(l)} + 2\Gamma_{\alpha} \right) t \right] \cos[(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})] - \cos[(\omega^{(k)} + \omega^{(l)}) t - (\alpha_j^{(k)} + \alpha_j^{(l)})] \right\} \]  \hspace{1cm} (3-5-4)

The time average is now performed on the above equation in an time interval \( T \), such that

\[ \omega_0^{-1} << T << \max[\Delta \omega^{(k)}]^{-1}. \]  \hspace{1cm} (3-5-5)

Typically, we have \([\Delta \omega^{(k)}]^{-1} \sim 1-100\text{ns}\) and \(\omega_0^{-1} \sim 10^{-10}\text{ns}\). For example, if we choose

\[ T = 10^{-4}\text{ns}, \]  which is well within the range set by Eq. (3-5-5), carrying out the averaging process \( \frac{1}{T} \int_t^{t+T} K_j(t) \), we get,

\[ <K_j(t)> = \frac{1}{4} m \omega_0^2 \left\{ \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}||A_l X_j^{(l)}| \exp\left[-\frac{1}{2} \left( \Gamma_{c}^{(k)} + \Gamma_{c}^{(l)} + 2\Gamma_{\alpha} \right) t \right] \cos[(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})] \right\} \]

\[ \int_t^{t+T} \exp\left[-\frac{1}{2} \left( \Gamma_{c}^{(k)} + \Gamma_{c}^{(l)} + 2\Gamma_{\alpha} \right) t \right] \cos[(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})] dt \]
\[ \frac{1}{\Gamma_0} \int_t^{t+T} \exp [- \frac{1}{2} (\Gamma_c^{(k)} + \Gamma_c^{(l)} + 2 \Gamma_\alpha) t] \cos \left[ (\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)}) \right] dt \} \] (3-5-6)

Now because \( \Gamma_c^{(k)} \ll 10^2 \Gamma \ll 1/T \), the decay factor \( \exp [- \frac{1}{2} (\Gamma_c^{(k)} + \Gamma_c^{(l)} + 2 \Gamma_\alpha) t] \) remains practically constant during the time period \( t \) to \( t+T \) and therefore can be pulled out of the integral. Similarly, \( \cos \left[ (\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)}) \right] \) remains practically constant during that same period of time because \( (\omega^{(k)} - \omega^{(l)}) \ll 10^2 \Gamma \ll 1/T \). On the other hand, \( (\omega^{(k)} + \omega^{(l)}) = 2 \omega_0 \gg 1/T \), so the contribution from the second integral is practically zero. Thus,

\[ \langle K_j(t) \rangle = \frac{1}{4} m \alpha_0^2 \exp(-\Gamma_\alpha t) \]

\[ \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}|^2 |A_l X_j^{(l)}|^2 \exp [- \frac{1}{2} (\Gamma_c^{(k)} + \Gamma_c^{(l)} + 2 \Gamma_\alpha) t] \cos \left[ (\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)}) \right] \].

(3-5-7)

We see that the final outcome of the average does not depend on \( T \), as long as \( T \) satisfies condition set by Eq. (3-5-5).

We also note,

\[ |X_j(t)|^2 = \sum_{k=1}^{N} |A_k X_j^{(k)}|^2 \exp [-i(\omega^{(k)} t - \alpha_j^{(k)})] \]

\[ = \exp(-\Gamma_\alpha t) \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}| |A_l X_j^{(l)}| \exp [- \frac{1}{2} (\Gamma_c^{(k)} + \Gamma_c^{(l)} + 2 \Gamma_\alpha) t] \exp [-i(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})] \]
\[ = \exp(-\Gamma_\alpha t) \sum_{k=1}^N \sum_{l=1}^N |A_k X_j^{(k)}| |A_l X_j^{(l)}| \exp[-\frac{1}{2}(\Gamma_c^{(k)} + \Gamma_c^{(l)}) t] \]

\[ \{ \cos[(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})] - i \sin[(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})] \}. \] (3-5-8)

Because \( \sin(x) \) is an odd equation, the second term sums up to zero. So we have,

\[ |X_j(t)|^2 = \exp(-\Gamma_\alpha t) \]

\[ \sum_{k=1}^N \sum_{l=1}^N |A_k X_j^{(k)}| |A_l X_j^{(l)}| \exp[-\frac{1}{2}(\Gamma_c^{(k)} + \Gamma_c^{(l)}) t] \cos[(\omega^{(k)} - \omega^{(l)}) t - (\alpha_j^{(k)} - \alpha_j^{(l)})]. \] (3-5-9)

Combining Eqs. (3-5-7) and (3-5-9), the time averaged kinetic energy of the \( j \)th oscillator is

\[ <K_j(t)> = \frac{1}{4} m\omega_0^2 |X_j(t)|^2. \] (3-5-10)

(B) Potential Energy of the \( j \)th Oscillator

The oscillators move under the influence of the strong linear restoring force \(-m\omega_0^2\)

\( X_j(t) \) and the weak radiative and non-radiative damping force \(-m\Gamma \dot{X}_j(t)\), which is down by a factor of \( \Gamma/\omega_0 \sim 10^{-12} \) in magnitude compared to the restoring force. The instantaneous potential energy of the oscillator is well defined,

\[ U_j(t) = \frac{1}{2} m\omega_0^2 \text{Re} X_j(t)^2. \] (3-5-11)

Combining with Eq. (3-3-7), we get,

\[ U_j(t) = \frac{1}{2} m\omega_0^2 \left( \sum_{k=1}^N |A_k X_j^{(k)}| \exp[-\frac{1}{2}(\Gamma_c^{(k)} + \Gamma_\alpha) t] \cos(\omega^{(k)} t - \alpha_j^{(k)}) \right)^2. \] (3-5-12)

Performing the same kind of time average as we did with the kinetic energy in Eq. (3-5-6), we get,
\[<U_j(t)> = \frac{1}{4} m\alpha_0^2 \exp(-\Gamma_\alpha t)\]

\[
\sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}||A_l X_j^{(l)}| \exp[-\frac{1}{2}(\Gamma_c^{(k)} + \Gamma_c^{(l)})t] \cos[(\omega^{(k)} - \omega^{(l)})t - (\alpha_j^{(k)} - \alpha_j^{(l)})].
\]

(3-5-13)

Comparing with Eq. (3-5-9), we get,

\[<U_j(t)> = \frac{1}{4} m\alpha_0^2 |X_j(t)|^2.\]

(3-5-14)

(C) Mechanical Energy of the jth Oscillator

The total mechanical energy of the jth oscillator \(E_j(t)\) is the sum of \(U_j(t)\) and \(K_j(t)\). Therefore,

\[<E_j(t)> = <U_j(t)> + <K_j(t)>\]

\[= \frac{1}{2} m\alpha_0^2 \exp(-\Gamma_\alpha t) \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}||A_l X_j^{(l)}| \exp[-\frac{1}{2}(\Gamma_c^{(k)} + \Gamma_c^{(l)})t] \cos[(\omega^{(k)} - \omega^{(l)})t - (\alpha_j^{(k)} - \alpha_j^{(l)})].\]

(3-5-15)

giving,

\[<E_j(t)> = \frac{1}{2} m\alpha_0^2 |X_j(t)|^2.\]

(3-5-16)

For the kth normal mode motion, we have,

\[|X_j(t)|^2 = |X_j^{(k)}(t)|^2 = |X_j^{(k)}|^2 \exp[-(\Gamma_c^{(k)} + \Gamma_\alpha)t],\]

(3-5-17)

which makes the mechanical energy of the jth oscillator in a pure normal mode system,
\[
\langle \mathcal{E}_j^{(k)}(t) \rangle = \frac{1}{2} m \omega_0^2 |X_j^{(k)}|^2 \exp[-(\Gamma_c^{(k)}+\Gamma_\omega)t].
\] (3-5-18)

So the mechanical energy of the \( j \)th oscillator decays exponentially with time.

However, if the initial state is a superposition of the normal modes, then \(|X_j(t)|^2\) is given by Eq. (3-5-9), and the energy will be transferred back and forth between different oscillators, with the modulation rates being the differences of the frequencies of different modes,

\[
\Omega_B(k,l) = |\omega^{(k)}-\omega^{(l)}|.
\] (3-5-19)

The decay will no longer be pure exponential. We will give an explicit example of this type of behavior for a system of 2 oscillators in the following chapter.

(D) The Mechanical Energy of the System

For the system, we have,

\[
\langle \mathcal{E}(t) \rangle = \sum_{j=1}^{N} \frac{1}{2} m \omega_0^2 |X_j(t)|^2
\]

But we have \( \sum_{j=1}^{N} |X_j(t)|^2 = |X(t)|^2 \), So the total mechanical energy is,

\[
\langle \mathcal{E}(t) \rangle = \frac{1}{2} m \omega_0^2 |X(t)|^2
\] (3-5-20)

For reference, \(|X(t)|^2 = \sum_{j=1}^{N} |X_j(t)|^2\) is, from Eq. (3-3-4) and (3-5-9), given by

\[
|X(t)|^2 = |\sum_{k=1}^{N} A_k X^{(k)} \exp[-i \omega^{(k)} t]|^2
\]
\[= \exp(-\Gamma_{\alpha}t) \sum_{j=1}^{N} \sum_{k=1}^{N} \sum_{l=1}^{N} |A_k X_j^{(k)}| A_{l} X_l^{(l)} \exp[-\frac{1}{2} (\Gamma_{c}^{(k)} + \Gamma_{c}^{(l)} ) t] \cos[(\omega_c^{(k)} - \omega_c^{(l)}) t - (\alpha^{(k)}_j - \alpha^{(l)}_l)] \]

\[(3.5.21)\]

where the expansion coefficient \(A_k\) is given by Eq. (3.3-3) and \(\alpha^{(k)}_j\) is the phase angle of the complex quantity \(A_k X_j^{(k)}\).

For pure normal mode motion, \(|X(t)|^2 = |X^{(k)}|^2 \exp[-(\Gamma_{c}^{(k)} + \Gamma_{\alpha}) t]\), therefore,

\[<\varepsilon^{(k)}(t)> = \frac{1}{2} m \alpha_0^2 |X^{(k)}|^2 \exp[-(\Gamma_{c}^{(k)} + \Gamma_{\alpha}) t] \]

\[(3.5.22)\]

decreases exponentially with time at the rate \((\Gamma_{c}^{(k)} + \Gamma_{\alpha})\).

If the initial excitation is a superposition of more than one normal mode, then \(X(t)\) is a superposition of normal mode motions [see Eq.(3.3.4)]. As we illustrated earlier in Eq. (3-2-3), the eigenvectors are transpose orthogonal instead of being Hermitian orthogonal. As the result, \((X^{(k)})^T \cdot (X^{(l)}) \neq \delta_{kl}\). Thus, \(|X(t)|^2\) has non-vanishing cross terms between normal modes, which introduces modulations at the various frequency differences \(\Omega_B(k,l)\), as shown explicitly in Eq. (3.5-22). \(<\varepsilon(t)>\) is still monotonically decreasing, but now the decay rate is no longer uniform, but instead will be modulated at the frequencies \(\Omega_B(k,l)\).

If a very large number of modes are initially excited, then the initial decay of \(<\varepsilon(t)>\) \((=|X(t)|^2)\) will be due in part to a "de-phasing" effect arising from the spread \(\Delta \omega\) of normal mode frequencies (affecting \(|X(t)|^2\) because of the non-Hermitian-orthogonality of the normal modes) as well as to the intrinsic decay rate \((\Gamma_{c}^{(k)} + \Gamma_{\alpha})\) of each normal mode. The "de-phasing" contribution can in fact be the dominant factor in the initial decay. As we will see in Chapter V, this is the situation for a nuclear exciton state \(|\Psi_e(k_0)>\), induced by a synchrotron pulse, with phasing vector \(k_0\) off-Bragg.
§ 3-6. Radiated Intensity and Radiated Power.

(A) Poynting Flux

Performing the same kind of time average as we did to the kinetic energy in Eq. (3-5-6), we get the Poynting flux for a system undergoing kth normal mode motion,

\[ <S^{(k)}> = \frac{c}{4\pi} <\text{Re}(E^{(k)}) \times \text{Re}(H^{(k)})> = \frac{c}{8\pi} \text{Re}[E^{(k)} \times H^{(k)*}], \quad (3-6-1) \]

where \( E^{(k)} \) and \( H^{(k)} \) are given by Eqs. (3-4-2) and (3-4-3). When the system dimension is much smaller than the distance between the observation point and the center of the system, we use Eqs. (3-4-4) and (3-4-5) as our field expressions. The Poynting flux expression then simplifies to

\[ <S^{(k)}> = \frac{c}{8\pi} |E^{(k)}|^2, \quad (3-6-2) \]

with \( E^{(k)} \) given by Eq. (3-4-4), giving,

\[ <S^{(k)}> = \frac{3}{16\pi} m \omega_0^2 \Gamma \gamma \frac{\sin^2(\theta)}{r^2} n \exp[-(\Gamma_c^{(k)} + \Gamma_\alpha(t - \frac{|R|}{c}))] \]

\[ \sum_{i=1}^{N} \sum_{j=1}^{N} |X_i^{(k)}| |X_j^{(k)}| \cos(k \cdot (R_i - R_j) - (\beta_i^{(k)} - \beta_j^{(k)})), \quad (3-6-3) \]

where \( \beta_i^{(k)} \) and \( \beta_j^{(k)} \) are the phase angles of \( X_i^{(k)} \) and \( X_j^{(k)} \), \( \theta \) is the angle between \( n \) and \( \hat{x} \) and \( k = k_0 n \).

For a general excitation, the Poynting flux is,

\[ <S> = \frac{c}{4\pi} <\text{Re}(E) \times \text{Re}(H)> = \frac{c}{8\pi} \text{Re}[E \times H^*], \quad (3-6-4) \]

where \( E \) and \( B \) are given by Eqs. (3-4-6) and (3-4-7). Again, far from the source, the Poynting flux simplifies to,
\[ <S> = \frac{c}{8\pi} n |E|^2, \]  

(3-6-5)

giving,

\[
\frac{3}{16\pi} m_0^2 \Gamma_c \sin^2(\theta) \frac{N}{r^2} \sum_{k=1}^{N} \sum_{l=1}^{N} \left\{ \exp[-\frac{1}{2}(\Gamma_c^{(k)}+\Gamma_c^{(l)}+2\Gamma_\alpha)(t-\frac{|R|}{c})] \cos[(\omega^{(k)}-\omega^{(l)})(t-\frac{|R|}{c})] \right\} 
\sum_{i=1}^{N} \sum_{j=1}^{N} |A_{k}X_i^{(k)}| |A_{l}X_j^{(l)}| \cos[k \cdot (R_i-R_j)-(\alpha_i^{(k)}-\alpha_j^{(l)})], \]  

(3-6-6)

where \( \alpha_i^{(k)} \) and \( \alpha_j^{(l)} \) are the phase angles of \( |A_{k}X_i^{(k)}| \) and \( |A_{k}X_j^{(l)}| \) respectively. If all \( A_k \)'s are zero except \( k=k \), then we are back at Eq. (3-6-3) where only the kth normal mode is being excited.

Comparing Eq. (3-6-2) with Eq. (3-6-3), we see an important distinction between a pure normal mode excitation and a general excitation which is superposition of many normal modes: \( <S^{(k)}> \) is a purely exponentially decaying function with a well defined decay rate \( \Gamma_c^{(k)}+\Gamma_\alpha \). On the other hand, \( <S> \) is not a pure exponentially decaying function in two senses. First, it contains components that have different decay rates \( \frac{1}{2}(\Gamma_c^{(k)}+\Gamma_c^{(l)}+2\Gamma_\alpha) \). More importantly, there are modulating factors \( \cos[(\omega^{(k)}-\omega^{(l)})(t-\frac{|R|}{c})] \), so that the emitted flux will pulse periodically, with the periodic modulation occurring at the frequencies \( \Omega_B(k,l) = (\omega^{(k)}-\omega^{(l)}) \). This leads to a time dependent instantaneous radiative decay rate \( \Gamma_c^{(l)} \), as we will discuss in § 3-7.
(B) Radiated Power

The power output of the system is the integration of the Poynting flux over the spherical surface,

\[ P = \int \mathbf{S} \cdot d\mathbf{\sigma} = \int \langle \mathbf{S} \rangle \cdot n r^2 d\Omega. \]  

\[(3-6-7)\]

For pure normal mode motion,

\[ p^{(k)} = \frac{3}{16\pi} m \omega_0^2 \Gamma \exp[-(\Gamma_c + \Gamma_\alpha)(t - \frac{|R|}{c_c})] \sum_{i=1}^{N} \sum_{j=1}^{N} \left| X_i^{(k)} \right| \left| X_j^{(k)} \right| \int d\Omega [1 - (n \cdot \hat{r})^2] \cos[\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j) - (\beta_i^{(k)} - \beta_j^{(k)})], \]

\[(3-6-8)\]

and for general motion,

\[ P = \frac{3}{16\pi} m \omega_0^2 \Gamma \exp[-\Gamma_\alpha(t - \frac{|R|}{c_c})] \sum_{k=1}^{N} \sum_{l=1}^{N} \left\{ \exp[-\frac{1}{2}(\Gamma_c + \Gamma_c')(t - \frac{|R|}{c_c})] \cos[(\omega_c^{(k)} - \omega_c^{(l)})(t - \frac{|R|}{c_c})] \right\} \sum_{i=1}^{N} \sum_{j=1}^{N} \left| A_i^{(k)} \right| \left| A_j^{(l)} \right| \int d\Omega [1 - (n \cdot \hat{r})^2] \cos[\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j) - (\alpha_i^{(k)} - \alpha_j^{(l)})]. \]

\[(3-6-9)\]

We again note from Eq. (3-6-9) that the radiated power will generally have pulsed modulations at the the rates \( \Omega_B(k,l) = (\omega_c^{(k)} - \omega_c^{(l)}) \).

These expressions will be used below to determine instantaneous radiative decay rate.

§ 3-7 Instantaneous Decay Rate

(i) Radiation and Decay

The instantaneous decay rate of the system is given by,
\[
\Gamma(t) = \Gamma_c(t) + \Gamma_\alpha = -\frac{1}{\langle \mathcal{E}(t) \rangle} \left\{ \frac{d\langle \mathcal{E}(t) \rangle}{dt} \right\}. \quad (3-7-1)
\]

The non-radiative decay rate, \( \Gamma_\alpha \), is independent of time, while the instantaneous radiative decay rate, \( \Gamma_c(t) \), is generally time dependent. Considering this, the time development of the mechanical energy is generally not an exponentially decaying function, but rather,

\[
\langle \mathcal{E}(t) \rangle = \mathcal{E}_0 \exp\left\{ -[\Gamma_\alpha + \frac{1}{t} \int_{t=0}^{t} \Gamma_c(t') dt'] \cdot t \right\}. \quad (3-7-2)
\]

\( \Gamma_c(t) \) can be determined either from the instantaneous radiated power \( P(t) \), or from the calculated time dependent energy \( \langle \mathcal{E}(t) \rangle \).

(A) Calculation of \( \Gamma_c(t) \) from the Radiated Power \( P(t) \)

The most intuitive approach is to calculate the radiative decay rate directly from the radiated power \( P(t) \),

\[
\Gamma_c(t) = -\frac{1}{\langle \mathcal{E}(t) \rangle} \left\{ \frac{d\langle \mathcal{E}(t) \rangle}{dt} \right\}_{\text{radiative}} = \frac{P(t)}{\langle \mathcal{E}(t) \rangle}, \quad (3-7-3)
\]

where \( P(t) \) is given by Eq. (3-6-8) for a pure normal mode excitation or by Eq. (3-6-9) for a general excitation.

Thus if the system is undergoing \( k \)-th normal mode motion, we have,

\[
\Gamma_c^{(k)} = \frac{P^{(k)}(t)}{\langle \mathcal{E}^{(k)}(t) \rangle}
\]

\[
= \frac{3 \Gamma_\gamma}{8 \pi |X^{(k)}|^2} \sum_{i=1}^{N} \sum_{j=1}^{N} |X_i^{(k)}|^2 |X_j^{(k)}|^2 \int d\Omega [1 - (n \cdot \hat{k})^2] \cos[k \cdot (R_i - R_j) - (\beta_i - \beta_j)],
\]

\( (3-7-4) \)
where we have substituted Eq. (3-5-22) in as the expression for $<\mathcal{E}^{(k)}(t)>$ and Eq. (3-6-8) in as the expression for $P^{(k)}$. We see that the resulting $\Gamma^{(k)}_c$ is no longer function of time.

Therefore when we substitute Eq. (3-7-4) into Eq. (3-7-2), it becomes,

$$<\mathcal{E}(t)> = \mathcal{E}^{(k)}_0 \exp\left[-(\Gamma_\alpha^{(k)}+\Gamma_c^{(k)})t\right].$$  \hspace{1cm} (3-7-5)

which is a truly exponentially decaying quantity with the decay constant,

$$\Gamma = \Gamma_\alpha + \Gamma_c^{(k)}.$$

On the other hand, when the system motion is the superposition of more than one normal modes, we have to use Eq. (3-5-21) as our $<\mathcal{E}(t)>$ and use Eq. (3-6-9) as our $P(t)$, which gives us a time dependent instantaneous decay rate $\Gamma_c(t)$, with periodic modulations at various frequency differences $\Omega_B(k,l)$. In turn, Eq. (3-7-2) can no longer be reduced to an exponential decay function. Instead, the average decay rate from $t=0$ to $t$ will be,

$$\Gamma(t) = \Gamma_\alpha^{(k)} + \frac{1}{t} \int_{t=0}^{t} \Gamma_c(t) dt.$$

(B) Calculation of the Instantaneous Decay Rate from $|X(t)|^2$

To derive $\Gamma_c(t)$ from the radiated power involves an integration $\int d\Omega(k)$ over the $4\pi$ solid angle to calculate $P(t)$. It's in fact much simpler to determine $\Gamma_c(t)$ directly from the mechanical energy $<\mathcal{E}(t)>$.

As given by Eq. (3-5-20), $<\mathcal{E}(t)> = \frac{1}{2} m \omega_0^2 |X(t)|^2$ with $X(t)$ given by Eqs. (3-3-4) and (3-3-5). We note from Eqs. (3-3-4) and (3-3-5) that the non-radiative decay factor $[\exp(-\frac{1}{2} \Gamma_\alpha t)]$ can be separated out of the expression $X(t)$, giving,

$$|X(t)|^2 = \exp(-\Gamma_\alpha t) |X_c(t)|^2.$$  \hspace{1cm} (3-7-6)

Here $X_c(t)$ gives the development of the system if we were to have $\Gamma_\alpha = 0$. From Eq. (3-7-6), the system mechanical energy is then,

$$<\mathcal{E}(t)> = \exp(-\Gamma_\alpha t) \left[ \frac{1}{2} m \omega_0^2 |X_c(t)|^2 \right].$$  \hspace{1cm} (3-7-7)
Substituting Eq. (3-7-7) into Eq. (3-7-1), the instantaneous radiative decay rate is then given by,

\[ \Gamma_c(t) = \frac{1}{|X_c(t)|^2} \frac{d}{dt} |X_c(t)|^2 = \frac{d}{dt} \ln(|X_c(t)|^2). \]  \hspace{1cm} (3-7-8)

Having solved the system motion under the general excitation, we know what \( X_c(t) \) will be at any given time \( t > 0 \). Therefore, we know what the instantaneous radiative decay rate will be at any moment \( t > 0 \).

Explicitly from Eq. (3-5-21), \( |X_c(t)|^2 \) is given by

\[ |X_c(t)|^2 = \sum_j |X_{cj}(t)|^2 \]

\[ = \sum_{j=1}^{N} \sum_{k=1}^{N} \sum_{l=1}^{N} |A_{kj}X_j^{(k)}||A_{jl}X_l^{(l)}|\exp[-\frac{1}{2}(\Gamma_c^{(k)}+\Gamma_c^{(l)})t] \cos[(\omega_j^{(k)}-\omega_j^{(l)})t - (\alpha_j^{(k)}-\alpha_j^{(l)})], \]

\hspace{1cm} (3-7-9)

and consequently,

\[ \frac{d}{dt} |X_c(t)|^2 = - \sum_{j=1}^{N} \sum_{k=1}^{N} \sum_{l=1}^{N} |A_{kj}X_j^{(k)}||A_{jl}X_l^{(l)}|\exp[-\frac{1}{2}(\Gamma_c^{(k)}+\Gamma_c^{(l)})t] \]

\[ \{(\omega_j^{(k)}-\omega_j^{(l)}) \sin [(\omega_j^{(k)}-\omega_j^{(l)})t - (\alpha_j^{(k)}-\alpha_j^{(l)})] + \frac{1}{2}(\Gamma_c^{(k)}+\Gamma_c^{(l)})\cos[(\omega_j^{(k)}-\omega_j^{(l)})t - (\alpha_j^{(k)}-\alpha_j^{(l)})]\}

\hspace{1cm} (3-7-10)

From Eqs. (3-7-8), (3-7-9) and (3-7-10), we can calculate the instantaneous decay rate for the system at any given moment \( t > 0 \). In the next chapter, we will use this to calculate and plot several examples with different parameters.
§ 3-8 Non-Identical Oscillators

Now we consider the situation in which the oscillators do not all share the common natural resonance frequency $\omega_0$. Quantum mechanically, it originates from the existence of hyperfine splittings of the nuclear levels and/or from the shifts between the levels of nuclei located in different chemical sites. Generally, there will only be a few of the different $\omega_j$'s within the system and the differences in natural resonance frequencies are at most $10^2\Gamma$ ($\sim 10^3\Gamma_\gamma$), which are only $10^{-11}\omega_0$ to $10^{-6}\omega_0$.

Just as in the identical oscillator case, we proceed to look for the radiative normal modes in which all oscillators move with a common eigenfrequency and decay rate. After pursuing similar lines of reasoning, we get the result that the equation of motion for the jth oscillator within a system undergoing the normal mode motion with complex eigenfrequency $\omega$ is given by,

$$m(\omega_j^2-\omega^2-i\omega\Gamma)X_j = \sum_{i \neq j} g_{ji}X_i, \quad (j=1, 2, \ldots, N) \quad (3-8-1)$$

where $X_j$ is the oscillation amplitude of the jth oscillator, $\omega_j$ is the natural resonance frequency of the jth oscillator, $\Gamma=\Gamma_\gamma+\Gamma_\alpha$ (assuming this quantity is universal for all oscillators) and $g_{ji}$ is defined in Eq.(3-1-7) as the interaction coefficient between the ith and the jth oscillators.

Because, the radiative interactions are extremely weak compared to the linear restoring forces [see discussion in § 3-1 (E)], we are able to linearize Eq. (3-8-1) into the form,

$$2m\omega_0(\omega_0+\Delta_j-\omega-i\frac{\Gamma_j}{2})X_j = \sum_{i \neq j} g_{ji}X_i, \quad (j=1, 2, \ldots, N) \quad (3-8-2)$$

where now $\omega_0$ is the average of the resonance frequencies,

$$\omega_j \equiv \frac{1}{N} \sum_{j=1}^{N} \omega_j \quad (3-8-3)$$

and $\Delta_j$ is the deviation of the resonance frequency of jth oscillator from $\omega_0$,

$$\Delta_j \equiv \omega_j - \omega_0. \quad (3-8-4)$$
With certain limitations [see footnote on page 15], we further make the approximation of replacing $\omega$ with $\omega_0$ on the right hand side of Eq. (3-8-2). With some notation changes, we are able to write Eq. (3-8-2) in the form of,

$$
(\omega_0 + \Delta_j - i \frac{\Gamma}{2}) X_j = \frac{1}{2} \Gamma \sum_{i \neq j} \kappa_{ji} \frac{\exp(ik_0\Gamma k_{ji})}{k_0^2 r_{ji}} X_i,
$$

where $\kappa_{ji}$ is defined in Eqs. (3-1-15), (3-1-16) and (3-1-17) as the interaction coefficient. Comparing to Eq. (3-1-13), we note that the only difference is the $\Delta_j$ on the left hand side.

As $j$ runs from 1 through $N$, we obtain $N$ linear equations with respect to $X_j$ ($j=1,2,\ldots,N$). Putting it into matrix form, we have,

$$
\mathbf{H} \mathbf{X} = \omega \mathbf{X},
$$

which looks the same as Eq. (3-2-1) except now the the diagonal elements of $\mathbf{H}$ matrix is re-defined as $h_{ij} = \omega_0 + \Delta_j - i \frac{\Gamma}{2}$.

With $\mathbf{H}$ redefined, the eigenfrequency function symbolically remains the same,

$$
\text{determinant } [\mathbf{H} - \omega I] = 0,
$$

which gives us $N$ eigenfrequency solutions $\omega^{(k)}$ with $k$ running from 1 through $N$. There exists the relation between these $\omega^{(k)}$s and the trace of $\mathbf{H}$,

$$
\sum_{k=1}^{N} \omega^{(k)} = -i \frac{N}{2} \Gamma + \sum_{j=1}^{N} \omega_j = N(-i \frac{1}{2} \Gamma + \omega_0).
$$

Taking the real and imaginary parts of Eq. (3-8-8), we get the new sum rules for the system of non-identical oscillators,

$$
\frac{1}{N} \sum_{k=1}^{N} \omega^{(k)} = \frac{1}{N} \sum_{k=1}^{N} \omega_j = \omega_0.
$$
and,

$$\frac{1}{N} \sum_{k=1}^{N} \Gamma^{(k)}_o = \Gamma_\gamma \quad (3-8-10)$$

The normal modes we get from Eq. (3-8-6) are again transpose orthogonal because the new \( \tilde{H} \) remains symmetric. So Eq. (3-2-3) applies here.

In addition, as long as the differences \( (\omega_j - \omega_{e_j}) \) between the natural resonance frequencies of different oscillators remain much smaller than \( \omega_0 \), the system of non-identical oscillators will inherit all the expressions we've obtained for the mechanical energy [Eqs. (3-5-20), (3-5-22), etc.], for radiation fields [Eqs. (3-4-6), (3-4-7), etc.], for Poynting flux [Eqs. (3-6-2) and (3-6-5), etc.], for radiative power output [Eqs. (3-6-8) and (3-6-9)] and for the radiative decay rate [Eqs. (3-7-4), (3-7-8), etc.].

It is important to note, however, the same symbolic expressions do not mean that systems of non-identical oscillators behave the same as systems of identical oscillators. The crucial difference starts from the eigenmode equations, Eqs. (3-8-6) and (3-8-7), in which the diagonal elements \( h_{jj} \) of the "effective Hamiltonian" \( \tilde{H} \) are defined differently than in the identical oscillator case.

As an example, we take a look at one extreme case: Suppose we have a small system which consists of only a few oscillators. Assume each oscillator has a different natural resonance frequency \( \omega_j = \omega_0 + \Delta_j \) and that the differences between those natural resonance frequencies are much more significant than the radiative coupling. In other words, this system satisfies the conditions,

\[ |\Delta_j| \gg |h_{ij}| (i \neq j) \text{ for all } \Delta_j \text{'s and } h_{ij} \text{'s} \]

and

\[ |\Delta_i - \Delta_j| \gg |h_{ij}| (i \neq j) \text{ for all } \Delta_j \text{'s and } h_{ij} \text{'s.} \]

The effective Hamiltonian \( \tilde{H} \) for such a system becomes nearly diagonal and the eigenfrequencies are close to \( \omega^{(k)} = \omega_k - \frac{\Gamma}{2} \). In other words, the eigenfrequencies are very
nearly the same as the natural resonance frequencies and the damping rate for each mode is very nearly the same as the damping rate for each isolated oscillator. In the kth mode, the kth oscillator moves approximately at its natural frequency $\omega_k$ with a large amplitude while all other oscillators, driven far off resonance by the radiation field produced by the kth oscillator, "jiggle" with small amplitudes. The general motion of such a system will very nearly be the "independent" motions of different oscillators. The radiation generally will be the superposition of the radiation fields from different oscillators moving with their own natural resonance frequencies. We will see "quantum beats" in the radiation of such a system with the beats frequencies being $\Omega_B(i,j) = |\omega_i - \omega_j|$, which can be fast compared to the characteristic decay time $\Gamma^{-1}$.

On the opposite limit, if $|\Delta_j| < |\epsilon_{ij}|$ for all oscillators, we are close to the case of identical oscillators. Then, the eigenfrequency shifts are determined primarily by the radiative coupling between the oscillators. The radiative interaction is what shapes the configurations of the normal modes. Under the most general circumstances, the system will be the superposition of various normal modes. Therefore the system radiation will also be modulated by rates $\Omega_B(k,l) = |\omega^{(k)} - \omega^{(l)}|$, only this time, $\Omega_B(k,l)$ will not reflect the differences in the natural resonance frequencies. Further more, the beat periods ($\sim \Omega_B^{-1}$) will now generally be on the order of the decay time, and the effect of the the $\Delta \omega$ spread of normal mode frequencies now tends to give a "de-phasing" decay of the initial excitation rather than a pattern of pronounced quantum beats.

In the next chapter, we will see this explicitly in an example of two coupled oscillators.
IV. Two Coupled Nuclei

We have developed the general approach to deal with a system of N coupled oscillators in Chapter III. As a specific example, we will first discuss two coupled oscillators. This is the simplest coupled system, yet we can already see features of "superradiance" and "subradiance", and, for non-identical oscillators, strong quantum beat modulations in the instantaneous decay rate.

§4-1 Two identical oscillators: Normal Mode Solutions

A. Eigenmode Solutions

Let's consider a system consisting of two identical oscillating dipoles. In this case, the eigenvalue equation Eq. (3-2-2) becomes,

$$\det \begin{pmatrix} \omega_0 - \omega - \frac{\Gamma}{2} & \frac{\Gamma}{2} \kappa_{12} \left( e^{ik_{12} r_{12}} - k_{012} \right) \\ \frac{\Gamma}{2} \kappa_{12} \left( e^{ik_{12} r_{12}} - k_{012} \right) & \omega_0 - \omega - \frac{\Gamma}{2} \end{pmatrix} = 0,$$

(4-1-1)

which gives us two eigen-solutions,

$$\omega^{(\pm)} = \omega_0 - \frac{\Gamma}{2} \mp \frac{\Gamma}{2} \kappa_{12} \frac{e^{ik_{12} r_{12}}}{k_{12}}.$$

(4-1-2)

The corresponding eigenvectors are,

$$X^{(\pm)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \mp 1 \end{pmatrix}.$$

(4-1-3)

The two eigenmodes correspond to symmetric and anti-symmetric motions. In the $X^{(+)}$ eigenmode, the two oscillators move together in phase, while in the $X^{(-)}$ eigenmode, the oscillators are moving $180^\circ$ out of phase. This result does not depend on further details of $\kappa_{12}$ and $k_{012}$.

To simplify the subsequent formalisms, we will replace $k_0$ with $k$ with the understanding that henceforth $k = k_0 = \omega_0/c$.

Letting the distance between the two oscillators $r_{12} = a$, we have
$$\kappa_{12} = \kappa_{21} = \frac{3}{2} \left\{ \sin^2(\theta_0) + [3\cos^2(\theta_0)-1][\frac{1}{(ka)^2} - \frac{i}{(ka)}]\right\},$$

(4.1-4)

where $\theta_0$ is the angle between $n_{12}$ and $\hat{n}$. From Eq. (4.1-2), we immediately get,

$$\omega^{(\pm)} = \omega_0 - i \frac{\Gamma}{2} \mp \frac{3\gamma}{4} (\frac{e^{ika}}{ka})(\sin^2(\theta_0) + [3\cos^2(\theta_0)-1][\frac{1}{(ka)^2} - \frac{i}{(ka)}]),$$

(4.1-5)

Recalling that $\Gamma = \Gamma_{\gamma} + \Gamma_{\alpha}$, we can re-write Eq. (4.1-5) in the form,

$$\omega^{(\pm)} = \omega_0 - \Gamma_{\gamma} \left[ \frac{i}{2} \pm \frac{3\sin^2(\theta_0)e^{ika}}{4ka} \pm \frac{3[3\cos^2(\theta_0)-1]}{4(ka)^3}(1-ika)e^{ika} \right] - \frac{\Gamma_{\alpha}}{2},$$

(4.1-6)

Let's look at some of the implications the above expressions have. First, the coherent radiative decay rates of the two modes are

$$\Gamma_c^{(+)} = \Gamma_{c}^{(-)} - \Gamma_{\alpha} = -2\text{Im}(\omega_{\pm}) - \Gamma_{\alpha}$$

$$= \Gamma_{\gamma} \left\{ 1 \pm \frac{3\sin^2(\theta_0)}{2ka} \frac{\sin(ka)\pm \frac{3}{2(ka)^3}(\sin(ka)-k\cos(ka))[3\cos^2(\theta_0)-1]}{2} \right\}.$$  

(4.1-7)

The average of the radiative decay rates of the + and - modes gives the decay rate $\Gamma_{\gamma}$ of an isolated oscillator,

$$\frac{1}{2} [\Gamma_c^{(+)} + \Gamma_c^{(-)}] = \Gamma_{\gamma}$$

(4.1-8)

consistent with the general sum rule in Eq. (3.2-7). In the near field region, $ka << 1$. Eq. (4.1-7) can be expanded in terms of $ka$, giving

$$\Gamma_c^{(\pm)} = \Gamma_{\gamma} \left\{ 1 \pm \frac{1}{10} (ka)^2 (1+ \sin^2\theta_0) + ... \right\}.$$  

(4.1-9)

We see that the limit is $\Gamma_c^{(\pm)} = \Gamma_{\gamma} (1\pm 1)$ as $ka$ approaches zero. This result is easy to understand. For the symmetric mode, the two dipoles oscillate together in phase. If they are so close together that the distance between them is much shorter than the wavelength, their radiation fields add constructively so that the output power ($P$) is 4 times as large as that of a single oscillator whereas the system energy ($\mathcal{E}$) is two times that of a single oscillator, so the radiative decay rate ($=P/\mathcal{E}$) is twice as large. For the anti-symmetric
mode on the other hand, they oscillate 180 degrees out of phase and their fields nearly cancel for $ka<<1$, so the system is only weakly radiating in this mode. For $ka << 1$, the system in the anti-symmetric state can be viewed as a electric quadrupole oscillator, with radiative decay rate $\Gamma_\gamma(E2) = \frac{1}{10} (ka)^2 (1+\sin^2\theta_0)\Gamma_\gamma(E1)$.

Fig. 1 gives plots of the radiative decay rates $\Gamma_c(\pm)$ as functions of $ka$. The solid curve is $\Gamma_c(+) = \Gamma(+) - \Gamma_\alpha$ while the dashed curve is $\Gamma_c(-) = \Gamma(-) - \Gamma_\alpha$. The flat line in the middle is their average. The vertical unit is $\Gamma_\gamma$, the radiative decay rate for an isolated oscillator. We show the results for two configurations: $\theta=0^0$ and $\theta=90^0$, corresponding respectively to the line of centers directions $n_{12}$ being parallel or perpendicular to the oscillation direction $\hat{x}$.

As just noted, when $ka$ goes to zero, the + mode's radiative decay rate is twice as great as a single oscillator, $\Gamma_c(+) = 2\Gamma_\gamma$ while the - mode becomes nonradiative, $\Gamma_c(-) = 0$. However, when $ka$ becomes large (which means $a>>\lambda$), the decay rates of both modes approach the single oscillator rate. In between, the two modes alternate between $\Gamma_c(+) > \Gamma_\gamma$ and $\Gamma_c(-) < \Gamma_\gamma$.

![Fig. 1](image)

Fig. 1  The radiative decay rates $\Gamma_c(\pm)$ as a functions of $ka$, for (a) $\theta_0 = 0^0$ and (b) $\theta_0 = 90^0$.

The vertical unit: $\Gamma_\gamma$.

Comparing the results for $\theta_0 = 0^0$ and $\theta_0 = 90^0$, we see that there is little difference in the near field region ($ka<<1$) (this will be discussed in the following section B), but in the far field region ($ka>>1$), the deviations of $\Gamma_c(\pm)$ are far more pronounced for $\theta_0 = 90^0$. 
than for $\theta_0 = 0^0$. This is because for $\theta_0 = 90^0$, the oscillations are coupled by the transverse radiation field which is proportional to $ka^{-1}$, while for $\theta_0 = 0^0$, the transverse fields are zero, and the coupling is only through the near zone longitudinal fields which fall off proportional to $ka^{-3}$.

Now let's look at the frequency shifts. Taking the real part of Eq. (4-1-6), we get,

$$\omega^{(\pm)} = \text{Re}[\omega^{(\pm)}] = \omega_0 \left[ 1 + \frac{3\Gamma_\gamma}{4\omega_0} \frac{\sin^2(\theta_0) \cos ka}{ka} + \frac{\cos(ka) + ka \sin(ka)}{(ka)^3} (3\cos^2(\theta_0) - 1) \right].$$

(4-1-10)

For $ka \ll 1$,

$$\omega^{(\pm)} = \omega_0 \left[ 1 + \frac{3\Gamma_\gamma}{4\omega_0} \frac{1}{(ka)^3} (3\cos^2(\theta_0) - 1) \right].$$

(4-1-11)

Fig. 2 gives plots of the frequency shifts $\Delta^{(\pm)} = \omega^{(\pm)} - \omega_0$ as functions of $ka$ (for $\theta_0 = 0^0$ and $90^0$). The solid line gives $\Delta^{(+)}$, the dashed line gives $\Delta^{(-)}$. The straight line in the middle is their average. We see that as $ka$ goes to zero, the frequency shifts become large compared to $\Gamma_\gamma$, while as $ka$ becomes large, the frequency shifts of both modes approach zero (i.e., the frequencies $\omega^{(\pm)}$ approach $\omega_0$). For all $ka$ values, the average of the frequency shifts remains zero, consistent with the sum rule Eq. (3-2-5).

![Frequency Shifts Diagram](image)

Fig. 2. The frequency shifts $\Delta^{(\pm)} = \omega^{(\pm)} - \omega_0$ as functions of $ka$, for (a) $\theta_0 = 0^0$ and (b) $\theta_0 = 90^0$. The vertical unit is $\frac{\Gamma_\gamma}{2}$. 
It is not surprising that the frequency shifts become large for \(ka<<1\). It is the direct result of the strong increase of the interaction between the two oscillators for \(ka<<1\). This is discussed further in section B. Notice that we are still using the small shift expansion assuming the correction terms are much smaller than \(\omega_0\) [i.e., we are still using the linearized eigenvalue equation Eq. (3-1-13)]. This is because in reality \(\frac{\Gamma}{\omega_0}\) is so small that until two oscillators are extremely close together the corrections are still negligible as compared to \(\omega_0\). For \(^{57}\)Fe 14.4 kev resonance, \(\Gamma\gamma=5\times10^{-9}\)eV/\(\hbar\) and \(\omega_0=14.4\) kev/\(\hbar\), so the ratio between \(\Gamma\gamma\) and \(\omega_0\) is approximately \(3\times10^{-13}\) to 1. Thus \(ka\) has to be \(3\times10^{-4}\) for the correction terms to be comparable to \(\omega_0\) which corresponds to \(a=4\times10^{-5}\) A, which is already "inside the nucleus"! The conclusion is that for our purposes, the small frequency shift approximation works very well for all regions of interest.

Comparing the results of \(\Delta^{(\pm)}\) for \(\theta_0=0^0\) and \(90^0\), we draw the following two conclusions.

(1). For \(ka>>1\), we see that the shifts are much more pronounced for \(\theta_0=90^0\) than it is for \(\theta_0=0^0\). The reason is precisely the same as what we have discussed for \(\Gamma_c^{(\pm)}\): the absence of transverse radiation field in the coupling for \(\theta_0=0^0\).

(2). For \(ka<<1\), the frequency shifts have a strong dependence upon \(\theta_0\), in contrast to the near zone behavior of \(\Gamma_c^{(\pm)}\): for \(\theta_0=0^0\), the symmetric mode is the low frequency mode when \(ka<<1\), while for \(\theta_0=90^0\) it becomes the high frequency mode. The reason for the strong angular dependence is discussed in the following section.

B. Near Zone Behavior

The near zone behaviors of \(\Gamma_c^{(\pm)}\) and \(\Delta^{(\pm)}\) are easy to understand by examining the dipole field Eq. (3-1-4) in the near zone. For a harmonically oscillating dipole moment \(d_1(t) = d_1^0 e^{-i\omega t}\), the radiation field in the near zone limit \(kr<<1\) can be obtained by expanding Eq. (3-1-4) in terms of \(kr\). The result is,

\[
E(r,t) = k^2 \left\{ \frac{1}{(kr)^3} \left[ 3(n \cdot d_1) - d_1 \right] + \frac{1}{2kr} \left[ n(n \cdot d_1) + d_1 \right] + \frac{2}{3} i d_1 \right\}, \quad (4-1-12)
\]
where we have neglected terms proportional to \((kr)^n\) \((n \geq 1)\).

Eq. (4-1-12) consists of three terms. The first two terms give the field component which is always in phase with \(d_1\). The directions of \(n\) and \(d_1\) determine the direction of the field from these two contributions. The third term gives the field contribution always \(90^0\) out of phase with \(d_1\) along the direction of \(d_1\), regardless of \(n\) and \(r\).

First, let's look at the in-phase contribution. Among the two terms, the first is dominant when \(ka<<1\). For that reason, we neglect the second term in the discussion. We can easily see that the first term is just the "instantaneous" electric dipole field due to the dipole moment \(d_1\) at the instant of time \(t\). Fig. 3 illustrates such a field distribution at a time when \(|d_1|\cos(\omega_1 t) > 0\).

Now consider a second dipole \(d_2\) moving in phase with \(d_1\), i.e., symmetric mode motion. The oscillating dipole is a positive-negative charge pair bound together by the linear restoring force \(-\lambda_0^2 X_2(t)\). If the second dipole is located at position \(A\) corresponding to \(\theta_0=0^0\) (see Fig. 3), then the field \(E_{21}\) from \(d_1\) gives a force \(F_{21}\) which pulls the two dipole charges apart, and hence acts to oppose the linear restoring force of \(d_2\), thus reducing the oscillation frequency \(\omega^{(1)}\) to be less than \(\omega_0\). In contrast, at position \(B\), for which \(\theta_0=90^0\), \(F_{21}\) acts in concert with the linear restoring force, giving \(\omega^{(+)} > \omega_0\). For the antisymmetric mode in which two oscillators are moving \(180^0\) out of phase, the situation is just the opposite. We have a higher mode frequency for \(\theta_0=0^0\) and a lower mode frequency for \(\theta_0=90^0\).
Fig. 3 Instantaneous electric dipole field Also illustrated is the interaction acting on the second oscillator in the vicinity.

Now, we turn to the third term in Eq. (4-1-12), which generates the 90° out-of-phase field with respect to the motion of \( \mathbf{d}_1 \). This field will exert a damping force on \( \mathbf{d}_2 \) no matter where it's located. To see this more clearly, we note that \( \mathbf{d}_1(t) = qX_1(t) \mathbf{\hat{X}} = qX_1^0 \exp(-i\omega_1 t) \mathbf{\hat{X}} \) and \( \Gamma_\gamma = \frac{2q^2 \omega_1^2}{3mc^3} \), so that the contribution can be rewritten as,

\[
\frac{2}{3} i k^3 d_1^0 \exp(-i\omega_1 t) = -\frac{m}{q} \Gamma_\gamma \dot{X}_1 \mathbf{\hat{X}}. \tag{4-1-13}
\]

The force exerted on charge \( q \) of dipole 2 at either position (A) or (B) is,

\[
\mathbf{F}_{21} \text{(damping)} = -m\Gamma_\gamma \dot{X}_1 \mathbf{\hat{X}}. \tag{4-1-14}
\]
This force will add or subtract from the self damping force $F_{22}^{\text{(damping)}} = -m_{\gamma} \dot{\gamma}_{2} \hat{\gamma}$, depending upon the modes. For the symmetric mode, $X_{1}(t) = X_{2}(t)$, so $\dot{X}_{1} = \dot{X}_{2}$ and the total damping force on dipole 2 becomes $F_{21}^{\text{(damping)}} + F_{22}^{\text{(damping)}} = -m(2\Gamma_{\gamma}) \dot{\gamma}_{1} \hat{\gamma}$, giving an enhanced radiative decay rate of $2\Gamma_{\gamma}$ in agreement with our earlier discussion and plots. For the anti-symmetric mode, $\dot{X}_{1} = - \dot{X}_{2}$, so that $F_{21}^{\text{(damping)}}$ cancels the self damping force $F_{22}^{\text{(damping)}}$, leading to an undamped motion.

§4-2 Normal Mode Radiation and Calculation of $\Gamma_{c}^{(e)}$.

In this section, we calculate the normal mode emission patterns for the identical oscillators. We also show we get the same result for the normal mode radiative decay rates $\Gamma_{c}^{(e)}$ by using the total radiated power, as given by Eq. (3-6-8).

The radiation fields for a normal mode excitation have been given in Eqs. (3-4-2) and (3-4-3) generally and in Eqs. (3-4-4) and (3-4-5) for the far field approximation. We consider a spherical surface of radius $r >> |R_{1} - R_{2}|$ and use the far field approximation. Directly from Eqs. (3-4-4) and (3-4-5), we have,

$$E^{(e)} =$$

$$\frac{1}{\sqrt{2}} q_{0}^{2} \left[ \hat{\gamma} - (n \cdot \hat{\gamma}) n \right] X_{0} \exp[-i\omega^{(e)} t'] \exp(-i \frac{1}{2} \mathbf{k} \cdot \mathbf{a} \pm \exp(i \frac{1}{2} \mathbf{k} \cdot \mathbf{a})], \quad (4-2-1)$$

where $\mathbf{a} = R_{1} - R_{2}$ is the separation between the two oscillators, $t' = t - \frac{r}{c}$ is the retarded time and $X_{0}$ is the amplitude of the normal mode excitation*. The point of origin is chosen to be at $\frac{1}{2} (R_{1} + R_{2})$. In our case, we have $|\mathbf{a}| << r$.

From Eq. (3-6-2), the time averaged Poynting flux is,

$$<S^{(e)}> = \frac{3}{8\pi} \left( \frac{1}{2} m \omega_{0}^{2} X_{0}^{2} \right) \Gamma_{\gamma} \frac{1}{r^{2}} \sin^{2} \theta \mathbf{n} \exp[-\Gamma^{(e)} t'][1 \pm \cos(k \cdot a)]. \quad (4-2-2)$$

* In other words, the normal mode excitations now look like this: $X^{(e)} = \frac{1}{\sqrt{2}} \left( \frac{1}{\pm 1} \right) X_{0}$.
The factor \([1 \pm \cos(k \cdot a)]\) gives the usual interference modulation factor for the intensity in the \(k\) direction of the radiation emitted from two dipoles oscillating in phase (+) or out of phase (−).

The total radiated power

\[
P(t') = \int <S^{(±)}> \cdot \mathbf{r}^2 d\Omega, \quad (4-2-3)
\]

is then given by,

\[
P^{(±)}(t') = \frac{3}{8\pi} \left( \frac{1}{2} m \omega_0^2 x_0^2 \Gamma_\gamma \exp[-\Gamma^{(±)} t'] \right) \int d\Omega \sin^2 \theta \left[ 1 \pm \cos(k \cdot a) \right]
\]

\[
= \frac{3}{8\pi} \left( \frac{1}{2} m \omega_0^2 x_0^2 \Gamma_\gamma \exp[-\Gamma^{(±)} t'] \right) \\
\int_0^\pi \int_0^{2\pi} d\theta d\phi \sin^3 \theta \left[ 1 \pm \sum_{n=0}^{\infty} (ka)^n (\sin \theta \sin \phi \cos \phi + \cos \phi \cos \theta)^n \right]
\]

\[
= \left( \frac{1}{2} m \omega_0^2 x_0^2 \Gamma_\gamma \exp[-\Gamma^{(±)} t'] \right) \\
\left( 1 \pm \frac{3}{8\pi} \int_0^\pi \int_0^{2\pi} d\theta d\phi \sin^3 \theta \sum_{n=1}^{\infty} (ka)^n (\sin \theta \sin \phi \cos \phi + \cos \phi \cos \theta)^n \right) \]

\[
= \left( \frac{1}{2} m \omega_0^2 x_0^2 \Gamma_\gamma \exp[-\Gamma^{(±)} t'] \right) \left( 1 \pm \frac{1}{10} (ka)^2 (1 + \sin^2 \theta_0) + \ldots \right) \quad (4-2-4)
\]

where \(\theta_0\) is the angle between the \(\hat{x}\) axis (the oscillation direction) and \(a\).

The time averaged system energy \(<E(t)>\) is given by Eq. (3-5-20) in general. For our case, with \(X^{(±)}\) given in the footnote on the previous page, the energy is,

\[
<E^{(±)}(t)> = \frac{1}{2} m \omega_0^2 x_0^2 \exp[-\Gamma^{(±)} t]. \quad (4-2-5)
\]

Substitute Eqs. (4-2-4) and (4-2-5) into Eq. (3-7-3), we get the radiative damping rate,

\[
\Gamma_c^{(±)} = \Gamma_\gamma \left( 1 \pm \frac{1}{10} (ka)^2 (1 + \sin^2 \theta_0) + \ldots \right),
\]

which is in agreement with the result from the direct eigenmode approach in Eq. (4-1-9).
§4-3 Calculation of $\Gamma^{(\pm)}$ from Rate of Work Considerations

As a final check of consistency, we calculate the normal mode decay rates $\Gamma^{(\pm)} = \Gamma_c^{(\pm)} + \Gamma_\alpha$ directly from the rate at which work is being done on the system:

Taking oscillator 2's perspective, it experiences two forces. One is its own damping force which gives the natural decay $\Gamma_c^{(\pm)} + \Gamma_\alpha$ (discussed in Appendix I and II),

$$F_2^{(\pm)} = -m(\Gamma_c^{(\pm)} + \Gamma_\alpha)v_2^{(\pm)}, \quad (4-3-1)$$

where $v_2$ is the velocity of the oscillator. If the system is in one of the normal modes, we have the instantaneous displacements of the two oscillators,

$$X_1^{(\pm)} \hat{x} = \frac{X_0}{\sqrt{2}} \exp[-\frac{1}{2} \Gamma_c^{(\pm)} t] \exp[-i\omega^{(\pm)} t] \hat{x}$$

$$X_2^{(\pm)} \hat{x} = \pm \frac{X_0}{\sqrt{2}} \exp[-\frac{1}{2} \Gamma^{(\pm)} t] \exp[-i\omega^{(\pm)} t] \hat{x},$$

which gives the velocities,

$$v_1^{(\pm)} = -i \frac{\omega_0}{\sqrt{2}} X_0 \exp[-\frac{1}{2} \Gamma_c^{(\pm)} t] \exp[-i\omega^{(\pm)} t] \hat{x}$$

$$v_2^{(\pm)} = \mp i \frac{\omega_0}{\sqrt{2}} X_0 \exp[-\frac{1}{2} \Gamma^{(\pm)} t] \exp[-i\omega^{(\pm)} t] \hat{x},$$

where we have made the approximation of replacing the complex $\omega^{(\pm)}$ with $\omega_0$ outside the exponential function. The second force on oscillator 2 is due to the radiation field from the first oscillator, which, according to Eq. (3-1-6), is given by,

$$(F_2^{(\pm)}_x) = \frac{X_0}{\sqrt{2}} q_0^2 k^2 \frac{1}{a} \left[ (1 - (\hat{a} \cdot \hat{x})^2) + [3(\hat{a} \cdot \hat{x})^2 - 1] \frac{1}{(ka)^2} \frac{i}{ka} \right] \exp[-\frac{1}{2} \Gamma^{(\pm)} t] \exp[i(ka - i\omega^{(\pm)} t)], \quad (4-3-2)$$

where $\hat{a} = a/|a|$ is the unit vector along the direction of $a$. Here we only need the $\hat{x}$ component of the force because the motion is constrained to move along the $\hat{x}$ direction.

The time averaged rate at which work is done on the second oscillator is then,
\[ \frac{d}{dt} \langle \xi_2^{(\pm)} \rangle = \langle ( F_{22}^{(\pm)} + F_{21}^{(\pm)} ) \cdot v_2 \rangle = \frac{1}{2} \text{Real}[( F_{22}^{(\pm)} + F_{21}^{(\pm)} ) \cdot v_2^{(\pm)*}] \]

\[ \approx -\frac{1}{4} m_o \omega_0^2 x_0^2 \exp[-\Gamma^{(\pm)}t] \left\{ (\Gamma_y + \Gamma_\alpha) \pm \frac{3}{2} \Gamma_y \left( \sin^2 \Theta_0 \frac{\sin(ka)}{ka} + [3\cos^2 \Theta_0 - 1] \frac{\sin(ka)}{(ka)^3} - \frac{\cos(ka)}{(ka)^2} \right) \right\}. \quad (4-3-3) \]

The energy of the second oscillator, according to Eq. (3-5-16), is time averaged to,

\[ \langle \xi_2^{(\pm)}(t) \rangle = \frac{1}{4} m_o \omega_0^2 x_0^2 \exp(-\Gamma^{(\pm)}t). \]

Therefore the decay rate of the second oscillator is,

\[ \Gamma_2^{(\pm)} = -\frac{\frac{d}{dt} \langle \xi_2^{(\pm)} \rangle}{\langle \xi_2^{(\pm)}(t) \rangle} = \Gamma_y + \Gamma_\alpha \]

\[ \pm \frac{3}{2} \Gamma_y \left( \sin^2 \Theta_0 \frac{\sin(ka)}{ka} + [3\cos^2 \Theta_0 - 1] \frac{\sin(ka)}{(ka)^3} - \frac{\cos(ka)}{(ka)^2} \right). \quad (4-3-4) \]

Similarly, we find the decay rate of the first oscillator,

\[ \Gamma_1^{(\pm)} = \Gamma_2^{(\pm)} = \Gamma_y + \Gamma_\alpha \]

\[ \pm \frac{3}{2} \Gamma_y \left( \sin^2 \Theta_0 \frac{\sin(ka)}{ka} + [3\cos^2 \Theta_0 - 1] \frac{\sin(ka)}{(ka)^3} - \frac{\cos(ka)}{(ka)^2} \right). \quad (4-3-5) \]

We see from Eqs. (4-3-4) and (4-3-5) that the two oscillators are decaying with the same rate, \( \Gamma^{(\pm)} = \Gamma_1^{(\pm)} = \Gamma_2^{(\pm)} \). The result agrees with Eq.(4-1-7).

§4-4 General Motion of Two Identical Oscillators

A. General Motion of the System

Suppose the two oscillators are initially at \( X_1(t=0)=\xi_A \) and \( X_2(t=0)=\xi_B \) so that the initial state of the system can be expressed as,*

\[ \xi_A \text{ and } \xi_B \text{ are two complex quantities. They actually specify both the initial displacements and the initial velocities of the two oscillators. The initial displacements of the two oscillators are } (\text{Re}(\xi_A), \text{Re}(\xi_B)). \]
\[ X(t=0) = \begin{pmatrix} \xi_A \\ \xi_B \end{pmatrix} \]

Expanding Eq. (4-4-1) with eigenmodes \( X^{(\pm)} \) of Eq. (4-1-3), we have,

\[ X(t=0) = \frac{1}{\sqrt{2}} (\xi_A + \xi_B) X^{(+)} + \frac{1}{\sqrt{2}} (\xi_A - \xi_B) X^{(-)} . \]

At times \( t > 0 \), the state of the system will be,

\[ X(t) = \frac{1}{\sqrt{2}} (\xi_A + \xi_B) X^{(+)} \exp[-\frac{\Gamma^{(+)}}{2} t] \exp[-\omega^{(+)} t] \]
\[ + \frac{1}{\sqrt{2}} (\xi_A - \xi_B) X^{(-)} \exp[-\frac{\Gamma^{(-)}}{2} t] \exp[-\omega^{(-)} t] , \]

so that the motion of oscillator \( j (j=1, 2) \) is,

\[ X_1(t) = \frac{1}{2} (\xi_A + \xi_B) \exp[-\frac{\Gamma^{(+)}}{2} t] \exp[-\omega^{(+)} t] + \frac{1}{2} (\xi_A - \xi_B) \exp[-\frac{\Gamma^{(-)}}{2} t] \exp[-\omega^{(-)} t] \]
\[ X_2(t) = \frac{1}{2} (\xi_A + \xi_B) \exp[-\frac{\Gamma^{(+)}}{2} t] \exp[-\omega^{(+)} t] - \frac{1}{2} (\xi_A - \xi_B) \exp[-\frac{\Gamma^{(-)}}{2} t] \exp[-\omega^{(-)} t] . \]

The mechanical energy of oscillator \( j \) is

\[ < \varepsilon_j(t) > = \frac{1}{2} m \omega_j^2 |X_j(t)|^2 , \]

with \( X_j(t) \) now given by Eq.(4-4-4), and the total energy of the system is

\[ < \varepsilon(t) > = \frac{1}{2} m \omega_0^2 |X(t)|^2 \]
\[ = \frac{1}{4} m \omega_0^2 (|\xi_A + \xi_B|^2 \exp[-\Gamma^{(+) t}] + |\xi_A - \xi_B|^2 \exp[-\Gamma^{(-)} t]) . \]

The initial velocities of the two oscillators are, to very good approximation, \( \{ \omega_0 \text{Im}(\xi_A), \omega_0 \text{Im}(\xi_B) \} \), i.e., replacing \( \omega^{(\pm)} \) by \( \omega_0 \).
From Eq. (4-4-6), we see that the total mechanical energy of the two identical oscillator system monotonically and smoothly decays. It is not a true exponential decay because of the mixture of two modes with different decay rates.

Although the total energy $\langle \varepsilon(t) \rangle = \sum_j \langle \varepsilon_j(t) \rangle$ decays smoothly, if the initial excitation is not a normal mode, then the energies $\langle \varepsilon_j(t) \rangle$ of individual oscillators will pulse with time at the normal mode frequency difference $\Omega_B$,

$$\Omega_B = |\omega^{(+)} - \omega^{(-)}|$$

$$\Omega_B = \frac{3\Gamma_\gamma}{2} \sin^2(\theta_0) \cos ka - \frac{\cos(ka) + ka \sin(ka)}{(ka)^3} \cdot (3\cos^2(\theta_0) - 1)$$  \hspace{1cm} (4-4-7)

with the energy being transferred back and forth between the oscillators. This interesting behavior is particularly pronounced if initially only one oscillator is displaced from equilibrium, for example, $X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$. Fig. 4 shows the subsequent mechanical energies $|X_1(t)|^2$ and $|X_2(t)|^2$ for (a) a near zone case $ka=0.1\pi$, for which $\Omega_B=46.2\Gamma_\gamma$ and for (b) a far zone case with $ka=\pi$, for which $\Omega_B=0.574\Gamma_\gamma$.

(a) $ka = 0.1\pi$

(b) $ka = \pi$

Fig. 4 Energy of the jth oscillator $j=1$ for solid line, $j=2$ for dashed line. The horizontal axis is time with unit $(1/\Gamma_\gamma)$. $\theta_0=90^0$ and $X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ for both (a) and (b).
As we see from the examples, appreciable energy transfer will only be observed for the near zone case \((ka<<1)\) where \(\Omega_B > \Gamma_\gamma\). For the case of Fe\(^{57}\) 14.4kev resonance, if \(a=1\)A, then \(ka=7.3\). When \(\theta_0=90^\circ\), then \(\Omega_B=0.082\Gamma_\gamma\), leading to very weak \(\Omega_B\) modulation during the life time of the Fe\(^{57}\) 14.4kev resonance.

B. The Instantaneous Decay Rate

In Chapter III, we discussed the instantaneous decay rate for a general excitation. Directly applying the formulas into our two oscillator case, we get the instantaneous decay rate of the system,

\[
    \Gamma(t) = \frac{1}{\langle \mathcal{E}(t) \rangle} \left\{ \frac{d\langle \mathcal{E}(t) \rangle}{dt} \right\} = \frac{(\xi_A + \xi_B)\Gamma^{(t)} \exp[-\Gamma^{(t)}t] + (\xi_A - \xi_B)\Gamma^{(-)} \exp[-\Gamma^{(-)}t]}{(\xi_A + \xi_B)\exp[-\Gamma^{(t)}t] + (\xi_A - \xi_B)\exp[-\Gamma^{(-)}t]},
\]

which is a time dependent quantity.

We note however that for two identical oscillators, there are no periodic modulation of \(\langle \mathcal{E}(t) \rangle\) and \(\Gamma(t)\) at the rate \(\Omega_B=|\omega^{(+)} - \omega^{(-)}|\), as was predicted in §3-5 and §3-7. This occurs because the normal mode vectors \(X^{(2)}\) given by Eq. (4-1-3) are real, and hence are Hermitian orthogonal as well as transpose orthogonal. As a consequence, there are no interference terms between the normal modes in \(\langle \mathcal{E}(t) \rangle \propto |X(t)|^2\). This situation is special to the system of 2 identical oscillators.

C. Differential Decay Rate and Beats

The total radiation field from the system of initial excitation Eq. (4-4-1) is, in the far field region,

\[
    E = \frac{1}{\sqrt{2}} (\xi_A + \xi_B)E^{(+)} + \frac{1}{\sqrt{2}} (\xi_A - \xi_B)E^{(-)}
    = \frac{1}{\sqrt{2}} \frac{k_0 \mathbf{n}}{\Gamma} \left[ \mathbf{\hat{k}} - (\mathbf{n} \cdot \mathbf{\hat{k}}) \mathbf{n} \right]
    \left\{ (\xi_A + \xi_B)\exp[-i\omega^{(+)}t]\cos(\frac{1}{2} k \cdot a) - i (\xi_A - \xi_B)\exp[-i\omega^{(-)}t]\sin(\frac{1}{2} k \cdot a) \right\},
\]

(4-4-9)
where again $t'=t-\tau/c$ and $\omega^{(\pm)}=\omega^{(\pm)}-i\frac{1}{2} \Gamma^{(\pm)}$. The factor $\cos (\frac{1}{2} k \cdot a)$ gives the interference modulation of the field due to the motion in the symmetric mode, and $\sin (\frac{1}{2} k \cdot a)$ gives the modulation due to the anti-symmetric mode.

The differential decay rate \( \frac{d\Gamma_c}{d\Omega} \) due to the rate at which energy is radiated into the solid angle \( d\Omega(n) \) is,

\[
\frac{d\Gamma_c}{d\Omega} d\Omega(n) = \frac{|<S>|^2 r^2 d\Omega(n)}{<E(t)>},
\]

(4-4-10)

where \( <S> = \frac{c}{8\pi} n |E|^2 \) with \( E \) now given by Eq. (4-4-9). The interesting point is that there can now be beats in the emitted intensity in the \( k \) direction, arising from the cross term in \( |E|^2 \), at the rate \( \Omega_B = |\omega^{(+)\Omega^{(-)}|} \).

Assuming \( ka = 0.1\pi \), \( \theta_0 = 90^0 \) and the initial excitation is \( X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \). Fig. 5 plots the instantaneous differential decay rate \( d\Gamma_c/d\Omega \) as the function of time in the \( n = a/|a| \) direction.

![Instantaneous Differential Decay Rate](image)

Fig. 5 Instantaneous differential decay rate as the function of time along direction of \( n = a/|a| \) when \( \theta_0 = 90^0 \), \( ka = 0.1\pi \) and initial condition: \( \begin{pmatrix} 1 \\ 0 \end{pmatrix} \)
We note however that although there can be appreciable beating in the instantaneous differential decay rate in a particular direction, for two identical oscillators, the beats disappear from the integrated instantaneous decay rate $\Gamma_c$, as we've already concluded in Section B. Here we can understand the reason for this disappearance from a different aspect. From Eq. (4-4-10) we see that the differential decay rate $d\Gamma_c/d\Omega(n)$ is proportional to $|S(n)|^2 = \frac{c}{8\pi} |E(n)|^2$. The cross terms in $|E|^2$ which produce the beats is proportional to $\cos(\frac{1}{2} k \cdot a) \sin(\frac{1}{2} k \cdot a) = \frac{1}{2} \sin(k \cdot a)\sin(kn \cdot a)$, which is an odd function [see Eq. (4-4-9)]. When we integrate over the sphere, $n$ and $-n$ directions always cancel each other. In other words, whenever the radiation intensity in one direction reaches strongest peak, its opposite direction is necessarily experiencing the weakest point. The net effect is that we see no beating in the integrated instantaneous decay rate $\Gamma_c$ and the system energy $\langle \varepsilon(t) \rangle$.

D. Validity of the Born Approximation Calculation of $\Gamma_c$

Another interesting point that we can comment on here is the fact that a Born approximation calculation of the radiative decay rate will correctly give the initial decay rate of the system energy even for the most general situations in which the instantaneous decay rate is time dependent.

Returning to Eq. (4-4-9) for the radiation field, we note that for $t'<<(\omega(\pm)-\omega_0)^{-1}$, we may ignore both the damping and the differences between $\omega'(\pm)$, $\omega'(\mp)\omega_0$ and $\omega_0$, and put Eq. (4-4-9) into

$$E = q_k_0^2 \frac{1}{\Gamma} [\hat{\mathbf{k}} \cdot (\mathbf{n} \cdot \hat{\mathbf{k}}) \mathbf{n}] \exp[-i\omega_0 t'] \left[ \xi_A \exp(-i \frac{1}{2} k \cdot a) + \xi_B \exp(-i \frac{1}{2} k \cdot a) \right]$$

(4-4-11)

What Eq. (4-4-11) in fact represents is the field coming from two identical dipole oscillators independently oscillating at the natural frequency $\omega_0$, with amplitudes $\xi_A$ and $\xi_B$. Calculating the initial decay rate $\Gamma_c$ from Eq. (3-7-3) with $E$ given by Eq. (4-4-11) is just the "first Born approximation" calculation of $\Gamma_c$, such as given in Eq. (2-5) for the radiative decay of the exciton state $|\psi_e(k_0)\rangle$. The two oscillators we have here are coupled by their radiative fields, but since the radiative coupling between them is extremely weak,
there is a relatively long period of time in which the existence of this radiative coupling can be ignored. This is the reason why we can still use the Born approximation and obtain accurate results for the initial decay.

As a typical example, if we have $|\omega^{(2)}-\omega_0|^{-1} \sim 10^{-8}$ sec, then we require $t' \ll 10^{-8}$ sec for the Born Approximation to be of good accuracy. $10^{-8}$ sec seems to be a short time, but compared to $1/\omega_0 \sim 10^{-19}$ sec, it is such a long time!

Of course, however weak, the coupling does exist and for $t'>|\omega^{(2)}-\omega_0|^{-1}$, the Born approximation [or Eq. (4-4-11)] no longer represents the correct result. At that time, we have to turn to Eq. (4-4-9) for the radiation fields.

This argument can be extended to systems of more than two identical oscillators and to systems of non-identical oscillators. The Born approximation gives an accurate calculation of the initial radiative decay rate $\Gamma_c$ over a relatively long period of time $t' < [\text{Max} |\omega^{(k)}-\omega^{(0)}|]^{-1}$.

§4-5 Two Non-identical Oscillators.

In this section, we discuss two oscillators with different natural resonance frequencies $\omega_0+\Delta$ and $\omega_0-\Delta$. We assume that $\Delta<\omega_0$ and that $\Delta < c/|R_1-R_2|$, but $\Delta$ can be large compared to $\Gamma_y$. We've already discussed the case of $N$ non-identical oscillators in §3-8. The discussion here is a direct application of that section.

(A) Normal Modes and Eigenfrequencies

From Eqs. (3-8-5), we get the equation of motion for the system of two non-identical oscillators,

\begin{align*}
(\omega_0+\Delta - \omega - i \frac{\Gamma}{2})X_1 &= \frac{\Gamma}{2} \kappa_{12} \left( e^{ik_0 a} \right) X_2 \\
(\omega_0-\Delta - \omega - i \frac{\Gamma}{2})X_2 &= \frac{\Gamma}{2} \kappa_{12} \left( e^{ik_0 a} \right) X_1.
\end{align*}

(4-5-1)

where $\kappa_{12}$ is defined the same as in Eq. (4-1-4). For the sake of simplicity in notation, we introduce

\begin{equation}
r = -\kappa_{12} \left( e^{ik_0 a} \right), \tag{4-5-2}
\end{equation}
and

\[ s = \frac{2\Delta}{\Gamma \gamma}. \]  

(4-5-3)

Then we get two eigenfrequency solutions,

\[ \omega^{(\pm)} = \omega_0 - i \frac{\Gamma_\alpha}{2} - \frac{1}{2} \Gamma \gamma \left[ \sqrt{r^2 + s^2} \pm \frac{1}{\sqrt{r^2 + s^2}} \right]. \]  

(4-5-4)

Substituting Eq.(4-5-4) into Eq. (4-5-1), we get the eigenstates corresponding respectively to \( \omega^{(\pm)} \),

\[
X^{(\pm)} = \begin{pmatrix} X_1^{(\pm)} \\ X_2^{(\pm)} \end{pmatrix} = \frac{1}{\sqrt{N^{(\pm)}}} \begin{pmatrix} r \\ -s \mp \sqrt{r^2 + s^2} \end{pmatrix},
\]  

(4-5-5)

where

\[ N^{(\pm)} = r^2 + (-s \pm \sqrt{r^2 + s^2})^2 \]

is the normalization constant.

We see from Eq. (4-5-4) the following:

1. The complex eigenfrequencies \( \omega^{(\pm)} \) satisfy the "sum rules" [Eqs. (3-8-9) and (3-8-10)]:

\[ \frac{1}{2} [\omega^{(+) + \omega^{(-})} = \omega_0 - i \frac{1}{2} (\Gamma_\alpha + \Gamma \gamma) = \omega_0 - i \frac{1}{2} \Gamma. \]

2. When \( s \ll |r| \), the eigenfrequencies and normal modes go back to the identical oscillator case. In particular, when \( s=0 \), Eq. (4-5-5) gives \( X^{(\pm)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm 1 \end{pmatrix} \) and Eq. (4-5-4) gives \( \omega^{(\pm)} = \omega_0 - i \frac{\Gamma_\alpha}{2} - \frac{1}{2} \Gamma \gamma \left[ \pm \kappa_{12} \frac{\xi \pm k_0 a}{k_0 a} \right] \), agreeing with Eqs. (4-1-3) and (4-1-2) in the identical oscillator case discussed in §4-5.

3. On the other hand, when \( s \gg |r| \), we approach the limit of \( \omega^{(\pm)} = \omega_0 - i \frac{\Gamma}{2} \pm \Delta. \)

That is, the two eigenfrequencies are approaching respectively to the natural resonance frequencies of the two oscillators. \( \omega^{(+)} \Rightarrow \omega_1 = \omega_0 \pm \Delta, \omega^{(-)} \Rightarrow \omega_2 = \omega_0 - \Delta.\)
(4) When \( s >> \| \ell \| \) the eigenmodes become, \( X^{(+)} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \) and \( X^{(-)} = \begin{pmatrix} 0 \\ -1 \end{pmatrix} \). Combining with (3), we see that as the difference in natural frequencies gets much larger than the radiative coupling, the eigenmodes become virtually the single-oscillator motion, that is, the "+" mode becomes very nearly the oscillation of the first oscillator with its own natural resonance frequency \( \omega_0 + \Delta \) while the "-" mode becomes very nearly the independent motion of the second oscillator with its natural resonance frequency. This illustrates the comments made earlier in the last two paragraphs of Chapter III where we discussed the similar situation for the system of \( N \) oscillators. Consequently, the general motion of the system becomes essentially the independent motion of the two oscillators with their individual natural resonance frequencies. The radiation of the system will be the superposition of the radiations from two such oscillators with different frequencies. Because of the strong frequency differences, there will be pronounced quantum beats in the emitted radiation at the beat frequency \( \Omega_B = |\omega^{(+)} - \omega^{(-)}| = |\omega_2 - \omega_1| \), and in contrast to the identical oscillator case, we will see that there are quantum beats in the instantaneous decay rate \( \Gamma_c(t) \).

(B) The Time Evolution of the System

Similar to the identical oscillator system, we are able to expand any arbitrary initial condition with the normal modes obtained above. The motion of the system can be most generally represented as,

\[
X(t) = A^{(+)} X^{(+)} \exp[-i \omega^{(+)} t] + A^{(-)} X^{(-)} \exp[-i \omega^{(-)} t],
\]

where \( X^{(\pm)} \) and \( \omega^{(\pm)} \) have already been expressed in Eq. (4-5-5) and Eq. (4-5-4), and \( A^{(\pm)} \) are the expansion coefficients,

\[
A^{(\pm)} = X(0) \cdot X^{(\pm)},
\]

where \( X(0) \) is the initial condition as in Eq. (4-4-1).

We give two examples of the time evolution of the system. In example 1, we let the natural resonance frequency splitting \( 2\Delta \) be small compared to the radiative coupling between the two oscillators, i.e., \( s << \| \ell \| \). In example 2, we take \( s >> \| \ell \| \). We start with the same initial condition, \( X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \) which means the first oscillator initially has a unit
displacement from its equilibrium position and the second oscillator's initial position is at the equilibrium position, and both oscillators have zero initial velocity.

Example 1: \(ka = 0.1\pi, \ s=5\). \(r = 46.17 - 0.98i, \ \Omega_\beta = 46.43\Gamma_\gamma, \ \Gamma^{(+)} = 1.9747\Gamma_\gamma \) and \(\Gamma^{(-)} = 0.0253\Gamma_\gamma\). The condition \(|r| > s\) is satisfied.

In Fig. 6, the energies of both oscillators are plotted (with \(\Gamma_\alpha\) assumed to be zero). The energies are represented by \(|X_1(t)|^2\) and \(|X_2(t)|^2\). The solid line is the energy of the first oscillator, the dashed line is that of the second oscillator.

![Graph showing energy evolution]  

**Fig. 6** Time evolution of the system for \(ka = 0.1\pi, \ s=5\), initial condition \(X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix}\). The condition \(|r| > s\) is satisfied.

We see the energy goes back and forth between oscillator 1 and 2 at the frequency \(\Omega_\beta\), showing a strong coupling between them. Because we have \(|r| >> |s|\), the radiative coupling plays a significant role in determining both the normal mode configurations \(X^{(d)}\), and the final frequency shifts of the two normal modes. Hence the case is very close to the identical oscillator situation plotted in Fig.(4a).
Example 2: $ka = 0.1\pi$, $s=1000$. $\Rightarrow r = 46.17 - 0.98i$, $\Omega_B = 1001.06\Gamma_\gamma$, $\Gamma^{(+)} = 1.045\Gamma_\gamma$ and $\Gamma^{(-)} = 0.955\Gamma_\gamma$. The condition $|r| < s$ is satisfied.

In this example, we put our parameters in such values that the natural resonance frequency difference $2\Delta$ is much bigger than the frequency shift caused by the radiative coupling. The evolution is shown in Fig. 7. In order to show the fine structure of the plot, we include two extra plots with expanded scales.

![Graphs showing the time evolution of the system for $ka = 0.1\pi$, $s=1000$, initial condition $X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$. The condition $|r| >> s$ is satisfied.]

As we discussed earlier, for $s >> |r|$, the normal mode are approximately independent single oscillator motion at the frequencies $\omega^{(\pm)} = \omega_0 \pm \Delta$. Thus the initial condition $X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ is very nearly a normal mode 1 excitation. There is very little excitation of oscillator 2, and
the first oscillator oscillates and decays almost by itself, with very little influence from oscillator 2. However, the fine structure oscillation shown in the expanded scale plots occur because $X(0)$ does contain a small admixture of mode 2, and so that $|X_1(t)|^2$ and $|X_2(t)|^2$ are modulated at the beat frequency $\Omega_B=|\omega^{(+)}-\omega^{(–)}|=2\Delta$.

In Fig. 8, we plot the system energy of the two cases. What is actually plotted is the quantity $|X_1(t)|^2+|X_2(t)|^2$. In Chapter III, we proved that this quantity is directly proportional to the mechanical energy of the system. The solid curve is for $ka=0.1\pi$, $s=5$. The dashed curve is for $ka=0.1\pi$, $s=1000$. An interesting feature is that the two case initially have the same decay rate, but subsequently, the energy lose is slower for the weak splitting $s=5$ case:

When $s=1$, $ka=0.1\pi$, things are very close to the identical oscillator system. The motion of the system consists of the "anti-symmetric mode" and the "symmetric mode". The initial condition that we have here makes the "symmetric mode" and the "anti-symmetric mode" share half the total system energy when $t=0$. But the symmetric mode has the decay rate close to $2\Gamma_\gamma$ while the anti-symmetric mode has almost zero decay rate. The net result is: (1) Initially, the decay rate of the whole system has the radiative decay rate $\Gamma_\gamma$ (2) The half energy that the symmetric mode has decays rapidly, while the antisymmetric mode tends to hold on to its share of the energy. Therefore as time goes on, the decay rate of the system falls closer and closer to the decay rate of the anti-symmetric mode, which is very small.

When $s=1000$, $ka=0.1\pi$, the motion of the system is nearly the independent motion of each oscillator. Since initially we give no energy to the second oscillator, the motion of the system is very nearly the motion of the first oscillator alone with its natural decay rate $\Gamma_\gamma$. So the system decay rate stays at the value of $\Gamma_\gamma$. Therefore we have in this case, a bigger decay rate than in the $s=1$ case.
The Energy of the System

\[ k_a=0.1\pi, \ s=5 \]

\[ k_a=0.1\pi, \ s=1000 \]

\[ \text{Time } (1/\Gamma) \]

Fig. 8 Mechanical energy of the system. Initial condition \( X(0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \).

(C) The Radiation from the System

We've discussed the radiation for a system of \( N \) oscillators in Chapter III. These equations (such as Eqs. (3-4-11), (3-4-14)) are equally applicable to the system of non-identical oscillators, as we have already argued in § 3-5. As an example, we use Eq. (3-4-11) in our two-non-identical oscillator system to study the angular distributions of the radiation from the system.

As shown in Fig. 9, we take a look at the power flow of the radiation in \( x, y, z \) three directions. In all three directions, the observing points \( A, B, C \) are assumed to be far away from the system so that the dimension of the system is neglectable. \( O_1 \) and \( O_2 \) are two oscillators moving along \( x \) direction. The difference in their natural resonance frequencies is assumed to be \( \Delta=5\Gamma \). And we assume the system's initial condition is

\[ X(0) = \begin{pmatrix} X_1(0) \\ X_2(0) \end{pmatrix} = \begin{pmatrix} 1 \\ \exp(ika) \end{pmatrix} \]
Observation Point A: In this direction, \( n = z, [\hat{\mathbf{x}} - (n \cdot \hat{\mathbf{x}}) n] = \hat{\mathbf{x}} \). From Eq. (3-6-5) with \( E \) defined in Eq. (3-4-8), we get the radiative power (Poynting flux) as the function of time for various \( ka \) values. The results are plotted in Fig. 10.

Fig. 10 Radiated intensity as function of retarded time for the system of 2 non-identical oscillators at point A. (The vertical unit is \( \frac{3}{16\pi^2} m \omega_0^2 \Gamma, \) the horizontal unit is \( 1/\Gamma \).)
When \( k_\alpha = 0.1\pi, |l| = 46.2 \gg s = 5 \). So the output radiation is modulated mainly by the radiative coupling between two oscillators. The modulation rate is \( \Omega_B = |\omega^{(+) - \omega^{(-)}| = 47.2\Gamma_\gamma \). The modulation period is \( 2\pi/(47.2\Gamma_\gamma) = 1/(7.5\Gamma_\gamma) \), so from \( t = 0 \) to \( t = 0.5(1/\Gamma_\gamma) \), we should see \( 0.5 \times 7.5 = 3.5 \) ups-and-downs, as shown on Fig. 10 (a).

On the other hand, when \( k_\alpha = 0.5\pi, \Omega_B = |\omega^{(+) - \omega^{(-)}}| = 10.2\Gamma_\gamma = 2\Delta \) (showing that the natural resonance frequency shift dominates), which corresponds to a modulation period of \( 2\pi/(10.2\Gamma_\gamma) \). So from \( t = 0 \) to \( t = 0.5(1/\Gamma_\gamma) \), we should only see \( 0.5 \times 10.2/(2\pi) = 0.8 \) ups-and-downs, as shown on Fig. 10 (b).

**Observation Point B.** In this direction, \( n = y, [\hat{x} - (n \cdot \hat{x})n] = \hat{x} \). From Eq. (3-6-5) with \( \mathbf{E} \) defined in Eq. (3-4-8), we calculate and plot the time evolution of the radiation power in that particular direction. The results are shown in the following pictures.

![Graphs](image)

**Fig. 11** Radiated intensity as function of retarded time for the system of 2 non-identical oscillators at point B (The vertical unit is \( \frac{3}{16\pi^2}m_0^2\omega_0^2\Gamma_\gamma \), the horizontal unit is \( 1/\Gamma_\gamma \))
In Fig. 11, the top-left shows the evolution of the radiation when \(ka=0.1\pi, \Delta=5\Gamma\) (\(|r|>>s\) case) The top right shows the evolution of the radiation when \(ka=-0.1\pi, \Delta=5\Gamma\). As we know, \(|k|a=0.1\pi\) and \(\Delta=5\Gamma\) gives us a relatively big frequency shift between two normal modes (\(\Omega_B=47.2\Gamma\), comparing to \(|\omega_1-\omega_2|=10\Gamma\) shows that the radiative coupling makes dominating contribution the final \(\Omega_B\)). On the other hand, the bottom left and the bottom right pictures show us the evolution of radiation power when \(ka=\pm 0.5\pi\) and \(\Delta=5\Gamma\) (\(|r|<<s\) case). The increase in \(ka\) value reduces the coupling strength between two oscillators so that now the difference in natural resonance frequencies dominates the frequency shifts of the normal modes (\(|\omega_1-\omega_2|=10\Gamma\), comparing with \(\Omega_B=|\omega^+_1-\omega^-_1|=10.2\Gamma\)). Corresponding to the above pictures, the top ones have a much greater modulating rate than the curves at the bottom.

We note that although the modulation is faster when \(|r|>>s\) than when \(|r|<<s\), yet the modulation range is smaller. We realize from this that only when in the \(|r|<<s\) case (that is, when there is appreciable difference between natural resonance frequencies), can we really expect the "turn-on and shut-off" effect, which we call "quantum beats".

The other point we have to take note here is that in both \(ka>0\) cases, we see enhanced initial radiation in this direction and in both \(ka<0\) cases, we see destructive effect. Moreover, \(ka=-0.5\pi\) gives us zero initial radiation in this direction. If we refer back to Chapter II where we discussed the initial excitation, we realize that the initial condition \(X(0)=\begin{pmatrix} X_1(0) \\ X_2(0) \end{pmatrix} = \begin{pmatrix} 1 \\ \exp(ika) \end{pmatrix} \) (\(ka>0\)) is actually the result of the interaction between the system and the electric magnetic wave pulse traveling from -y to +y direction (when \(ka<0\), that pulse travels in the opposite direction). What the results show is that the forward scattering of such a pulse is always constructive and therefore strong. In the mean time, the scattering in backward directions is generally weaker.

Observation Point C. In this direction, \([\dot{\mathbf{\hat{X}}}-\mathbf{n}\cdot\mathbf{\hat{X}}]n(\mathbf{n}\cdot\mathbf{\hat{X}})]=0\), so that there is no dipole radiation. The higher order radiation would be much weaker than the dipole radiation in other directions.

(D) Instantaneous Decay Rate

Finally, we show the instantaneous decay rate of the system of two non-identical oscillators.
The situation of two nonidentical oscillators is similar to the identical oscillator case. If the system happens to be in one of the eigenmodes, we have a true exponential decay. The decay rate remains constant all the time.

However, in general, the system excitation will not be one of the eigenmodes. In that case, we do not have a truly exponential decay. The instantaneous decay rate will be a function of time given by Eq. (3-6-17). Not only will it involve the decay rates $\Gamma^{(+)}$ and $\Gamma^{(-)}$ of the two modes, it will be modulated at the rate of $\Omega_B = |\omega^{(+)} - \omega^{(-)}|$ as well.

Starting from the initial excitation $\begin{pmatrix} 1 \\ 1 \end{pmatrix}$, we calculate and plot the instantaneous decay rates for three different situations.

**Example 1.**

The instantaneous decay rate for system of $ka=0.1\pi$, $s=2\Delta/\Gamma_\gamma=5$.

When $ka=0.1\pi$, $|r|=46.2$. So now we have $s=5$ giving $s<|r|$. As was discussed earlier in this section, this is very close to the identical oscillator case. If it were exactly identical oscillator system, we would see no modulation in the instantaneous decay rate. Here the two oscillators are not quite identical, we do see small modulation, which is what is shown in Fig. 12. Notice we have expanded the vertical scale of the plot in order to make the modulation visible.
The Instantaneous Decay Rate as the Function of Time

\( ka=0.1\pi, s=5 \)

![Graph showing the decay rate over time for a specific set of parameters](image)

**Fig. 12** Instantaneous decay rate as the function of time for \( ka=0.1\pi, s=5 \).

On the other hand, we see from Fig. 12 that the value of the instantaneous decay rate is very close to \( 2\Gamma_y \). This is because (1) the system is very close to an identical oscillator system and therefore has two normal modes very close to the symmetric mode (which has decay rate close \( 2\Gamma_y \) when \( ka \) is small) and anti-symmetric mode (which has decay rate close to zero when \( ka \) is small), (2) and that the initial excitation \( \begin{pmatrix} 1 \\ 1 \end{pmatrix} \) puts the system almost totally in the symmetric mode.

From Fig. 12, we are able to determine the modulation rate. According to Fig. 12, \( \Omega_B \) should be approximately \( 7.5 \times 2\pi \Gamma_y = 47\Gamma_y \), which agrees with the calculated \( \Omega_B = |\omega^{(+)} - \omega^{(-)}| = 46.4\Gamma_y \).
Example 2.

The instantaneous decay rate for system of $ka=0.1\pi$, $s=2\Delta/\Gamma_\gamma=50$.

Recall that $ka=0.1\pi$ gives us $|r|=46.2$. So $s=50$ gives us $|r| \sim s$, which means that the frequency shift due to the radiative coupling is roughly the same as the natural frequency split. We interpret the situation now as "the two oscillators are quite different, but not completely".

As is show in Fig. 13, we get a much more significant modulation for the instantaneous decay rate. The decay rate starts off at $2\Gamma_\gamma$ because of our special initial condition. Then it bounces back and forth between $\Gamma=2\Gamma_\gamma$ and $\Gamma<\Gamma_\gamma$. It reaches the top whenever two oscillators have the chance to get in phase again. The immediately, the "dephasing" process starts again and the decay rate drops.

For such a significant modulation, we begin to call it "beats". The whole process can be vividly viewed as if the system were a radiation source periodically turned on and off.

The modulation rate, or beats frequency is just as predicted by the calculation: $\Omega_B=(11x2x\pi/1)\Gamma_\gamma=69.1\Gamma_\gamma=(\omega^+ - \omega^-) = 68.0\Gamma_\gamma$.

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**The Instantaneous Decay Rate as the Function of Time**

$ka=0.1\pi$, $s=50$

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Fig. 13 Instantaneous decay rate as the function of time for $ka=0.1\pi$, $s=50$
Example 3.

The instantaneous decay rate for system of $ka=0.1\pi$, $s=2\Delta/G\gamma=1000$.

Fig. 14  Instantaneous decay rate as the function of time for $ka=0.1\pi$, $s=1000$

When $s=1000$, $ka=0.1\pi$, we have $|\omega^{(+)\omega^{(-)}}|=|\omega_1-\omega_2|=1000G\gamma$. The condition $s \gg |r|$ is satisfied. Now, the radiative coupling is not important any more. The system becomes virtually the independent motion of two radiation sources with different frequencies. As is shown in Fig. 11, the instantaneous decay rate periodically bounces between $\Gamma = 2G\gamma$ (when two oscillators are moving in phase) and $\Gamma = 0$ (when they are 180 degrees out of phase). It is like a turning on and shutting off process.
The modulation rate is, according to Fig. 14, $\Omega_B = (6.5 \times 2 \pi / 0.04) \Gamma_\gamma = 1007 \Gamma_\gamma$, which agrees with the estimated $\Omega_B = 2 \Delta = 1000 \Gamma_\gamma$.

From these examples, we show in detail with the two-oscillator system, how we construct the normal modes, get the eigenfrequencies and then, solve the motion of such a system under general initial conditions. In the next Chapter, we will try to study the system of $N$ coupled oscillators.
V. Crystalline Arrays of Identical Oscillators

In this chapter, we discuss the system of N coupled identical oscillators arranged in two and three dimensional crystalline arrays. We use the general development of Chapter III to obtain the radiative normal modes, the complex eigenfrequencies and the normal mode radiation fields.

We first solve for a 2-dimensional array. The normal mode excitations are shown to be the two dimensional Bloch waves with a phasing factor $e^{i\mathbf{q}\cdot\mathbf{R}_j}$, and the radiation is emitted into a finite number of plane waves forming symmetric pairs of channels "above" and "below" the plane. The concept of opened and closed channels is discussed, and we discuss the strong enhancement that occurs if a mode $\mathbf{q}$ has a channel which is "just opened".

With the 2-dimensional array as our base, we move on to systems of 2 and 3 layers, and set up the coupled equations between the different plane layers. The eigenmodes are obtained using the direct determinant approach.

A more general approach is used to solve for systems of M plane layers. Here the general coupled equations are first simplified to a 2-wave approximation, consisting of the "transmission" and "reflection" channels which make an angle $\phi$ relative to the crystal planes. These equations give M radiative normal modes which have either a sine or cosine modulation coming in from the crystal faces, with a complex wave vector $k'$. As illustrative examples, we solve for $M=3$ and $M=10$. The mode frequency shifts $\Delta\omega^{(n)}$ and radiative decay widths $\Gamma^{(n)}$ are plotted as a function of $\phi$. For $\phi$ satisfying a Bragg angle for $\omega_0$ radiation, only one mode is superradiant with width

$$\Gamma^{(n)} = \Gamma_c = \Gamma_c 2\pi n^2 k^2 L_{||}(\phi),$$

(5-1)

and the frequency shift $\Delta\omega^{(n)}=0$. The remaining M-1 modes are non-radiative. When $\phi$ is off-Bragg, all M modes are radiative although most of them are subradiant with $\Gamma^{(n)}<<\Gamma_r$. The frequencies of all modes are shifted either above or below $\omega_0$, obeying the sum rule $\sum_n \Delta\omega^{(n)}=0$, while the widths obey the sum rule $\frac{1}{M} \sum_n \Gamma^{(n)} = \Gamma_0$, where $\Gamma_0$ is the the radiative decay width of a single layer for decay into two open channels, $k_0^{(\pm)}$. 


We then return to consider the single exciton states \( |\Psi_e(k_0)\rangle \) created by synchrotron radiation pulses. We expand \( |\Psi_e(k_0)\rangle \) as a superposition of normal mode states, solve for the subsequent time dependence of the state of the system and for the emitted radiation fields, and examine the nature of the superradiant decay. We will show that if the phasing vector \( k_0 \) is on-Bragg with respect to the crystal planes, then the system is excited into a single superradiant eigenmode. The subsequent emission goes symmetrically into the forward and the "Bragg reflection" directions, with enhanced decay rates \( \Gamma_c(\text{Bragg}) \) given by Eq. (5-1). For \( k_0 \) off-Bragg, more than one eigenmode is excited, and the subsequent development is superradiant nonexponential decay into the forward \( k_0 \)-direction, with an initial decay rate \( \Gamma_c(\text{off-Bragg}) = \frac{1}{2} \Gamma_c(\text{Bragg}) \), consistent with the Born approximation calculation, Eq. (1-6). An interesting point here is that for large \( M \), \( \Gamma_c(\text{off-Bragg}) \) greatly exceeds any of the eigenmode decay rates \( \Gamma^{(n)} \), so that it is the nonorthogonality of the eigenmodes and the spread of eigenfrequencies which determine the superradiant decay of \( |\Psi_e(k_0)\rangle \) off-Bragg.

§ 5-1 2-Dimensional Array.

We first consider a 2-dimensional square array system. The lattice constant is \( a \) and the equilibrium positions of the oscillators are \( R_i = n_1 a \hat{x} + n_2 a \hat{y} \). We assume that all the oscillators are restricted to move the \( \hat{x} \) direction in the plane. Again, we consider the radiative interaction between individual oscillators and look for the radiative normal modes.

(A). Eigenmodes

For normal mode motion of the system, the equation of motion for the \( j \)th oscillator is

\[
(i\omega - \omega - \frac{\Gamma}{2}) X_j = -\frac{q^2}{2m\omega_0} \hat{x} \cdot \sum_{i \neq j} \left\{ \nabla_j \nabla_j^* \exp(-ik_0|R_i - R_j|) \frac{\exp(-ik_0|R_i - R_j|)}{|R_i - R_j|} \right\} X_i,
\]

as was given by Eq. (3-1-11). In principle, we can get the eigensolutions by solving the associated determinant equation, just as we did in Chapter III. But that is impractical and not necessary because the 2-D translational symmetry dictates that the eigenmodes should be 2-D Bloch waves.
(1) Radiation from a Bloch Wave State

Assume a Bloch wave solution,

\[ X_i^{(q)}(t) = X^{(q)} \exp(iq \cdot r_i) \exp(-i\omega t), \tag{5-1-1} \]

where \( q \) is a vector in the x-y plane, \( X_i^{(q)}(t) \) is the displacement of the \( i \)th oscillator and \( \omega = \omega^{(q)} \) is the frequency associated with the vector \( q \). We will later prove by consistency that the above form is indeed a normal mode.

For a 2-D array of oscillators undergoing the motion in the Bloch mode \( q \), we've calculated in Appendix III the field at the observation point \( R = z\hat{z} + \rho \) (where \( \rho = x\hat{x} + y\hat{y} \) is the projection of \( R \) in the plane),

\[
E^{(q)} = e \sum_i \nabla_x \left( \nabla_x \frac{\exp(-ik_0 |R - R_i|)}{|R - R_i|} \hat{x} \right) X_i^{(q)} \\
= \sum_{\mu} E^{(q)}_{\mu} \\
= -i \frac{2\pi e}{a^2} X^{(q)} \exp(-i\omega t) \sum_{\mu} \exp[ig_{\mu} |z| + i(\tau_\mu + q) \cdot \rho] \frac{k^{(\pm)}_\mu \times (k^{(\pm)}_\mu \times \hat{x})}{g_{\mu}}, \tag{5-1-2}
\]

where

\[ g_{\mu} = \sqrt{k_0^2 + (\tau_\mu + q)^2}, \tag{5-1-3} \]

\[ \tau_\mu = \frac{2\pi}{a} (m\hat{x} + n\hat{y}) \tag{5-1-4} \]

and

\[ k^{(\pm)}_\mu = \pm g_{\mu} \hat{z} + (\tau_\mu + q). \tag{5-1-5} \]

In the above expressions, we take + for \( z > 0 \) and – for \( z < 0 \), (see Fig. 15). The \( \tau_\mu \) is the set of planar reciprocal lattice vectors for the square lattice. The factor \( g_{\mu} \), which gives the
component of the wave vector $k_{\mu}^{(\pm)}$, is real if $|\tau_\mu + q| < k_0$, and is to be taken positive imaginary if $|\tau_\mu + q| > k_0$.

We see from Eq. (5-1-2) that the field consists of the sum of various components ($E_\mu$) corresponding to different reciprocal vectors $\tau_\mu$:

![Diagram of radiation from a 2-D array](image)

**Fig. 15.** Radiation from a 2-D array.

- Plane wave channels.

Those $\tau_\mu$ for which $|\tau_\mu + q| < k_0$ give a real $g_\mu$. Consequently those $E_\mu$'s are true plane waves. Each such $\tau_\mu$ gives two plane waves coming off symmetrically "above" (-z) and "below" (+z) the plane in the directions of $k_{\mu}^{(+)}$ and $k_{\mu}^{(-)}$ respectively, which each makes an angle of $\phi_\mu = \sin^{-1}(g_\mu/k_0)$ with respect to the x-y plane, as indicated schematically in Fig. 15. We call each of these a "plane wave channel" or an "open channel". There are finite number of $\tau_\mu$ that satisfy this condition, which lead to finite number of open channels. These channels contribute to the energy decay and therefore the damping rate of the system in mode $q$.

- Exponentially decaying channels.

Those $\tau_\mu$ for which $|\tau_\mu + q| > k_0$ give positive imaginary $g_\mu$. These $E_\mu$'s symmetrically exponentially decay in the $\pm \hat{z}$ directions, and we refer to these as "closed channels". There are infinite number of $\tau_\mu$'s that satisfy this condition. As we will see
later, the closed channels do not contribute to the damping, but do contribute to the the frequency shift of mode $q$.

(2) Equation of Motion for the jth Oscillator.

Now consider the equation of motion of the jth oscillator assuming the system is in the normal mode motion of the form expressed by Eq. (5-1-1). We have,

$$m \dddot{x}^{(q)}_j + m\omega_0^2 x^{(q)}_j + m\Gamma \dot{x}^{(q)}_j = e \hat{\Phi} \cdot E^{(q)}_j = e \hat{\Phi} \cdot \sum_i E^{(q)}_{ij}, \quad (5-1-6)$$

where $\sum'_i$ means the sum over i's that are not equal to j, and according to Eq. (3-1-5),

$$E^{(q)}_{ij} = e \hat{\Phi} \cdot \nabla x \{ \frac{\exp(-ik_0 |R_j - R_i|)}{|R_j - R_i|} \hat{\Phi} \} X^{(q)}_i.$$  

In order to be able to use the planar sum of Eq. (5-1-2), we must add and subtract the field contribution from the jth oscillator: We first take the result $E^{(q)}(r)$ of Eq. (5-1-2) in the vicinity (that is, at $R=R_j+r$) of the jth oscillator, which includes the field oscillator j creates. Then, we subtract the dipole field $E^{(q)}_{rj}$ from the jth oscillator at that point

$$E_{rj}^{(q)} = e \nabla x \{ \frac{\exp(-ik_0 |r|)}{|r|} \hat{\Phi} \} X^{(q)}_j. \quad (5-1-7)$$

Taking the limit of $r \to 0$, we get the field the rest of the system creates at the site of the jth oscillator,

$$\sum_i E^{(q)}_{ji} = \lim_{r \to 0} [ \sum_i E^{(q)}_{ri} - E^{(q)}_{rj} ] = \lim_{r \to 0} [E^{(q)}(r) - E^{(q)}_{rj}], \quad (5-1-8)$$

where now we use the result of Eq. (5-1-2) in $\sum_i E^{(q)}_{ri} = \sum_{\mu} E^{(q)}_{\mu}(r) = E^{(q)}(r)$. The equation of motion thus becomes,

$$m \dddot{x}^{(q)}_j + m\omega_0^2 x^{(q)}_j + m\Gamma \dot{x}^{(q)}_j = e \hat{\Phi} \cdot \lim_{r \to 0} [E^{(q)}(r) - E^{(q)}_{rj}]. \quad (5-1-9)$$
For \( r \to 0 \), the dipole field \( E_{rj}^{(q)} \) is the near field solution given by Eq. (4-1-12),

\[
E_{rj}^{(q)} = k_0^3 \left( \frac{1}{k_0 r} \right)^3 \left[ 3n(n \cdot d_j^{(q)}) - d_j^{(q)} \right] + \frac{1}{2k_0 r} \left[ n(n \cdot d_j^{(q)}) + d_j \right] + \frac{2}{3} i d_j^{(q)},
\]

(5-1-10)

where \( n = r/|r| \) and \( d_j^{(q)} = e X_j^{(q)} \). It contains three contributions, the first two of which give the field in phase with \( X_j^{(q)} \),

\[
E_{rj}^{(q)\text{(in phase)}} = ek_0^3 \left( \frac{1}{k_0 r} \right)^3 \left[ 3n(n \cdot \hat{k}) - \hat{k} \right] + \frac{1}{2k_0 r} \left[ n(n \cdot \hat{k}) + \hat{k} \right] X_j^{(q)},
\]

(5-1-11)

which gives rise to a divergent frequency shift as \( r=|r| \) goes to zero and it depends on the path of \( r \) relative to \( \hat{k} \) when the limit is taken. The last term in Eq. (5-1-10) gives us a field \( 90^0 \) out of phase with \( X_j^{(q)} \),

\[
E_{rj}^{(q)\text{(90^0 out of phase)}} = -\frac{m}{e} \Gamma \dot{X}_j^{(q)} \hat{k},
\]

(5-1-12)

which gives a damping force on the \( j \)th oscillator. Note that this term is independent of \( r \), and substituting Eq. (5-1-12) back into Eq. (5-1-9), it subtracts from the \( \Gamma \) term on the left hand side of Eq. (5-1-9). The result is that \( \Gamma \) is replaced by \( \Gamma - \Gamma = \Gamma_\alpha \).

The force equation of the \( j \)th oscillator can then be written as,

\[
m \ddot{X}_j^{(q)} + m \omega_0^2 X_j^{(q)} + m \Gamma_\alpha \dot{X}_j^{(q)} = e \hat{k} \cdot \lim_{r \to 0} [E^{(q)}(r) - E_{rj}^{(q)\text{(in phase)}}].
\]

(5-1-13)

(3) Normal Mode Equation of Motion.

With the substitution of \( X_j = X_j^{(q)} = X^{(q)} \exp(iq \cdot R_j) \), and with \( E^{(q)}(r, t) \) given by Eq. (5-1-2), the left hand side and the right hand side of Eq. (5-1-13) are both proportional to \( \exp(iq \cdot R_j) \), verifying the Bloch wave nature of the eigenmode, and the normal mode equation can be put into the "Hamiltonian form" (for more details, see Appendix 4)
\[ \hbar^{(q)} \chi^{(q)} = \omega \chi^{(q)}, \quad (5-1-14) \]

where

\[ \hbar^{(q)} = \omega_0 + \delta \omega^{(q)} - i \frac{1}{2} (\Gamma^{(q)} + \Gamma^{(q)},) \quad (5-1-15) \]

where \( \Gamma^{(q)}_c \) gives the the radiative decay rate for the Bloch mode \( q \),

\[ \Gamma^{(q)}_c = \sum_{\mu<} \Gamma^{(q)}_{\mu} \quad (5-1-16) \]

where

\[ \Gamma^{(q)}_{\mu} = 3 \pi n_{(2)} \frac{\hbar^2}{\epsilon^2} \frac{k_0^2 - (\tau_{\mu} + q_x)^2}{k_0 \epsilon_{\mu}} \Gamma_\gamma \quad (5-1-17) \]

gives the radiative decay rate due to the emission into the two open channels \( k_{\mu}^{(2)} \). (Note for \( \mu<, \Gamma_{\mu} > 0 \).) Here \( n_{(2)} = 1/a^2 \) is the planar density of oscillators and

\[ \delta \omega^{(q)} = \lim_{r \to 0} [S^{(q,\mu)}(r) - S_0(r)], \quad (5-1-18) \]

where

\[ S^{(q,\mu)}(r) = \frac{e}{2m\omega_0} \hat{\mathbf{r}} \cdot \mathbf{E}^{(q,\mu)}(r) \]

\[ = \frac{3}{2} \pi n_{(2)} \frac{\hbar^2}{\epsilon^2} \Gamma_\gamma \sum_{\mu>} \frac{(	au_{\mu} + q_x)^2 - k_0^2}{|\epsilon_{\mu}|} \exp[-|\epsilon_{\mu}| r^2 + i (\tau_{\mu} + q_x) \cdot \mathbf{r}] \quad (5-1-19) \]
gives the "planar field self-energy", and

\[ S_0(r) = \frac{e}{2m\omega_0} \hat{A} \cdot \mathbf{E}_r^{(q)} \text{(in phase)} \]

\[ = -\frac{3}{4} \Gamma_\gamma \left\{ \frac{1}{(k_0r)^2} [3(\mathbf{n} \cdot \hat{A})^2 - 1] + \frac{1}{2k_0r} [(\mathbf{n} \cdot \hat{A})^2 + 1] \right\} \quad (5-1-20) \]

gives the "dipole self-energy".

We note that both \( S^{(q,\mu)}(r) \) and \( S_0(r) \) are divergent as \( r \) goes to zero, but the difference of the two divergent quantities \( \delta \omega^{(q)} \) must be finite. Furthermore, \( \delta \omega^{(q)} \) must be independent of the path taken in the limit \( r \to 0 \) (although it is not at all obvious that this is true since \( S_0(r) \) is clearly path dependent as discussed in §4-1-B). Assuming path independence, then the limit could also be taken by first averaging \( S_0(r) \) and \( S^{(q,\mu)}(r) \) over a spherical surface, and then letting the radius \( \mathbf{r} \to 0 \). The \( d\Omega \) average over \( S_0(r) \) is particularly simple,

\[ <S_0(r)> = \frac{\Gamma_\gamma}{2} \left( \frac{1}{k_0r} \right). \quad (5-1-21) \]

But \( S^{(q,\mu)}(r) \) does not appear to be that simple. We will not pursue the evaluation of \( \delta \omega^{(q)} \) further in this thesis, but plan to do so later. For our purpose now, it suffices to note that the resonance frequency for the mode \( q \) is shifted,

\[ \omega_0 \to \omega_0^{(q)} = \omega_0 + \delta \omega^{(q)}, \quad (5-1-22) \]

and that \( \delta \omega^{(q)} \) will generally be small, \( \delta \omega^{(q)} < (1-10^3)\Gamma_\gamma \ll \omega_0 \).

Returning to the first normal mode equation of motion Eq. (5-1-14), this can also be written as,

\[
Q(\omega) X^{(q)} = \frac{1}{2} \sum_{\mu} \Gamma_\mu^{(q)} X^{(q)}, \quad (5-1-23)
\]

where

\[
Q(\omega) = \omega_0 - \omega - i \frac{1}{2} \Gamma_\alpha, \quad (5-1-24)
\]
where \( \omega_0 = \omega_0 - S_0(r) \) (\( S_0 \) and \( \frac{1}{2} \sum_{\mu} \Gamma_{\mu}^{(q)} \) combine to give \( \delta \omega^{(q)} \)). We will find this form more convenient when we extend to muti-layer systems.

We also note for future reference that

\[
\frac{e}{2m\omega_0} \hat{\mathbf{x}} \cdot E^{(q)}(r,t) = \frac{1}{2} i X^{(q)} \exp[-i\omega^{(q)} t] \sum_{\mu} \Gamma_{\mu}^{(q)} \exp[i(k_{\mu}^{(\pm)} \cdot \mathbf{r})],
\]

(5-1-25)

where we have used \( \hat{\mathbf{x}} \cdot [k_{\mu}^{(\pm)} \times (k_{\mu}^{(\pm)} \times \hat{\mathbf{x}})] = (\tau_{\mu \alpha} + q_{\alpha})^2 - k_0^2 \).

(4) Normal Mode Frequencies.

Returning to Eqs. (5-1-14) and (5-1-15), we see that the complex eigenmode frequency is

\[
\omega^{(q)} = \omega^{(q)} - i\omega^{(q)} = \omega_0 + \delta \omega^{(q)} - i \frac{1}{2} (\Gamma^{(q)}_\alpha + \Gamma^{(q)}_c).
\]

(5-1-26)

The mode frequency is shifted to,

\[
\omega^{(q)} = \omega_0 + \delta \omega^{(q)},
\]

(5-1-27)

and the decay rate \( \Gamma^{(q)} \) is

\[
\omega^{(q)} = 2 \omega^{(q)} = \Gamma^{(q)}_\alpha + \Gamma^{(q)}_c.
\]

(5-1-28)

(5) Sum Rules

In Chapter III, we derived the general sum rules for the normal mode frequencies and radiative decay rates, given by Eqs. (3-2-5) and (3-2-7). For our 2-D crystalline array, the sum rules become,

\[
\sum_q \delta \omega^{(q)} = 0,
\]

(5-1-29)

and
\[ \lim_{N \to \infty} N \Gamma_c^{(q)} = \Gamma_\gamma \quad , \]

(5-1-30)

where \( N \) is the number of oscillators in the square lattice (and hence the number of normal modes in the system). The sum is over the Bloch wave normal mode vector \( q \). As discussed in the next section, the \( q \)'s uniformly fill the first Brillouin zone, with the density \( \frac{L}{2\pi} \). Thus the normal mode sum rules become,

\[
\int_{-\pi/a}^{+\pi/a} dq_x \int_{-\pi/a}^{+\pi/a} dq_y \delta \omega^{(q)} = 0 ,
\]

(5-1-31)

and,

\[
\frac{1}{(2\pi a)^2} \int_{-\pi/a}^{+\pi/a} dq_x \int_{-\pi/a}^{+\pi/a} dq_y \Gamma_c^{(q)} = \Gamma_\gamma
\]

(5-1-32)

As before, the frequency sum rule says that the mode are distributed above and below \( \omega_0 \), with average being zero. The decay rate sum rule shows that either all modes have radiative decay rates \( = \Gamma_\gamma \), or else a few modes \( q \) are superradiant with \( \Gamma_c^{(q)} >> \Gamma_\gamma \) and the remaining modes are subradiant with \( \Gamma_c^{(q)} << \Gamma_\gamma \), such that the average over all modes is still \( \Gamma_\gamma \). As we'll discuss below, it is the later alternative that holds: most modes are subradiant but there will be some superradiant modes \( q \) which contain "just opened channels" for which \( \Gamma_c^{(q)} >> \Gamma_\gamma \).

(6) Allowed Bloch Vector \( q \)

For a finite 2-D array, the normal modes will be standing waves rather than travelling Bloch wave \( \exp(iq \cdot r) \). From symmetry, we would expect that in a normal mode, symmetrically arranged oscillators on the opposite edges of the 2-D array will be in phase or \( 180^0 \) out of phase. Thus wave vectors associated with the standing wave must satisfy the \( \frac{\lambda}{2} \) condition \( q_x = n_{xL} \frac{\pi}{L} \) and \( q_y = n_{yL} \frac{\pi}{L} \), with \( n_x , n_y = 0, 1, 2, ..., \sqrt{N}-1 \). In the limit
of large $L$, this gives a continuum of states filling the $q$-space quadrant ($q_x, q_y \approx 0$ to $\frac{\pi}{a}$) with mode density $\rho(q)dq_x dq_y = \frac{(L)^2}{\pi} dq_x dq_y$ or $\rho(q)dq = \frac{1}{4}(L)^2 2\pi q dq = \frac{L^2}{2\pi} q dq$.

Alternatively, for $L\to\infty$, the modes can be treated as travelling Bloch waves which as usual means taking the periodic boundary conditions on the finite array, giving

$$q_x = n_x \frac{2\pi}{L}, \quad (n_x = -\frac{\sqrt{N}}{2}, -\frac{\sqrt{N}}{2} + 1, ..., +\frac{\sqrt{N}}{2} - 1)$$

(5-1-33)

and

$$q_y = n_y \frac{2\pi}{L}, \quad (n_y = -\frac{\sqrt{N}}{2}, -\frac{\sqrt{N}}{2} + 1, ..., +\frac{\sqrt{N}}{2} - 1).$$

(5-1-34)

The $q$'s uniformly fill the first Brillouin zone with mode density $\rho(q) = \frac{(L)^2}{2\pi}$.

(7) Normal Mode Orthogonality

In Chapter III, we showed that for a finite system, the eigenvectors of the radiative normal modes are transpose orthogonal (rather than Hermitian orthogonal),

$$(X^{(k)})^T X^{(l)} = \delta_{kl}.$$  

(5-1-35)

For our infinite 2-D array we have found that the eigenmodes are Bloch waves

$$X_i^{(q)} = X^{(q)} \exp(iq \cdot r_i),$$

(5-1-36)

or in the vector form,

$$X^{(q)} = X^{(q)} \begin{pmatrix} \cdot \\ \cdot \\ \cdot \end{pmatrix} \exp(iq \cdot r_i)$$

(5-1-37)

These vectors are in fact Hermitian orthogonal. The product of Eq. (5-1-35) gives us,

* Here we have used the relation,
\[(X^{(q)})^T X^{(q')}\]
\[= X^{(q)} X^{(q')} \sum_{j=1}^{N \to \infty} \exp[i(q+q') \cdot r_j] = X^{(q)} X^{(q')} \left(\frac{2\pi}{a}\right)^2 \delta(q+q'), \quad (5-1-38)\]

rather than \(\delta(q-q')\). However we note that the state \(q\) is degenerate with the state \(-q\), since the replacement \(q \to -q\) and \(\tau_{\mu} \to -\tau_{\mu}\) leave \((q+\tau_{\mu})^2\) unchanged, so that \(\Gamma_{\mu}^{(-q)} = \Gamma_{\mu}^{(q)}\), and consequently \(\omega^{(-q)} = \omega^{(q)}\). Thus we can take the standing wave wave combination,

\[X^{c(q)}_j = \frac{X^{(q)}_j}{\sqrt{2}} \left( e^{iq \cdot r_j} + e^{-iq \cdot r_j} \right) \quad (5-1-39)\]
\[X^{c(q')}_j = \frac{X^{(q')}_j}{\sqrt{2}} \left( e^{iq \cdot r_j} - e^{-iq \cdot r_j} \right),\]

which are transpose orthogonal. For most purposes however, it will be convenient to use the travelling wave solutions, and then the planar modes orthogonality condition is,

\[(X^{(-q)})^T X^{(q')} = X^{(-q)} X^{(q')} \left(\frac{2\pi}{a}\right)^2 \delta(q-q'), \quad (5-1-40)\]

or equivalently, Hermitian orthogonality,

\[(X^{(q')})^\dagger X^{(q)} = |X^{(q)}|^2 \left(\frac{2\pi}{a}\right)^2 \delta(q-q'). \quad (5-1-41)\]

We will usually take the normalization factor \(X^{(q)}\) as

\[X^{(q)} = \frac{1}{\sqrt{N}},\]

where \(N\) is now the number of oscillators in the \(L \times L\) 2-D lattice, and the \(q\) satisfy the boundary condition. The normalized Bloch wave vector solutions are,

\[\sum_j \exp(iQ \cdot r_j) = \left(\frac{2\pi}{a}\right)^2 \sum_{\mu} \delta(Q - \tau_{\mu}).\]

Since \(q\) and \(q'\) are in the first Brillouin zero. So only \(\tau_{\mu} = 0\) channels contribute.
\[ X^{(q)} = \frac{1}{\sqrt{N}} \begin{pmatrix} \cdot \\ \cdot \\ \cdot \\ \cdot \end{pmatrix} \exp(iq \cdot r_i) \]

and the orthogonality condition becomes,

\[ (X^{(q')}\dagger) \cdot X^{(q)} = (X^{(-q')}\dagger) \cdot X^{(q)} = \left(\frac{2\pi}{L}\right)^2 \delta(q-q'). \] (5-1-43)

We note that because Bloch modes are Hermitian orthogonal, there will be no normal mode beats \((\Omega_B(q,q') = \omega^{(q)} - \omega^{(q')})\) in the total energy \(\langle E(t) \rangle = \frac{1}{2} m \omega_0^2 |X(t)|^2\) or in the instantaneous decay rate \(\Gamma(t) = -\frac{1}{\langle E(t) \rangle} \left(\frac{d\langle E(t) \rangle}{dt}\right)\).

(8) A Special Case: Just Opened and Just Closed Channels

\(|\tau_\mu + q| < k_0\) gives open channels and \(|\tau_\mu + q| > k_0\) gives closed channels. On the verge of opening or closing a channel, we have \(|\tau_\mu + q| = k_0\). This makes that particular channel especially important. If it is a "just opened" channel, it will give a large decay rate due to the denominator \(g_p\) going to zero [see \(\Gamma_\mu^{(q)}\) in Eq. (5-1-16)]. For the same reason, if it is a "just closed" channel, it will give a large frequency shift.

Let us look at an example of eigenmode \(q\) for which \(|q| < k_0\), with \(q \perp \hat{x}\). In Eq. (5-1-16), we drop all other terms except \(\tau_\mu = 0\) because now the \(\tau_\mu = 0\) channels \(k_0^{(\pm)}\) are just opened channels and they carry a large weight in the summation. The resulting width contribution from these channels is

\[ \Gamma_0^{(q)} = \frac{3\pi n(2)^2}{\sin \phi_0} \Gamma_{\gamma}. \] (5-1-44)
where $\sin \phi_0 \frac{q}{k_0} = \sqrt{1 - \left( \frac{|q|}{k_0} \right)^2}$. $\phi_0$ gives the (grazing) angle of $k^{(\pm)}_0$ with respect to x-y plane. Thus as $|q|$ goes to $k_0$, then $\phi_0$ goes to 0 and $\Gamma_0^{(q)}$ goes to $\infty$. For $|q| \approx k_0$ on the other hand, $\Gamma_0^{(q)}$ becomes imaginary and contributes a large normal mode frequency shift.

(B). General Excitations

A general initial excitation $X(0)$ of the 2-D array can be expanded in the Bloch normal modes,

$$X(0) = \sum_q A^{(q)} X^{(q)},$$

where the expansion coefficient is

$$A^{(q)} = (X^{(q)})^\dagger X(0) = (X^{(-q)})^T X(0).$$  \hfill (5-1-45)

The subsequent time development is then,

$$X(t) = \sum_q A^{(q)} X^{(q)} \exp[-i\omega^{(q)} t],$$  \hfill (5-1-46)

and the radiation field is a superposition of the normal mode fields,

$$E(r, t) = \sum_q A^{(q)} E^{(q)}(r, t).$$  \hfill (5-1-47)

Here $E^{(q)}(r, t)$ is the normal mode field given by Eq. (5-1-2).

§ 5-2. Two-Layer Crystalline Films

We now consider crystalline films of two layers of the 2-D arrays with a planar separation $d$. This is shown schematically in Fig. 16.
Fig. 16 Two-layer system.

(A). Normal Modes

The normal modes will be of the form*,

\[ X_{j(m)}^{(q,l)}(t) = X_{m}^{(q,l)} \exp[i(q \cdot r_{j(m)} - \omega t)], \]  

(5-2-1)

which represents the motion of the jth oscillator in the mth layer (m=0, 1) in the mode (q,l). Here q is the discrete set of planar Bloch mode vectors, and l represents the extra degrees of freedom brought in by the extra plane, l=1, 2 (or +, − for two layers). \( \omega = \omega^{(q,l)} \) is the complex eigenfrequency for normal mode (q,l). \( X_{m}^{(q,l)} \) is the amplitude of the oscillation of the mth layer in the mode (q,l). And \( r_{j(m)} \) is the position of the jth oscillator in the mth layer. We put the normal mode of the system into the vector form [see Eq. (5-1-42)],

\[ X_{m}^{(q,l)} = \frac{1}{\sqrt{N}} X_{m}^{(q,l)} \begin{pmatrix} \vdots \\ \exp[iq \cdot r_{j(m)}] \\ \vdots \end{pmatrix}, \]  

(5-2-2)

where N is the number of oscillators in the plane and the vector has N components.

* This form is dictated by the invariance of the system under translation by the planar lattice vectors \( R_{ij} = a(i\hat{x} + j\hat{y}) \).
(B). Equation of motion

Each oscillator is now coupled to both the field from its own plane and the field from the adjacent plane. Assuming normal motion of the form in Eq. (5-2-1) and using the result of Eq. (5-1-23), we immediately get the normal mode equation of motion for the system,

$$Q(\omega)X^{(q,1)}_m = \sum_\mu \frac{i}{2} \Gamma^{(q)}_\mu \sum_{n=0}^1 X^{(q,1)}_n \exp(i \omega_\mu n \cdot \text{mld}),$$  \hspace{1cm} (5-2-3)

where $Q(\omega)$ was given by Eq. (5-1-24). With this done, we have proved that the form we assumed in Eq. (5-2-1) is indeed the normal mode. Alternatively, Eq. (5-2-3) can be put into the effective "Hamiltonian" form,

$$\bar{h}^{(q)}_\mu X^{(q,1)} = \omega X^{(q,1)},$$ \hspace{1cm} (5-2-4)

where

$$X^{(q,1)} = \begin{pmatrix} X^{(q,1)}_0 \\ X^{(q,1)}_1 \end{pmatrix}$$ \hspace{1cm} (5-2-5)

and

$$\bar{h}^{(q)}_{mn} = \omega_0 + \delta^{(q)}_\omega \frac{1}{2} i (\Gamma^\alpha + \sum_{\mu<} \Gamma^{(q)}_\mu), \hspace{1cm} (m=0, 1)$$ \hspace{1cm} (5-2-6)

$$\bar{h}^{(q)}_{mn} = -\frac{1}{2} \sum_{\mu} \Gamma^{(q)}_\mu \exp(i \omega_\mu n \cdot \text{mld}). \hspace{1cm} (m\neq n)$$ \hspace{1cm} (5-2-7)

Here we note that for "$\mu<$", $\Gamma_\mu$ and $g_\mu > 0$; for "$\mu>$", $\Gamma_\mu = i \|\Gamma_\mu\|$ and $g_\mu = i |\Gamma_\mu|$. Also note that $\bar{h}^{(q)}_{mn} = \bar{h}^{(q)}_{nm}$ so that the effective "Hamiltonian" matrix $\bar{h}^{(q)}$ is a 2x2 symmetric matrix, which makes $X^{(q,1)}$ transpose orthogonal.
(C). Dispersion Relation

The normal mode dispersion equation is,

\[
\text{Determinant } \left[ H^{(q)} - \omega I \right] = 0, \quad (5-2-8)
\]

which immediately gives,

\[
\omega^{(q, \pm)} = \omega_0 + \delta \omega^{(q)} - \frac{1}{2} \left( \Gamma_\alpha^+ \sum_{\mu<} \Gamma^{(q)}_{\mu} + \frac{1}{2} \sum_{\mu} \Gamma^{(q)}_{\mu} \exp(i \varepsilon_{\mu} d) \right). \quad (5-2-9)
\]

 Breaking the above into real and imaginary parts \((\omega = \omega' - i \omega'')\), we get the normal mode frequencies,

\[
\omega^{(q, \pm)} = \omega_0 + \delta \omega^{(q)} \pm \frac{1}{2} \sum_{\mu<} \Gamma^{(q)}_{\mu} \sin(\varepsilon_{\mu} d) \pm \frac{1}{2} \sum_{\mu>} \Gamma^{(q)}_{\mu} \exp(-|\varepsilon_{\mu}| d), \quad (5-2-10)
\]

and the normal mode damping rates \([\omega'\Gamma^{(q, \pm)} = \frac{1}{2} \Gamma^{(q, \pm)}]\),

\[
\Gamma^{(q, \pm)} = \Gamma_\alpha^+ \sum_{\mu<} \Gamma^{(q)}_{\mu} [1 \pm \cos(\varepsilon_{\mu} d)]. \quad (5-2-11)
\]

From Eq. (5-2-11), we see that for two-layer case, just as for a single layer, "\(\mu<\)" channels (open channels) contribute to the total decay rates. However, the rate is modulated by an extra factor \([1 \pm \cos(\varepsilon_{\mu} d)]\). This factor can be easily explained once we have seen the mode configuration in the next section.

For the frequency shifts [Eq. (5-2-10)], we see that in contrast to the single layer case, the "open channels" (the \(\sum\) terms) now enter the frequency shift as well as the closed channels [the latter contribution is primarily from \(\delta \omega^{(q)}\) because the last term in Eq. (5-2-10) drops off quickly due to the exponential decay factor \(\exp(-|\varepsilon_{\mu}| d)\)].

(D). Normal Mode Configuration

With the eigenfrequencies \(\omega^{(q, \pm)}\) given by Eq. (5-2-9), the corresponding eigenmodes are [from Eq. (5-2-8)],
\[ X^{(q,\pm)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm 1 \end{pmatrix} \]  \hspace{1cm} (5-2-12)

If we give the normal mode motion explicitly, we have for layer zero \((m=0)\),
\[ X_{j(0)}^{(q,\pm)}(t) = \frac{1}{\sqrt{2N}} \exp[i(q \cdot r_{j(0)} - \omega^{(q,\pm)} t)] \], \hspace{1cm} (5-2-13)

and for layer one \((m=1)\),
\[ X_{j(1)}^{(q,\pm)}(t) = \pm \frac{1}{\sqrt{2N}} \exp[i(q \cdot r_{j(1)} - \omega^{(q,\pm)} t)] \]. \hspace{1cm} (5-2-14)

For each \(q\), the + mode will be the symmetric mode in the sense that corresponding oscillators (for example, oscillator \((j,0)\) of first plane and \((j,1)\) of second plane) move exactly in phase. On the other hand, the – mode will be the anti-symmetric mode in the sense that corresponding oscillators move exactly 180° out of phase.

We also note that the resulted normal modes vectors \(X^{(q,\pm)}\) are Hermitian orthogonal as well as transpose orthogonal. So for 2-layer systems, there will be no interference between the normal modes and therefore no normal mode beats in the system energy and instantaneous decay rates under general initial excitations.

(E). Normal Mode Radiation Fields

For each normal mode of the 2-layer system, we still have Bloch wave form inside each plane. Therefore the normal mode radiation fields are the superpositions of the planar fields given by Eq. (5-1-2).

For \(z<0\), we have,
\[ E^{(q,\pm)}(r,t) = -i \frac{2\pi c}{a^2} X_0 \exp(-i\omega^{(q,\pm)} t) \sum_{\mu} \exp[i k_\mu^{(-)} \cdot r] \frac{k_\mu^{(-)} \times (k_\mu^{(-)} \times \hat{n})}{\varepsilon_\mu^{(-)}} [1 \pm e^{i \delta^{(q,\pm)}}], \] \hspace{1cm} (5-2-15)

where \(k_\mu^{(-)} = -\varepsilon_\mu^{(-)} \hat{n} + i(\tau_\mu + q)\) [note that the superscript \((-)\) on \(k_\mu^{(-)}\) no longer represents the \((-)\) mode here]. For \(z > d\), we have,
\[ E^{(q,\pm)}(r,t) = -i \frac{2\pi e}{a^2} X_0 \exp(-i\omega^{(q,\pm)}t) \sum_{\mu} \frac{k_{\mu}^{(\pm)} \times (k_{\mu}^{(\pm)} \times \hat{r})}{g_{\mu}} [1 \pm e^{-i\phi_{\mu}d}], \]

where \( k_{\mu}^{(\pm)} = g_{\mu} \hat{z} \pm i(\tau_{\mu} \pm q) \).

We see from Eqs. (5-2-15) and (5-2-16) that only the "\( \mu^-\)" channels radiate just as in single layer case. But for each channel that does radiate, there is an extra factor \([1 \pm e^{i\phi_{\mu}d}]\) or \([1 \pm e^{-i\phi_{\mu}d}]\) depending on which side of the crystal we are on. The total averaged radiated power is \( \frac{1}{2} \left( |1 \pm e^{i\phi_{\mu}d}|^2 + |1 \pm e^{-i\phi_{\mu}d}|^2 \right) = 2|1 \pm \cos(g_{\mu}d)| \). On the other hand, the total system energy is twice as much as what it would be for single-layer system. Therefore the radiative decay rates for the two modes will have the factor \([1 \pm \cos(g_{\mu}d)]\) with respect to \( \Gamma_{\mu}^{(q)} \), the radiative decay rate contribution from single layer in this channel [see Eq. (5-2-11)].

(F) Comparison with Two Atom Case

We note that the two-layer case results are directly analogous to the 2-atom case: The normal modes are again the symmetric and anti-symmetric states \( X^{(q,\pm)} \). Also, when the separation \( d \) between the 2 planes is small compared to the wave length [so that \( g_{\mu}d \rightarrow 0 \)] then the radiative decay rate for the \((+)\) mode doubles while the decay rate for the \((-)\) mode drops to zero, just as for the 2-oscillator case. In the \((+)\) mode, the planar radiation fields from the 2-planes interferes constructively for \( d \ll \lambda \), leading to the enhanced decay rates, while in the \((-)\) mode, there is destructive interference and a suppressed decay rate. One obvious difference of the 2-layer case from the 2-oscillator case originates from the fact that now the radiation fields are plane waves instead of spherical waves in the 2-oscillator case. The difference leads to the periodic enhancement and shut-off for the two-layer system when \( g_{\mu}d \) reaches \( \pi, 2\pi, \ldots \), while in the 2-oscillator case, this does not happen.

§ 5-3. M Layers, Determinant Formalism

We can immediately generalize the 2-layer case to an M-layer crystal.

(A). Normal Modes

The normal mode will be of the form
\[ X_{j(m)}^{(q,l)}(t) = X_m^{(q,l)} \exp[i(q \cdot r_{jm} - \omega t)], \quad (m = 0, 1, 2, ..., M-1) \]  

(5-3-1)

which is same as Eq. (5-2-1) except now the layer index \( m \) can be 0 through \( M-1 \), corresponding to \( M \) layers. \( r_{jm} \) is the position of the \( j \)th oscillator in \( m \)th plane. Within each plane, the mode will be a 2-D Bloch wave with Bloch vector \( q \). For each \( q \), there will be \( M \) normal modes \((q,l), l=1, 2, ..., M\). For the \((q,l)\) normal modes, \( X_m^{(q,l)} \) is the planar amplitude for layer \( m \) and \( \omega = \omega^{(q,l)} \) is the normal mode frequency.

(B). Equation of Motion

The normal mode equation of motion is,

\[ Q(\omega) X_m^{(q,l)} = \sum_{\mu} \frac{i}{2} \Gamma_{\mu}^{(q)} \sum_{n=0}^{M-1} X_n^{(q,l)} \exp(ig_{\mu l n m} \omega), \]

(5-3-2)

where \( Q(\omega) \) is given by Eq. (5-1-24). Alternatively, the normal mode equation of motion can be put into the effective "Hamiltonian" form,

\[ \mathcal{H}^{(q)} X^{(q,l)} = \omega X^{(q,l)}, \]

(5-3-3)

where

\[
 X^{(q,l)} = \begin{pmatrix}
 X_0^{(q,l)} \\
 X_1^{(q,l)} \\
 \vdots \\
 X_{M-1}^{(q,l)}
 \end{pmatrix}
\]

(5-3-4)

is the eigenvector under the sub-space of fixed \( q \) where \( X_m^{(q,l)} \) \((m=0,1,2,...,M-1)\) is the amplitude of oscillators of plane \( m \) in mode \((q,l)\), and \( \mathcal{H}^{(q)} \) now is \( M \times M \) matrix, the elements of which are,
\[ h_{mn}^{(q)} = \omega_0 + \delta \omega^{(q)} - \frac{1}{2} i \left( \Gamma_\alpha + \sum_{\mu \leq n} \Gamma_\mu^{(q)} \right), \quad (m=0, 1, 2, \ldots, M-1)\]  
(5-3-5)

\[ h_{mn}^{(q)} = -\frac{i}{2} \sum_{\mu} \Gamma_\mu^{(q)} \exp(ig_{\mu \ln-md}). \quad (m \neq n)\]  
(5-3-6)

(C) Dispersion Relation

The dispersion relation is now,

\[ \text{Determinant } [ \mathcal{H}^{(q)} - \omega I ] = 0, \quad \text{(5-3-7)} \]

which is now an Mth order polynomial equation. Solving this equation will give the normal mode frequencies and damping rates in terms of \( \delta \omega^{(q)} \) and \( \sum_{\mu < n} \Gamma_\mu^{(q)} \), the frequency shift and damping rate for single layer.

(D) Planar Sum Rules

For each sub-space with fixed \( q \), we have a "new" set of sum rules,

\[ \sum_{l=1}^{M} [\omega^{(q,l)} - \omega_0^{(q)}] = 0, \quad \text{(5-3-8)} \]

where \( \omega_0^{(q)} = \omega_0 + \delta \omega^{(q)} \), and

\[ \frac{1}{M} \sum_{l}^{M} \Gamma^{(q,l)} = \Gamma_\alpha + \sum_{\mu < n} \Gamma_\mu^{(q)}, \quad \text{(5-3-9)} \]

where \( \Gamma^{(q,l)} = 2\omega^{(q,l)} \) and \( \Gamma_\alpha + \sum_{\mu < n} \Gamma_\mu^{(q)} \) is in fact the damping rate for the single layer.

Of course, the original "global" sum rules still apply, which can be verified if we take Eqs. (5-1-31) and (5-1-32) into account.
(E). Normal Mode Orthogonality

From Eqs. (5-3-5) and (5-3-6), we see that the effective Hamiltonian $\mathcal{H}^{(q)}$ is symmetric instead of Hermitian. Consequently, the normal modes $\mathbf{X}^{(q,l)}$ ($l=1,2,...,M$) we get from Eq. (5-3-3) are transpose orthogonal,

$$[\mathbf{X}^{(q,l)}]^T \mathbf{X}^{(q,l)} = \delta_{ll}. \quad (5-3-10)$$

One exception is the 2-layer case in which $\mathbf{X}^{(q,l)}$ is real except a global phase factor. In that case, $\mathbf{X}^{(q,l)}$ is both transpose and Hermitian orthogonal.

(F). Synchrotron Pulse Initial Excited State

With the discussion above, we are able to treat quite general initial excitations. But our particular interest is in an initial excitation $|\psi_e^{(k_0)}\rangle$ caused by a synchrotron radiation pulse as discussed in Chapter II. The corresponding classical initial state $\mathbf{X}(k_0)$ has components

$$X_{j(m)}(t=0) = X_{j(m)}(k_0) = X_0 e^{ik_0 \cdot r_j} = X_0 \exp[i\omega_0(m-1)d] \exp[iq_0 \cdot r_j], \quad (5-3-11)$$

where $k_0 = \frac{\omega_0}{c} \mathbf{n}$, $q_0 = k_0 \cdot \hat{\mathbf{z}}$ and $q_0 = (k_0)_{xy} = k_0 - g_0 \hat{\mathbf{z}}$ [See Fig. 17 on the next page].

Because of the special form the initial excitation has, only the modes $(q,l)$ [$l=1,2,...,M$] with $q = q_0$ are excited so that we have only to consider the sub-space with $q$ fixed to $q_0$ (reduced to first Brillouin zone). The initial excitation is then,

$$\mathbf{X}(0) = \mathbf{X}(k_0) = \begin{pmatrix} 1 \\ \exp(i\omega_0 d) \\ . \\ . \\ \exp[i(M-1)g_0 d] \end{pmatrix} \quad (5-3-12)$$
The subsequent motion is,

\[ X(t) = \sum_{l=1}^{M} A^{(q_0,l)}X^{(q_0,l)} \exp[-i\omega^{(q_0,l)}t], \]

(5-3-13)

with

\[ A^{(q_0,l)} = [X^{(q_0,l)}]^T \cdot X(0) \]

(5-3-14)

as the expansion coefficients.

(G) Radiation Fields

The fields from \((q,l)\) normal mode excitation again will be the superposition of the open channels \(k_\mu^{(b)}\) occurring for a single plane layer, with the contributions from various planes adding as \(\sum_{m=0}^{M-1} X^{(q,l)} \exp[\text{img}_\mu d]\) for the \(k_\mu^{(-)}\) channels \((z<0)\), and as \(\sum_{m=0}^{M-1} X^{(q,l)} \exp[-\text{img}_\mu d]\) for the \(k_\mu^{(+)}\) channels \([z>(M-1)d]\).

The \((q,l)\) normal mode radiation field "above" the crystal \((z<0)\) is then,
\[ E^{(q,l)}(r,t) = -i \frac{2\pi e}{a^2} \exp(-i\omega^{(q,l)}t) \sum_{\mu} \exp[ik^{(-)}_\mu \cdot r] \frac{k^{(-)}_\mu \times (k^{(-)}_\mu \times \hat{x})}{g_\mu} \sum_{m=0}^{M-1} X^{(q,l)}_m \exp[\text{imag} \mu d], \]

(5-3-15)

and the field below \((z>M-1)d\)

\[ E^{(q,l)}(r,t) = -i \frac{2\pi e}{a^2} \exp(-i\omega^{(q,l)}t) \sum_{\mu} \exp[ik^{(+)}_\mu \cdot r] \frac{k^{(+)}_\mu \times (k^{(+)}_\mu \times \hat{x})}{g_\mu} \sum_{m=0}^{M-1} X^{(q,l)}_m \exp[-\text{imag} \mu d]. \]

(5-3-16)

Following a synchrotron pulse, the initial excited state is given by Eq. (5-3-12) which can be expressed in terms of the linear superposition of different normal modes as shown in Eq. (5-3-13) and Eq. (5-3-14). The subsequent radiation field is then the superposition of all the normal mode radiation fields,

\[ E(r,t) = \sum_{i=1}^{M} A^{(q_0,l)} E^{(q_0,l)}(r,t), \]

(5-3-17)

where \(A^{(q_0,l)}\) is given by Eq. (5-3-14) and \(E^{(q_0,l)}(r,t)\) is gen by Eqs. (5-3-16) and (5-3-15) with \(q = q_0\).

(H) A Useful Relation for \(\hat{x} \cdot E^{(q,l)}\).

Finally, for future reference, we note a useful general relation for the \(\hat{x}\)-component of the total field in the mode \((q,l)\) at the point \(r(m)=(0,0,md)\) [of layer \(m\): From Eqs. (5-3-15, 16) and Eq. (5-1-27), we have

\[ \frac{e}{2m\omega_0} \hat{x} \cdot E^{(q,l)}[r(m),t] = \sum_{\mu} \frac{i}{2} \Gamma^{(q)}_\mu \sum_{n=0}^{M-1} X^{(q,l)}_n \exp[\text{imag} \mu d|m-nl]. \]

(5-3-18)

But the RHS of Eq. (5-3-18) is precisely the RHS of the normal mode equation of motion of Eq. (5-3-2), giving,
$$\frac{e}{2m\omega_0} \hat{\mathbf{k}} \cdot \mathbf{E}^{(q,1)}[\mathbf{r}(m),t] = Q(\omega^{(q,1)}) \chi_m^{(q,1)}.$$  (5-3-19)

Here $Q(\omega) = \omega_0 - S_0(r) - \omega - i \frac{1}{2} \Gamma_\alpha$ as given by Eq. (5-1-24). Eq. (5-3-19) is of course simply the force equation for an oscillator in the $m$th layer.

$E^{(q,1)}[\mathbf{r}(m),t]$ includes the self-field of layer $m$, and the contribution from all other layers (both open and closed channels). Because the closed channels from layers $n \neq m$ exponentiate off as $\exp(-i g_{\mu \lambda} m_{\mu \lambda})$, the radiation field at the $m$th layer satisfies, to good approximation,

$$\frac{e}{2m\omega_0} \hat{\mathbf{k}} \cdot \mathbf{E}^{(q,1)}_{RAD}[\mathbf{r}(m),t] = Q(\omega^{(q,1)}) \chi_m^{(q,1)},$$  (5-3-20)

where $\hat{\mathbf{k}} \cdot \mathbf{E}^{(q,1)}_{RAD}$ is given by Eq. (5-3-18) with the sum restricted to open channels ($\mu <$) and now,

$$Q(\omega) = \omega_0 + \delta \omega^{(q)} - \omega - i \frac{1}{2} \Gamma_\alpha.$$  (5-3-21)

Here we've used the fact that the closed channel's contributions $S^{(q,1)}$ of layer $m$ combine with $S_0$ in $Q(\omega)$ to give $\delta \omega^{(q)}$.

§5-4 Three-Layer Crystal.

As a specific example of the $M$-layer formalism, we discuss the 3-layer crystalline array.

(A) Eigenmodes

For the 3-layer system, the elements of the effective "Hamiltonian" become,

$$h^{(q)}_{mm} = \omega_0 + \delta \omega^{(q)} - \frac{1}{2} i \left( \Gamma_\alpha + \sum_{\mu <} \Gamma^{(q)}_\mu \right), \quad (m=0, 1, 2)$$  (5-4-1)

$$h^{(q)}_{12} = h^{(q)}_{21} = h^{(q)}_{23} = h^{(q)}_{32} = -\frac{1}{2} \sum_{\mu} \Gamma^{(q)}_\mu \exp(i g_{\mu \lambda} d),$$  (5-4-2)

$$h^{(q)}_{13} = h^{(q)}_{31} = -\frac{1}{2} \sum_{\mu} \Gamma^{(q)}_\mu \exp(2i g_{\mu \lambda} d).$$  (5-4-3)
To find the normal mode frequency, we use Eq. (5-3-7). The result is,

\[ \omega^{(q,1=1)} = \omega_0 + \delta \omega^{(q)} - \frac{1}{2} i \left( \Gamma_\alpha + \sum_{\mu<} \Gamma^{(q)}_\mu - \sum_{\mu} \Gamma^{(q)}_\mu \exp(2i \mu d) \right) \]  \hspace{1cm} (5-4-4)

\[ \omega^{(q,1=2,3)} = \omega_0 + \delta \omega^{(q)} - \frac{1}{2} i \left[ \Gamma_\alpha + \sum_{\mu<} \Gamma^{(q)}_\mu \right] + \frac{i}{4} \left[ \sum_{\mu} \Gamma^{(q)}_\mu \exp(2i \mu d) \right] \sqrt{\left[ \sum_{\mu} \Gamma^{(q)}_\mu \exp(2i \mu d) \right]^2 + 8 \left[ \sum_{\mu} \Gamma^{(q)}_\mu \exp(i \mu d) \right]^2} \]  \hspace{1cm} (5-4-5)

Comparing the normal mode frequency \( \omega^{(q,1)} \) of mode 1 with \( \omega^{(q,-)} \) given in Eq. (5-2-9) for the anti-symmetric mode of the 2-layer system, we find that they are almost the same except d is replaced by 2d. We can immediately conclude that the first mode in three-layer case is the anti-symmetric mode in which the top and the bottom layers move exactly 180° out of phase while the center layer remain motionless. (The \( \hat{\mathbf{k}} \) components of the radiation fields of he top and the bottom layers will cancel at the center layer.)

(B) Symmetric Two Wave Approximation

In order to see the physical trend in the second and the third modes, we first make a simplifying 2-wave approximation: We truncate the results of Eq. (5-4-5) by dropping all the \( \tau_\mu \) terms except one which we will take as \( \tau_0 = 0 \) corresponding to two open channels \( k^{(2)}_0 \) [If \( \tau_0 \neq 0 \), we can always redefine \( q \) as \( q' + \tau_0 \), so that with the new Bloch vector \( q' \), the two-open channels are \( k^{(2)}_0 \)]. The two open channels \( k^{(2)}_0 \) couple the three different plane layers. We will further more assume maximum coupling of the radiation fields to the oscillators (which are oscillating along \( \hat{\mathbf{k}} \) direction). This means that the two open channels \( k^{(2)}_0 \) are perpendicular to \( \hat{\mathbf{k}} \) as shown schematically in Fig. 18.
Fig. 18 Symmetric two wave approximation. Note here we have shown the system of M layers. For the three-layer case in this section, M = 3.

For the symmetric two wave approximation, we then have,

\[ q = q \hat{y} = k_0 \cos \phi \hat{y}, \]  \hspace{1cm} (5-4-6)

\[ g_0 = \sqrt{k_0^2 - q^2} = k_0 \sin \phi_0, \]  \hspace{1cm} (5-4-7)

\[ k_0^{(\pm)} = \pm g_0 \hat{y} + q, \]  \hspace{1cm} (5-4-8)

\[ \Gamma_0^{(q)} = \left[ \frac{3\pi n_0^{(2)} \Delta^2}{\sin \phi_0} \right] \Gamma_\gamma \]  \hspace{1cm} (5-4-9)

The normal mode complex eigenfrequencies are then,

\[ \omega_q^{(1,1)} = \left( \omega_0 + \delta \omega^{(q)} \right) - \frac{1}{2} i \Gamma_\alpha - \frac{1}{2} i \Gamma_0^{(q)} \left[ 1 - \exp(2ig_0d) \right], \]  \hspace{1cm} (5-4-10)

\[ \omega_q^{(1,2,3)} = \left( \omega_0 + \delta \omega^{(q)} \right) - \frac{1}{2} i \Gamma_\alpha - \frac{1}{2} i \Gamma_0^{(q)} \left( 1 - \frac{1}{2} \exp(2ig_0d) \exp(-2ig_0d) \right). \]  \hspace{1cm} (5-4-11)

The complex eigenfrequencies \( \omega_q^{(1,1)} \) are expressed as functions of \( g_0d \), or equivalently by Eq. (5-4-7), they can be expressed as functions of \( q = |q| \) or \( \phi_0 \).
In Fig. 19, we plot the frequency shift \( \Delta \omega^{(q,l)} = [\omega^{(q,l)} - (\omega_0 + \delta \omega^{(q)})] \) as functions of \( g_0d \) for all three modes, for \( g_0d \) from 0 to \( \pi \). In Fig 20, we give the corresponding plots for the decay rates, \( \Gamma^{(q,l)} \).

We note in particular that for those \( q \) that satisfy a "Bragg condition" \( g_0d = m\pi \), the frequency shifts \( \Delta \omega^{(q,l)} \) all go to zero, and only one mode has non-zero decay rate, which reaches \( 3\Gamma_0^{(q)} \), three times as big as the \( \tau_\mu = 0 \) channels contribution to the decay rate of a single layer. What has happened is that for one mode, the "Bragg mode", the fields in the \( k_0^{(l)} \) (or \( \tau_\mu = 0 \)) channels from all three layers interfere constructively. Relative to a single layer, the energy flux is increased 9-fold, while the total mechanical energy is increased 3-fold, giving an enhanced decay rate \( 3\Gamma_0^{(q)} \). For the remaining two modes, there is complete destructive interference of the fields external to the crystal.

Fig. 19. Plot of \( \omega^{(q,l)} - (\omega_0 + \delta \omega^{(q)}) \) as function of \( g_0d \). The horizontal axis goes from 0 to \( \pi \). The vertical unit is \( \frac{1}{2} \Gamma_0^{(q)} \).
Fig. 20 $\Gamma^{(q,l)} - \Gamma_\alpha$ as the function of $g_0d$

The vertical unit is $\Gamma_0^{(q)}$. $g_0d$ goes from 0 to $\pi$.

For example, if $g_0d = \pi$, one of the non-radiative modes is the l=1 "antisymmetric" mode (1, 0, -1) discussed earlier, and the other is the symmetric mode (1, 2, 1). In the (1, 0, -1) mode, the field outside the crystal will be proportional to $[1-\exp[i g_0d(2d)]] = [1-\exp(2i\pi)] = 0$. In the (1, 2, 1) mode, the field outside the crystal will be proportional to $[1+2 \exp[i g_0d] + \exp[2i g_0d]] = [1-2+1] = 0$. Therefore for those two modes, we get completely destructive interference from the three layers and thus there is no radiative damping for them when $g_0d = \pi$. On the other hand, the other mode configuration is (1, -1, 1). In this mode, the radiation fields outside the crystal is proportional to $\{1-\exp[g_0d] + \exp[2g_0d]\} = 1+1+1 = 3$. We get fully constructive interference from the radiation fields of the three layers. So this mode is the only strongly radiating mode.

Away from the Bragg condition, i.e., for $g_0d \neq m\pi$, the three modes are frequency shifted consistent with the frequency shift sum rule Eq. (5-3-8), as shown in Fig. 19, and all three modes are radiative, consistent with the decay rate sum rule Eq. (5-3-9), or written in another form,

$$\sum_{l=1}^{3} [\Gamma^{(q,l)} - \Gamma_\alpha] = 3 \Gamma_0^{(q)}.$$  \hspace{1cm} (5-4-12)

as shown in Fig. 20.
There are limits to this 2-wave approximation. It breaks down if \( k_0^{(+)} \) should satisfy a multi-beam Bragg condition from more than one set of Bragg planes. In those cases, we have to resort to "3-wave" approximation or more. Also, it relies on the assumption that the number of layers be large. In actual crystals, this condition is well satisfied. Here in the three-layer system, we used this approximation only to roughly describe the behavior of the system.

§5-5 M-layer Crystal in the Symmetric Two-Wave Approximation.

We now treat the M-layer crystal in the symmetric two-wave approximation introduced in the last section. The two-wave approximation assumes that among all the \( r_i \) channels that contribute to the final equation of motion Eq. (5-3-2), only the \( r_0=0 \) channels are significant to the final results, all others will be suppressed during the summation over the layers. Therefore we only have to retain the \( r_0=0 \) terms in Eq. (5-3-2) and subsequently only the \( r_0=0 \) terms in our final results of complex frequency shifts, radiations and so on.

The approximation should give good results for synchrotron pulse excitation if the resulting phasing vector \( k_0 = \frac{\omega_0}{c} n \) is off-Bragg for the crystal film (or if the film is an amorphous non-crystalline film), or \( k_0 \) satisfies a Bragg reflection from the x-y planes (i.e., if \( g_0 d = m \pi \)).

For an amorphous 2-D layer, the scattered radiation will go only into the \( k_0^{(\pm)} \) channels. Similarly, for an off-Bragg excitation of the 3-D crystal, the crystal structure is unimportant, so an "amorphous" 2-wave approximation coupling the \( k_0^{(\pm)} \) channels should lead to good results.

Near a Bragg condition \( g_0 d = m \pi \), the waves \( k_0^{(\pm)} \) will build up constructively within the crystal and dominate all other channels. So again a 2-wave approximation coupling the symmetric channels \( k_0^{(+)} \) and \( k_0^{(-)} \) should give good results.
As before, we will again take the simplest case of maximal coupling of the radiation fields to the oscillators, so that $k_0^{(e)}$ are perpendicular to $\mathbf{k}$ as shown in Fig. 18, and $q$, $g_0$, $k_0^{(e)}$ and $\Gamma_0^{(q)}$ are again given by Eqs. (5-4-6) ~ (5-4-9).

With this simplified 2-wave approximation, we will now develop a more general approach to solve for M-layer system. This is necessary because the determinant approach becomes intractable for $M>3$.

(A) Normal Mode Solution

(1) The normal modes and eigenfrequencies.

The equation of motion for M-layer system is given by Eq. (5-3-2). After making the two-wave approximation, the sum over $\tau_\mu$ is eliminated except only the $\tau_\mu=0$ term:

\[
Q(\omega) X_{m}^{(q,l)} = \frac{i}{2} \Gamma_0^{(q)} \sum_{n=0}^{M-1} X_{n}^{(q,l)} \exp(i g_0 |n-m| a), \quad (m = 0,1,2,\ldots M-1)
\]

(5-5-1)

where $g_0$ is given by Eq. (5-4-7) and $\Gamma_0^{(q)}$ is given by Eq. (5-4-9). And now by $Q(\omega)$ we mean $Q(\omega)$, which is given in Eq. (5-3-21).

Consider the trial solution of the form:

\[
X_m = e^{ikam} \pm e^{ik(a(M-1-m))}, \quad (m=0,1,2,\ldots M-1)
\]

(5-5-2)

where $k$ is a complex parameter yet to be determined. Note here we did not write out the planar part of the solution $\exp[iq \tau_j]$ and for simplicity we omit the superscript "(q,l)" on $X_m$ (=X_m^{(q,l)}).

We choose this form of trial solution based on the physical thinking that there exist two propagating waves inside the crystal. And the eigenmode is the stationary state formed by the superposition of the two waves, similar to the standing waves on a beaded string.
Since we have: $e^{ikma} + e^{ik(M-1-m)a} = 2 \exp\left(\frac{1}{2} i(M-1)ka\right) \cos\left(\frac{1}{2} k(M-1-2m)a\right)$ and $e^{ikma} - e^{ik(M-1-m)a} = -2i \exp\left(\frac{1}{2} i(M-1)ka\right) \sin\left(\frac{1}{2} k(M-1-2m)a\right)$, we call

$$X_m = e^{ikam} \pm e^{ik(M-1-m)a}$$

COS solutions

$$X_m = e^{ikam} - e^{ik(M-1-m)a}$$

SIN solutions

(5-5-3)

Substituting Eq. (5-5-2) back to Eq. (5-5-1), the left hand side yields,

$$\text{LHS} \: Q(\omega)X_m = Q(\omega)(e^{ikma} \pm e^{ik(M-1-m)a})$$

and the right hand side,

$$\text{RHS}$$

$$= \frac{i}{2} \Gamma_0^{(q)} \left\{ \left( \exp(i\omega a) \pm e^{ik(M-1-m)a} \right) \right.$$ 

$$+ \frac{\exp(ig_0a) - \exp(ikam)}{1 - \exp[i(k-g_0)a]} \pm \frac{\exp[i(M-1)ka] - \exp[i(M-m-1)ka]}{1 - \exp[-i(k+g_0)a]}$$

$$+ \frac{\exp[i(M-1)ka + i(M-1)g_0a] - \exp(ikam)}{1 - \exp[-i(k+g_0)a]}$$

$$\pm \frac{\exp[i(M-m)g_0a] - \exp[i(M-m-1)ka]}{1 - \exp[i(k-g_0)a]} \right\}$$

$$= \frac{i}{2} \Gamma_0^{(q)} \left( e^{ikam} \pm e^{ik(M-1-m)a} \right) \left\{ 1 - \frac{1}{1 - \exp[i(k-g_0)a]} \right.$$ 

$$+ \frac{1}{1 - \exp[-i(k+g_0)a]} + \frac{\exp[i(M-1)ka]}{1 - \exp[i(k-g_0)a]} + \frac{\exp[-i(M-1)ka]}{1 - \exp[-i(k+g_0)a]} \right\}$$

$$= \frac{i}{2} \Gamma_0^{(q)} X_m \left\{ 1 - \frac{1}{1 - \exp[i(k-g_0)a]} \right.$$ 

$$+ \frac{1}{1 - \exp[-i(k+g_0)a]} \right\}$$

$$+ \frac{i}{2} \Gamma_0^{(q)} \left( \exp(ig_0am) \pm \exp[i_0a(M-1-m)] \right) \left\{ 1 - \frac{1}{1 - \exp[i(k-g_0)a]} \right.$$ 

$$+ \frac{1}{1 - \exp[-i(k+g_0)a]} \pm \frac{\exp[i(M-1)ka]}{1 - \exp[i(k-g_0)a]} \right\} \right. \}.$$ 

(5-5-4)
In order for $X_m$ be the eigenvector, the second term in Eq. (5-5-4) has to be zero. That gives us the "boundary condition" to determine the $M$ allowed values of $k$,

$$\frac{1}{1-\exp[i(M-1)ka]} \pm \frac{\exp[i(M-1)ka]}{1-\exp[-i(k+g_0)a]} = 0. \tag{5-5-5}$$

Substituting Eq. (5-5-5) into Eq. (5-5-4), we get the dispersion relation to determine the eigenmode frequencies $\omega = \omega(k)$,

$$Q(\omega) = \frac{i}{2} \Gamma_0^{(q)} \left( \frac{1}{1-\exp[i(k+g_0)a]} - \frac{1}{1-\exp[i(k-g_0)a]} \right), \tag{5-5-6}$$

where $Q(\omega) = \bar{\omega}_0 - \omega - i \frac{1}{2} \Gamma_0 \omega$ for definition of $\bar{\omega}_0$ refer to Eq. (5-1-24).

The procedures is to first solve Eq. (5-5-5) for the various $k^{(l)}(g_0) \ [l=1,2,\ldots,M]$. Each $k^{(l)}(g_0)$ is then substituted into Eq. (5-5-6), giving $Q(\omega^{(l)})$ and consequently $\omega^{(l)}$. The parameter $g_0 = \sqrt{k_0^2 - q^2}$ where $q$ is the planar vector indicating the planar Bloch subspace. Thus solving the equations Eqs. (5-5-5) and (5-5-6) for different values of $g_0$, we get the eigenfrequencies for all the normal modes $(q,l)$.

We can rewrite the basic equation Eqs. (5-5-5) and (5-5-6) in a more convenient form. Letting $Z=\exp(ika)$, the boundary condition Eq. (5-5-5) becomes,

$$Z^{M+1} - \exp[i \omega a] \left( Z^M + \left(-Z \exp[i \omega a] + 1 \right) \right) = 0, \tag{5-5-7}$$

and the dispersion relation Eq. (5-5-6) becomes,

$$Q(\omega) = \frac{i}{2} \Gamma_0^{(q)} \left( \frac{1}{1-Z \exp[i \omega a]} - \frac{1}{1-Z \exp[-i \omega a]} \right). \tag{5-5-8}$$

The eigenmode configuration [Eq.(5-5-2)] becomes:

$$X_m^{(Z)} = Z^m \pm Z^{M-1-m}, \tag{5-5-9}$$
where \( m = 0, 1, 2, \ldots, M-1 \). The \( M \) components constitute an eigenvector:

\[
X^{(Z)} = \begin{pmatrix}
X_0^{(Z)} \\
X_1^{(Z)} \\
\vdots \\
\vdots \\
X_{M-1}^{(Z)}
\end{pmatrix}
\]  

(5-5-10)

We use the software package Mathematica\textsuperscript{TM} to solve Eq.(5-5-7) and get the results for \( \omega \) and \( X^{(Z)} \). The boundary condition Eq. (5-5-7) constitute two \( M+1 \) order equations, with (+) equations giving the COS solutions and the (−) equations give the SIN solutions. There are \( 2M+2 \) solutions to the boundary condition Eq. (5-5-7). Two of those solutions are "null" solutions giving "mode" \( X^{(Z)} \equiv 0 \). In particular, the two null solutions are: \( Z = 1 \) which is always a solution to the (−) equation, and \( Z = -1 \) which is a solution to the (−/+)-equation if \( M \) is (odd/even). The remaining \( 2M \) solutions are paired, with each pair being two identical solutions. Therefore if \( M \) is even, there are \( M/2 \) distinct COS and SIN solutions, while if \( M \) is odd, there are \( (M+1)/2 \) COS solutions and \( (M-1)/2 \) SIN solutions. In either cases, there are \( M \) distinct eigenmodes for an \( M \)-layer system in the \( q \)-sub-space, which will now be distinguished by the integer index \( l = 1, 2, \ldots M \).

(2) Normal Mode Radiation Fields

For each normal mode, the radiation fields emerging from the crystal are given by Eqs. (5-3-15) and (5-3-16). For our symmetric 2-wave approximation, only the \( k_0^{(\pm)} \) channels contribute. Further more, since we've assumed the simplest case of maximal coupling of the radiation to the oscillation, the \( k_0^{(\pm)} \) are perpendicular to \( \hat{x} \), and hence the normal mode radiation fields are polarized in the \( \hat{x} \)-direction. As a consequence, in the mode (l), the amplitudes of the fields emerging in the \( k_0^{(-)} \) and \( k_0^{(+) \text{ }} \) channels "above" the crystal (\( z < 0 \)) and below the crystal (\( z > L = (M-1) d \)) are,
\( E^{(l)}(z<0,t) = C \frac{1}{2} i \Gamma_0 \exp(i k_0^{(-)} \cdot r - i o^{(l)}(t)) \sum_{m=0}^{M-1} X_m^{(l)} \exp(i m_0 a), \)  \hspace{1cm} (5-5-11)

\( E^{(l)}(z>L,t) = C \frac{1}{2} i \Gamma_0 \exp(i k_0^{(+)\cdot r - i o^{(l)}(t)}) \sum_{m=0}^{M-1} X_m^{(l)} \exp(-i m_0 a), \)  

where \( C=(2m_0 c_0/e) \), and we've also made use of Eq. (5-3-18).

Using Eq. (5-5-1), the planar summation can be evaluated explicitly [compare Eqs. (5-3-19, 20)], giving,

\( E^{(l)}(z<0,t) = C \exp[-i o^{(l)} t] \exp[i k_0^{(-)\cdot r}] Q(o^{(l)}) X_0^{(l)} \)  \hspace{1cm} (5-5-12)

\( E^{(l)}(z>L,t) = C \exp[-i o^{(l)} t] \exp[i k_0^{(+)\cdot r - (M-1) g_0 a}] Q(o^{(l)}) X_{M-1}^{(l)} \)

\( X_0^{(l)} \) is the planar excitation amplitude in mode \((l)\) of the "top" layer, and \( X_{M-1}^{(l)} \) is the excitation amplitude of the "bottom" layer.

(B) General Initial Excitation.

Suppose there is an initial excitation within the \( q \) sub-space,

\[
X(0) = \begin{pmatrix}
X_0 \\
X_1 \\
\vdots \\
\vdots \\
X_{M-1}
\end{pmatrix}
\]  \hspace{1cm} (5-5-13)

It can be expanded in terms of all the linearly independent eigenvectors within the \( q \) sub-space.

\( X(0) = \sum_l A^{(l)} X^{(l)} \),  \hspace{1cm} (5-5-14)

where \( A^{(l)} \) are the expansion coefficients which can be determined by,
\[ A^{(0)} = (X^{(0)})^T X(0). \] (5-5-15)

The subsequent radiation field of such a system, according to Eq. (5-3-17), would then be, for \( z > (M-1)a \) and for \( z < 0 \),

\[
E[z > (M-1)a, t] = C \exp\{i[k_0^{(+)} \cdot r - (M-1)g_0 a]\} \sum_l A^{(l)Q(\omega^{(l)})} X^{(l)}_{M-1} \exp[-i\omega^{(l)} t] \quad (5-5-16)
\]

\[
E(z < 0, t) = E_0 \exp\{i[k_0^{(-)} \cdot r]\} \sum_l A^{(l)Q(\omega^{(l)})} X^{(l)}_0 \exp[+i\omega^{(l)} t]. \quad (5-5-17)
\]

1. Synchrotron radiation pulse induced phased state \( X(k_0) \).

We are particular interested in the nuclear excited states \( |\psi_e(k_0)\rangle \) produced by synchrotron pulses, as discussed in Chapter II. The corresponding classical initial state is,

\[
X(0) = X(k_0) = \begin{pmatrix}
1 \\
\exp(iga) \\
\cdot \\
\cdot \\
\exp[iq_0 \cdot r_j]
\end{pmatrix}, \quad (5-5-18)
\]

where \( k_0 = (\omega_0/c)n_0 = q_0 + \frac{g_0}{c} \) and \( n_0 \) is the direction of the incident synchrotron pulse.

We give two examples below on with a 3-layer system and a 10-layer system. In those examples, the radiated intensities in both of the above directions are plotted as functions of time for several typical \( g \) values. Note again that we only have to consider the \( \mathbf{q} = q_0 \) sub-space because only the modes in this sub-space are excited.

As just discussed, the general procedure is first to expand \( X(0) \) with the normal modes using Eq. (5-5-14). And then the total radiation fields are the superposition of the normal mode radiations as we did for the general case in Eq. (5-5-16) and Eq. (5-5-17).
2. Born approximation results for initial decay rates of the phased state $X(k_0)$.

Before proceeding, we first calculate the radiative decay rates for the phased state $X(k_0)$ in the Born approximation.

Eq. (5-1-2) gives the radiation fields for a single plane of phased oscillators. For the system of $M$-layer crystals, under the two wave approximation, only the two $\tau_{\mu}=0$ channels are kept. In the Born approximation, the total field for an $M$-layer crystal is simply the superposition of the fields from $M$ independent planes. Of course, there is a different phase factor associated with each different plane. For the $k_0^{(+)}$-direction [for $z>(M-1)a$], the additional phase factor for each plane, $\exp[ig_0(Z-md)]$, is just the inverse of the phasing factor $\exp(i g_0 md)$ in the initial excitation. Therefore the fields from all $M$ planes add up constructively in the $k_0^{(+)}$-direction. If we denote the field radiated from a single plane in one direction as 1, then the total field from $M$ planes in the $k_0^{(+)}$-direction is simply $1+1+\ldots+1 = M$. In the $k_0^{(-)}$-direction (for $z<0$) however, the phase factors are $\exp[ig_0(md-Z)]\exp(i g_0 md)$. So the field in the $k_0^{(-)}$-direction is

$$|1+e^{i2ga}+\ldots+e^{i(M-1)ga}| = |\frac{1-e^{2iMga}}{1-e^{2iga}}| = \frac{\sin(Mga)}{\sin(ga)}.$$

If we denote the mechanical energy of one plane as 1, then the energy of the system of $M$ layers is $M$. The radiative decay rate is then proportional to $\Gamma_c(M) = \frac{1}{M} \left[ M^2 + \frac{\sin^2(Mga)}{\sin^2(ga)} \right]$.

Thus, in the Born approximation, an $M$-layer system in the phased state $X(k_0)$ will have an initial radiative decay rate,

$$\Gamma_c(k_0) = \frac{1}{2M} \left[ M^2 + \frac{\sin^2(Mga)}{\sin^2(ga)} \right] \Gamma_0^{(q)},$$

(5-5-19)

From Eq. (5-5-19), we see that when $ga=0, \pi, \ldots, \Gamma(k_0) = M \Gamma_0^{(q)}$. In other cases, $\Gamma_c(k_0)$ is still greater or equal to $\frac{M}{2} \Gamma_0^{(q)}$. In all cases, $\Gamma_c(k_0)$ is roughly proportional to the number of layers.

Combining Eq. (5-5-19) and $\Gamma_0^{(q)}$ in Eq. (5-4-9), we have
\[
\Gamma_c(k_0) = \frac{3\pi\hbar^2}{2\sin\phi_0} \left[1 + \frac{\sin^2(Mga)}{M^2\sin^2(ga)}\right] M_0 \Gamma_r
\]  
(5-5-20)

where \(\phi_0\) is the angle between the direction of \(k_0\) and the plane. Note that \(\Gamma_r\), the thickness in the direction of \(k_0\), is

\[
L_\parallel = \frac{M_0}{\sin\phi_0}.
\]  
(5-5-21)

We may write Eq. (5-5-19) in the form of,

\[
\Gamma_c(k_0) = \pi\hbar^2 \left[1 + \frac{\sin^2(Mga)}{M^2\sin^2(ga)}\right] L_\parallel \Gamma_{coh},
\]  
(5-5-22)

where \(\Gamma_{coh} = \frac{3}{2} \Gamma_r\) [which corresponds to \(\frac{2J_1+1}{4J_0+2} \Gamma_r\) as given in Eq. (2-5) when \(J_1=1\) for the excited state and \(J_0=0\) for the ground state]. We note that when \(ga=0,\pi,2\pi,\ldots\), Eq. (5-5-22) gives us \(\Gamma_c(k_0) = 2\pi\hbar^2 L_\parallel \Gamma_{coh}\) which agrees with Eq. (2-8). When \(ga\) is far from those values, \(\left[1 + \frac{\sin^2(Mga)}{M^2\sin^2(ga)}\right] \approx 1\), therefore we have \(\Gamma_c(k_0) = \pi\hbar^2 L_\parallel \Gamma_{coh}\) which agrees with Eq. (2-6). In both of those situations, the decay rate of an \(M\)-layer system is proportional to the thickness of the crystal in the direction of the synchrotron pulse. So as the thickness of the crystal gets large, we will see appreciable enhancement in the radiative decay.

We have to note though the above analysis is obtained after ignoring the interaction between the layers. It is equivalent to the Born approximation result. As time passes by, the initial phasing between the different layers is destroyed because of the mixture of eigenmodes with different eigenfrequencies (see the discussion in section 4-4-D). However, this will not happen if the initial state satisfies the condition \(ga=n\pi\), which guarantees us a pure normal mode.

(C) Results for a three-layer crystal.

(1) Eigenfrequencies.

We'll first do the three layer problem again using the new method and compare with our previous results. What we are interested in are (1) the complex eigenfrequencies for
the eigenmodes, and the eigenmode configurations, (2) how the system evolves with time if the initial excitation is a "nuclear excitation" state $X(k_0)$.

As we did in the previous section, we plot the frequency shifts $\text{Real}[\omega^{(l)} - (\omega_0 + \delta\omega^{(q)})]$ and the decay rates $\Gamma^{(l)} - \Gamma_\alpha$ as a function of $g_0 a$, for the three eigenmodes, as $g_0 a$ goes from 0 to $\pi$. Since $g_0 = \sqrt{k_0^2 - q^2} = k_0 \sin \phi_0$, we can also view those plots as being plotted against $q = |q|$ or $\phi_0$.

The plots given in Fig. 21, Fig. 22 were made using the Mathematica library function "ListPlot". What has been done is that for every sample point of $g_0 a$, we solve Eq. (5-5-7) for all the $Z$ solutions and then substitute the resulted solutions into Eq. (5-5-8) and get the frequency shifts and the decay rates. Before we actually plot the data point, we first check to see if it gives non-zero eigenvectors, that is, substituting the resulting $Z$'s into Eq. (5-5-9) to see whether the resulted vector [Eq. (5-5-9)] has all zero components. If the "eigenvector" is zero, we ignore that solution and put the points onto the horizontal axis.

Ignoring the null solution points on the axis, we see three curves on both plots. It means that although Eq. (5-5-7) gives us $2(M+1) = 2(3+1) = 8$ solutions, it only gives three non-trivial sets of eigenfrequency solutions and therefore three distinguishable sets of normal modes. Two of those modes are "COS" solutions and one mode is "SIN" solution, consistent with our discussions in part (A).

Comparing Figs. 21, 22 with Figs. 18, 19, we find that they are identical. Therefore all the discussion made there apply here.
Fig. 21. Plot of Real[ω(l) - (ω₀ + δω(q)))] as function of g₀a. (l=1,2,3). The horizontal axis goes from 0 to π. The vertical unit is \( \frac{1}{2} \Gamma₀^{(q)} \).

Fig. 22. Plot of decay rates \( Γ^{(l)} - Γ_α \) as the function of g₀a for eigenmodes (l=1,2,3). The vertical unit is \( Γ₀^{(q)} \) and g₀a goes from 0 to π.

(2) Radiative decay from a "nuclear exciton" initial state \( X(k₀) \).

For the initial phased state \( X(k₀) \) expressed in Eq. (5-5-18), the initial phasing between three layers is,
\[
\begin{pmatrix}
X_{(0)} \\
X_{(1)} \\
X_{(2)}
\end{pmatrix} = \begin{pmatrix}
1 \\
\exp(iga) \\
\exp(2iga)
\end{pmatrix}
\]

(5-5-23)

Using Eqs. (5-5-13) through (5-5-17), we can calculate the radiation power in both the \(k_0^{(+)}\) and \(k_0^{(-)}\) directions for specific \(ga\) values.

In Fig. 23, we plot the radiated intensities as the function of time for \(ga=0.1\pi\). The dashed curve is the reference line decaying at the rate \(\frac{1}{6} \left[9 + \frac{\sin^2(0.3\pi)}{\sin^2(0.1\pi)}\right] \Gamma_0^{(q)}\), which, as given by Eq. (5-5-19), is the Born approximation result of the total radiative decay rate. The top solid curve is the radiated intensity in the \(k_0^{(+)}\) direction. The lower solid curve is the radiation in the \(k_0^{(-)}\) direction. From the Born approximation, the probabilities of decaying into the \(k_0^{(+)}\) and \(k_0^{(-)}\) directions are in the ratio of \(9 \frac{\sin^2(0.3\pi)}{\sin^2(0.1\pi)} = 6.86\), so for \(ga=0.1\pi\), we have strong radiation in both directions.

![Graph](image_url)

Fig. 23 Radiated intensity from 3-layer system as the function of time following an initial phased excitation with \(ga=0.1\pi\) as a function of time. The dashed curve (hidden under the top solid curve) is the reference line with a decay rate of \(\frac{1}{6} \left[9 + \frac{\sin^2(0.3\pi)}{\sin^2(0.1\pi)}\right] \Gamma_0^{(q)}\). The top solid curve is the radiation power in \(k_0^{(+)}\) direction. The lower solid curve is the radiated intensity in the \(k_0^{(-)}\) direction. The vertical unit is \(\frac{1}{4} (\Gamma_0^{(q)})^2\), the radiated intensity in the \(k_0^{(2)}\) directions from a single plane. The horizontal time unit is \(\frac{1}{\Gamma_0^{(q)}}\).
Fig. 24 Radiated intensity from 3-layer system as the function of time following an initial phased excitation with \( \gamma = 0.5\pi \) as the function of time. The dashed curve (hidden under the top solid curve) is the reference line with a decay rate of \( \frac{1}{6} [9 + \frac{\sin^2(1.5\pi)}{\sin^2(0.5\pi)}] \Gamma_0(q) \). The top solid curve is the radiation power in \( k_0^{(+)} \) direction. The lower solid curve is the radiation power in the \( k_0^{(-)} \) direction. The vertical unit is \( \frac{1}{4} (C \Gamma_0)^2 \), the radiated intensity in the \( k_0^{(\pm)} \) directions from a single plane. The horizontal time unit is \( \frac{1}{\Gamma_0(q)} \).

Fig. 24 gives the results for \( \gamma = 0.5\pi \). The phasing vector \( k_0 \) for the initial excitation is now "far off-Bragg". The initial radiation power in the \( k_0^{(-)} \) direction drops to one unit, that is to say, the radiation in this direction has only the intensity that a single layer emits. Comparing to the \( k_0^{(+)} \) direction intensity, which is 9 units initially, we can effectively ignore the radiation in \( k_0^{(-)} \) direction (this is more true if the number of layers gets bigger). The decay of the system is close to the dashed line, which represents the Born approximation reference curve that decays at the rate of \( \frac{1}{6} [9 + \frac{\sin^2(1.5\pi)}{\sin^2(0.5\pi)}] \Gamma_0(q) = \frac{1}{6} [9+1] \Gamma_0(q) \).
When \( g_a = 0, \pi, 2\pi, \ldots \), (shown in Fig. 25) \( X(k_0) \) is in fact one of the eigenmodes (this has been proven by substituting \( g_a \) into Eq. (5-5-7) and subsequently solving for the normal modes). As we see from Fig 22, at those \( g_a \) values only one mode is radiating. Therefore the system's decay rate is the same as the rate of the mode. There is no mixture of other modes and other eigenfrequencies or decay rates. Therefore the initial phase relation between the layers is kept all the time, although the overall oscillation amplitude decays as the function of time. Consequently, we see a truly exponentially decaying state, decaying at the enhanced rate \( \frac{1}{6}(9+9) \Gamma_0^{(q)} = 3\Gamma_0^{(q)} \), which is three times the decay rate of a single layer.

When \( g_a \) equals to \( m\pi \), the radiation from all planes add up constructively in both the \( k_0^{(+)} \) and the \( k_0^{(-)} \) directions. Therefore there is strong radiation in both directions. Since the initial phasing between the layers is maintained, the constructiveness in superposition of the planar radiation fields is also kept for both directions. This means that both waves contribute fully to the radiative decay of the system, and the resulting radiative decay rate is approximately twice the initial decay rate for the "off-Bragg" \( g_a = 0.5\pi \) case.

![Radiated Intensity From 3-layer System](image)

**Fig. 25** Radiated intensity from 3-layer system \((g=0,\pi,...)\). The units are the same as in Fig. 23. Here the solid curve represents both the intensities in \( k_0^{(\pm)} \) direction and Born approximation results.

In Fig. 25, the three curves for \( k_0^{(+)} \), \( k_0^{(-)} \) and the Born approximation merge into one, which means that the \( k_0^{(+)} \) and the \( k_0^{(-)} \) directions have exactly the same radiated
intensity, and that the Born approximation correctly predicts the radiative decay rate for the "Bragg mode excitation".

(D). Results for a 10-layer crystal.

(1) Eigenfrequencies.

We can immediately extend the consideration to an arbitrarily thick crystal (But of course for large M, the calculation times involved will become excessive). As a specific example, we take a ten layer crystal, M=10.

Fig. 26 and Fig. 27 give the frequency shifts $\text{Re} \{ \omega^{(l)} - (\omega_0 + \delta \omega^{(q)}) \}$ for the 10 normal modes as function of $g_0 a$. Fig. 26 plots the SIN solution branch and Fig. 27 plots the COS solution branch. There are 5 solutions for each branch.

Fig. 26. $\text{Re} \{ \omega^{(l)} - (\omega_0 + \delta \omega^{(q)}) \}$, for the five SIN solutions for the 10-layer system. The vertical unit is $\Gamma_0^{(q)}/2$ and $g_0 a$ goes from 0 to $\pi$. 
Fig. 27. $\text{Real}[\omega^{(l)} - (\omega_0 + \delta\omega^{(q)})]$, for the five COS solutions for the 10-layer system. The vertical unit is $\Gamma_0^{(q)}/2$, and $g_0a$ goes from 0 to $\pi$.

Fig. 28 and Fig. 29 plot the radiative decay rate $\Gamma^{(l)} - \Gamma_\alpha - \Gamma_\gamma$ for the different modes. Fig. 28 gives the plot for the SIN solutions and Fig. 29 plots the COS solutions.

Fig. 28. $\Gamma^{(l)} - \Gamma_\alpha$ for the five SIN solutions for 10-layer system. The vertical unit is $\Gamma_0^{(q)}$ and $g_0a$ goes from 0 to $\pi$. 
Fig. 29. $\Gamma^{(1)} - \Gamma_\alpha$ for the five COS solutions for the 10-layer system. The vertical unit is $\Gamma_0^{(q)}$ and $g_0 a$ goes from 0 to $\pi$.

From Figs. 26, 27 and Figs. 28, 29, we see again that although there are 2(M+1) roots for Eq. (5-5-6), we only obtain M eigenvalues for each ga value (here M=10, so the number of solutions is 10).

We note in particular that for those $q$ that satisfy a "Bragg condition" $g_0 d = m \pi$, the frequency shifts $\text{Real}[\omega^{(q)} - (\omega_0 + \delta\omega^{(q)})]$ all go to zero, and only one mode has non-zero decay rate, which reaches $10\Gamma_0^{(q)}$, ten times as big as the $\tau_\mu = 0$ channels contribution to the decay rate of a single layer. What has happened is that for the "Bragg mode", the fields in the $k_0^{(2)}$ (or $\tau_\mu = 0$) channels from all ten layers interfere constructively. Relative to a single layer, the energy flux is increased 100-fold, while the total mechanical energy is increased 10-fold, giving an enhanced decay rate $10\Gamma_0^{(q)}$. For the remaining two modes, there is complete destructive interference of the fields external to the crystal.

Away from the Bragg condition, i.e., for $g_0 d \neq m \pi$, all ten modes are frequency shifted consistent with the frequency shift sum rule Eq. (5-3-8), as shown in Figs. 26, 27, and all ten modes are radiative, consistent with the decay rate sum rule Eq. (5-3-9), or written in another form,
\[ \sum_{l=1}^{3} \left[ \Gamma_{(q,l)} - \Gamma_{(q)} \right] = 10 \Gamma_{(q)}^{(q)}, \]  

(5-5-24)

as shown in Figs. 28, 29.

(2) Synchrotron radiation pulse induced phased state \( X(k_0) \).

Just as we did for 3-layer system, we set up the system with a synchrotron pulse excitation initial condition,

\[ \{X_m(t=0)\} = \{\exp(ig_0am)\} \quad (m=0,1,2,...9) \]  

(5-5-25)

The jth oscillator in the nth layer has a complex oscillation amplitude,

\[ X_{(m)}(t=0) = \text{exp}(i q \cdot r_j) \hat{n}. \]

In Figs. 30 and 31, we give the results for the "off-Bragg" excitations, \( ga=0.1\pi \) and \( 0.5\pi \). The solid line in Fig 30 plots the radiated intensity in the \( k_0^{(+)} \) direction. The intensity in the \( k_0^{(+)} \) direction is down by 2 orders of magnitude and has been omitted. The vertical unit is the initial radiation power of a single-layer system. The dashed line is the Born approximation reference curve which has an exponential decay rate of \( \Gamma_{(q)}^{(q)} = \frac{1}{2M} \left[ M^2 + \frac{\sin^2(Mga)}{\sin^2(ga)} \right] \Gamma_0^{(q)} = 5\Gamma_0^{(q)}. \) (M=10, \( ga=0.1\pi \) or \( 0.5\pi \)). We see initially the two curves are very close together but after certain time (\( =2/\Gamma_0 \)), the solid line deviates from the dashed curve giving the Born approximation exponential decay. This is because the off-Bragg initial excitation \( X(k_0) \) is a superposition of all 10 normal modes, and following an initial superradiant decay in the forward direction, which is correctly given by the Born approximation, the system has a slow non-exponential decay.
Fig. 30 Radiated intensity for a 10-layer system in the $k_0^{(+)}$ direction following an "off-Bragg" initial excitation, $g\alpha=0.1\pi$. The units are the same as Fig. 23.

Fig. 31 Radiated intensity for a 10-layer system in the $k_0^{(+)}$ direction following an "off-Bragg" initial excitation, $g\alpha=0.5\pi$. The units are the same as Fig. 23.

To show more clearly the behavior of the decay curve, we put Figs. 30 and 31 in logarithmic scale which gives a more dramatic deviation of the eigenmode solution from the Born approximation at times $t \geq 2/\Gamma_c$; Fig. 32 and Fig. 33 plot the same case as in Fig. 30.
and Fig. 31, the radiated intensity in the $k_0^{(+)}$ direction for 10-layer case when $\alpha_0 = 0.1 \pi$ (Fig. 32) and $\alpha_0 = 0.5 \pi$ (Fig. 33). The difference is that we take the $\log_{10}$ of the intensities. What we see is a much more obvious deviation from the Born approximation (the dashed straight line) at $t \geq 2/T_c$.

![Log plot](image)

Fig. 32 Log plot of the radiated intensity for a 10-layer system in the $k_0^{(+)}$ direction following an "off-Bragg" initial excitation, $\alpha_0 = 0.1 \pi$. 
Fig. 33 Log plot of the radiated intensity for a 10-layer system in the $k_0^{(+)}$ direction following an "off-Bragg" initial excitation, $ga=0.5\pi$.

As shown by Figs. 30, 31, 32 and 33, the initial decay rate is given by the Born approximation (also see discussion in §4-4-D). Further more, when the Bragg condition is satisfied ($ga=m\pi$), a single superradiant "Bragg mode" is excited and the Born approximation gives the correct decay rate for all times $t > 0$.

The initial superradiant decay rate of a crystalline system after a synchrotron pulse excitation $X(k_0)$ is proportional to the thickness of the crystal in the direction of the initial synchrotron pulse, as given by Eq. (5-5-21). Thus for a thick enough crystal, there will be a very strong initial superradiant radiative decay that dominates the incoherent decay $\Gamma_\alpha$.

For delayed time $t \geq 2/\Gamma_\alpha$, if $k_0$ is off-Bragg, then the decay develops into a slow non-exponential decay. To understand this process, one has to take the interaction between oscillators into account and resort to the normal mode method that we discussed. We found that the interaction plays a profound role in the subsequent system development.
VI. Summary and Conclusion

In this thesis, we determined the normal modes and the collective radiative and decay characteristics for a system of nuclei coupled by their radiation fields.

Each nucleus is represented by a one-dimensional classical dipole oscillator which, on the one hand, bears the natural resonance frequency $\omega_0$, on the other hand, decays in mechanical energy through both the radiative ($\Gamma_\gamma$) and non-radiative processes ($\Gamma_\alpha$).

The motion of such a classical oscillator is affected by the radiation from the other oscillators surrounding it. This way, a system of $N$ such oscillators constitutes a coupled system. The coupling may greatly alter the radiative decay rate of the excited system relative to that of an isolated oscillator.

First, we discussed in general the system of $N$ one-dimensional coupled dipole oscillators. We found that it is always possible to establish radiative normal mode motion in the system. The eigenvectors associated with each normal mode are transpose orthogonal rather than Hermitian orthogonal. The eigenfrequencies we find contain a real part which gives the oscillation frequency $\omega^{(k)}$ for the mode, and an imaginary part which gives the decay rate $\Gamma^{(k)}$. The frequency shifts for all the modes satisfy the "sum rules",

$$\sum_k [\omega^{(k)} - \omega_0] = 0 \quad (3-2-5)$$

and

$$\frac{1}{N} \sum_k \Gamma^{(k)} = \Gamma_\alpha + \Gamma_\gamma \quad (3-2-6)$$

We can expand an arbitrary initial excitation of the system with the normal modes. By doing that, we can get the state of the system in any given time $t > 0$,

$$X(t) = \sum_{k=1}^{N} A_k X^{(k)} \exp[-i\omega^{(k)} t] \exp[-\frac{1}{2} \Gamma^{(k)} t]. \quad (3-3-4)$$

We discussed the mechanical energy of the system, the radiation fields and radiated intensity distribution. Unless the system is in a radiative normal mode motion, these results involve more than one frequency component and decay rate and are not simple
exponentially decaying functions. These "interference contributions" arise because the normal mode eigenvectors are not Hermitian orthogonal.

As a first example, we discussed the system of two coupled oscillators, both identical and nonidentical. We devoted a whole chapter to explore in detail the normal modes, eigenfrequencies and decay rates for the system. We can already see from the example the enhanced and suppressed radiative decay for the symmetric and the antisymmetric modes. We also illustrated how we determine the motion of the system at any time $t > 0$ with an arbitrary initial excitation at $t=0$. We obtained the system energy, instantaneous decay rate and radiation. For non-identical oscillators, we showed that the instantaneous decay rate $\Gamma_c(t)$ becomes time dependent and can pulse on and off, from superradiant to subradiant.

For a two dimensional array, the radiative normal modes were found to be two-dimensional Bloch waves. Each different Bloch vector corresponds to a different normal mode. The frequency shift and decay rate of each mode consists of contributions from many "channels". Each open channel corresponds to one plane wave component of the total radiation, and therefore, contributes to the total decay rate. Each closed channel corresponds to a non-propagative spatially decaying field distribution which contributes to the real frequency shift rather than the decay rate. The radiation field from such a system in one of the normal modes will have a single oscillation frequency and decay rate. But it includes a number of plane wave channels symmetrically above the below the plane. The contributions to the final frequency shift and decay rate from those channels are different. Those "just opened" and "just closed" channels will have a more significant contribution.

The M-layer system can be solved based on the results we obtained for the single-layer case. It is shown that the normal modes for the M-layer case are modified 2-D Bloch waves, with Bloch vector $\mathbf{q}$ within each plane, and with modulated planar amplitudes. And then within the sub-space of the Bloch vector $\mathbf{q}$, we are able to establish a "new" linear normal mode system with again a symmetric effective "Hamiltonian" based on the interaction between the different layers. We have a set of sum rules for the mode frequencies and decay rates within each $\mathbf{q}$ sub-space. The normal modes eigenvectors satisfy the transpose orthogonality condition.

We then made the "symmetric two-wave approximation" and truncated the general formalism, keeping only two plane wave channels symmetric with respect to the plane (which we call $\tau_{\mu}=0$ channels), assuming that those two channels are the major parts
contributing to the inter-planar coupling. Using this approximation, we discussed the system of $M$ layers and developed a general procedure to solve the coupled $M$-layer system. The modulation of the planar amplitudes for the planes $m=0,1,...,M-1$ are SIN or COS "standing wave"s with a complex wave vector.

As a specific example, we discussed the three layer system and ten layer system using this method, solving for the eigenfrequencies and decay rates for the various modes. Within each $q$ sub-space, there are $M$ different normal modes corresponding to the $M$ extra degrees of freedom brought in by $M$ layers. We calculated and plotted the normal mode frequency and decay rate results for the case of 3 and 10 layers. Actually, with this method, we can treat systems of any number of layers within the computing ability of the computer.

We then used the normal mode approach to solve for the radiative decay properties of an $M$-layer crystal which has an initial phased excitation $X(k_0)$ corresponding to the nuclear exciton state $\nu_e(k_0) >$ produced by a synchrotron radiation pulse. We show that there is a initial superradiant decay at an enhanced width $\Gamma_e(k_0)$, in agreement with the Born approximation,

$$\Gamma_e(k_0) = \pi n\chi^2 \left[ 1 + \frac{\sin^2(Mga)}{M^2 \sin^2(ga)} \right] L_{\parallel} \Gamma_{coh},$$

(5-5-22)

where $\Gamma_{coh} = \frac{3}{2} \Gamma_\gamma$ [corresponding to $\frac{2J_1+1}{4J_0+2} \Gamma_\gamma$ when $J_1=1$ for the excited state and $J_0=0$ for the ground state]. For a Bragg excitation $(ga=0,\pi,2\pi,...)$, $\Gamma_e(k_0) = 2\pi n\chi^2 L_{\parallel} \Gamma_{coh}$ which agrees with Eq. (2-8). For an off-Bragg excitation, $\left[ 1 + \frac{\sin^2(Mga)}{M^2 \sin^2(ga)} \right] = 1$, giving $\Gamma_e(k_0) = \pi n\chi^2 L_{\parallel} \Gamma_{coh}$ which agrees with Eq. (2-6).

We showed that the difference between the Bragg and off-Bragg situation is not only the value of the decay rate: When the Bragg condition is satisfied, the initial excitation $X(k_0)$ is a radiative normal mode with an unshifted normal mode frequency $\omega_0$ and the decay of the system is truly exponential with radiation emitted in both the $k_0^{(+)}$ and $k_0^{(-)}$ directions. On the other hand, if the Bragg condition is not satisfied, the phased state $X(k_0)$ is a superposition of various normal modes which now have a spread of eigenmode frequencies $\omega^{(0)}$ and width $\Gamma_e^{(0)}$. There is still an enhanced initial decay, but now the
radiation is only emitted into the forward $k_0^{(+)}$ direction, and the superradiant decay rate
$\Gamma_c(k_0) = \pi n^2 L_{\parallel} \gamma_{\text{coh}}$ is one-half the Bragg superradiant rate. For this off-Bragg case, the initial enhanced decay is due in part to the spread of normal mode eigenfrequencies: As we showed, the normal modes are not Hermitian orthogonal, and as a consequence, there are interferences between normal mode contributions to the radiation fields which act to "dephase" the signal, leading to an initially enhanced decay of the signal. Further more, as time develops, because $X(k_0)$ is a superposition of normal modes, the initial superradiant exponential decay changes into a slow non-exponential decay.

The discussion in this thesis is classical. But the same eigenvalue equations are obtained in the quantum mechanical approach. We intend to further develop the parallel quantum mechanical approach in the subsequent work.
Appendix 1. The Radiation from an Oscillating Dipole

In Eq. (3-1-4), we use the expression

\[ E = -k^2 \left[ n_x(n_x d^{(0)}_x) \right] \frac{1}{r} e^{i(kr - i\omega t)} + \left[ 3n(n \cdot d^{(0)}) - d^{(0)} \right] \left( \frac{1}{r^3} - \frac{1}{r^2} \right) e^{i(kr - i\omega t)} \]  \hspace{1cm} (A1-1)

to denote the field radiated from a radiating dipole and we claim this is valid provided the observing point is so far from the source that the size of the source is much smaller than the distance between the observing point and the radiation source. And the approximation is irrelevant with the radiation wave length. In other words, the equation is correct not only in the radiation zone. Also, \( \mathbf{n} \) is the direction pointing from the center of the source to the observing point. \( r \) is the distance from the observing point to the source. \( \omega \) is the frequency of the oscillator. \( k \) is the wave vector value which equals \( \omega / c \).

In the equation, \( d^{(0)} \) is the amplitude of the radiation dipole moment. In order to substantialize the problem, we assume the dipole consists of a harmonically oscillating charged particle. Without sacrificing generality, we let the oscillating direction be along the \( \hat{z} \) direction (like what we show in Fig. 34). And the amplitude of oscillation is \( Z \). Now, \( d^{(0)} = qZ \hat{z} \), where \( q \) is the charge the particle carries.

![Diagram of single oscillator radiation](image-url)

Fig. 34 Single oscillator radiation
The instant position of the particle is,

\[ x = z \hat{\omega} = Z\cos(\omega t) \hat{\omega}. \quad (A1-2) \]

The current density is,

\[ J(x,t) = \rho(x,t) v(x,t) = -qZ\omega \sin(\omega t) \delta(x) \delta(y) \delta(z - Z\cos(\omega t)). \quad (A1-3) \]

In the Lorentz gauge where there are no boundary surfaces present, the vector potential is,

\[ A(x,t) = \frac{1}{c} \int d^3x' \int dt' \frac{J(x',t)}{|x-x'|} \delta(t' + \frac{|x-x'|}{c} - t). \quad (A1-4) \]

Put (A1-3) into (A1-4), we have:

\[ A(x,t) \]

\[ = -\frac{1}{c} \int d^3x' \int dt' \frac{1}{|x-x'|} \delta(t' + \frac{|x-x'|}{c} - t) qZ\omega \sin(\omega t') \delta(x') \delta(y') \delta(z' - Z\cos(\omega t')) \hat{\omega} \]

\[ = -\frac{1}{c} \int d^3x' \frac{qZ\omega \sin[\omega(t - \frac{|x-x'|}{c})]}{|x-x'|} \delta(x') \delta(y') \delta(z' - Z\cos[\omega(t - \frac{|x-x'|}{c})]) \hat{\omega} \]

\[ = -\frac{1}{c} \frac{1}{|x|} qZ\omega \sin[\omega(t - \frac{|x|}{c})] \hat{\omega}. \quad (A1-5) \]

We assume that \(|x| >> |x'|\) and \(\frac{\omega}{c} |x'| << 1\) (small source approximation) so that (A1-5) can be simplified to the form,

\[ A(x,t) = -\frac{1}{c} \frac{1}{|x|} qZ\omega \sin[\omega(t - \frac{|x|}{c})] \hat{\omega} \]

\[ = \text{Real} \left\{ -ik \frac{1}{r} d^{(0)} \exp[i(kr - \omega t)] \right\}, \quad (A1-6) \]

where we have used \(d^{(0)} = qZ \hat{\omega}, k = \omega/c, r = |x|\).

If we decide to switch to the complex expression thereafter, we can drop that "Real" sign. Now we have,

\[ A(x,t) = -ik \frac{1}{r} d^{(0)} \exp[i(kr - \omega t)]. \quad (A1-7) \]
We have,

\[ \mathbf{B}(x,t) = \nabla \times \mathbf{A}(x,t) \]
\[ \mathbf{E}(x,t) = -\frac{i}{k} \nabla \times \mathbf{B}(x,t) = \nabla \times \left[ \nabla \times \frac{1}{r} \mathbf{d}^{(0)} \exp[ikr \omega t] \right]. \]  
(A1-8)

That puts us directly to:

\[ \mathbf{B} = k^2 (\mathbf{n} \times \mathbf{d}^{(0)}) \frac{1}{r} e^{i(kr - i\omega t)}(1 + \frac{1}{ikr}), \]
\[ \mathbf{E} = -k^2 (\mathbf{n} \times (\mathbf{n} \times \mathbf{d}^{(0)})) \frac{1}{r} e^{i(kr - i\omega t)} + 3\mathbf{n} \cdot \mathbf{d}^{(0)} \frac{1}{r^3} - \frac{ik}{r^2} e^{i(kr - i\omega t)}, \]  
(A1-9)

where \( \mathbf{n} = x/|x| \).

There is a static portion of the field that we did not include in (A1-9). That is because we have used the second part of (A1-8) which assumes the field we are dealing with does not have a static part in it. But the whole thing is okay because we are not in any way interested in the static field. We can for example put a negative sign charge of the same amount at the center of the oscillation to cancel the static field.

The time average of the Poynting vector is:

\[ \langle S \rangle = \frac{c}{8\pi} \text{Real} (\mathbf{E} \times \mathbf{B}^*) \]  
(A1-10)

The integral over the \( 4\pi \) solid angle gives us the power output the the radiation source,

\[ P = \int \langle S \rangle \cdot |n| r^2 d\Omega \]  
(A1-11)

The integration has several terms.

\[ P = \int r^2 d\Omega \left\{ k^2 (\mathbf{n} \times \mathbf{d}^{(0)})^2 \frac{1}{r^2} + A \frac{1}{r^3} + B \frac{1}{r^4} + C \frac{1}{r^5} \right\} \]  
(A1-12)

Where \( A, B, C \) are functions independent of \( r \). We see already that these terms will disappear when we take the limit of \( r \rightarrow \infty \). Furthermore by considering the energy conservation principle, we know \( P \) should not depend on where we do the integral as long as the big sphere is outside the source. For those reasons, we drop the 2nd, the 3rd and the 4th term. That leaves us,
\[ P = -\frac{dE}{dt} = \int r^2 d\Omega \frac{k^2}{r^2} (nx) \cdot (n\cdot d)(0)^2 \frac{1}{8\pi} \int \frac{1}{r^2} \sin^2 \theta d\Omega = \frac{C}{3} \frac{k^4}{r^2} (n\cdot d)(0)^2 \]

(A1-13)

In the radiation zone (\(kr >> 1\)), the electric field is:

\[ E = -k^2 \left[ n_x(n\cdot d)(0)^2 \right] \frac{1}{r^3} e^{i(kr-\omega t)} \]

(A1-14)

For an harmonically oscillating particle of mass \(m\) and amplitude \(Z\), the time average energy \(E\) is (include the potential and kinetic energies),

\[ E = \frac{1}{2} m(kc)^2 Z^2 \]

(A1-15)

If we assume that the energy is an exponentially decaying quantity, we have the decay constant \(\Gamma = \frac{-1}{E} \frac{dE}{dt} = \frac{P}{E}\). That gives us,

\[ \Gamma = \frac{2q^2 k^2}{3mc} = \frac{2q^2 \omega^2}{3mc^3} \]

(A1-16)

Here we get the decaying rate for a single oscillating dipole moment. We will be able to get the same result in Appendix 2 using another method.

In the intermediate zone, all the terms of (A1-9) are taken into account when we calculate the electric field.

In the near field zone, (\(ka << kr << 1\), where \(a\) is the size of the source),

\[ E = [3n(n\cdot d)(0) - d(0)] \frac{1}{r^3} e^{i(kr-\omega t)}. \]

(A1-17)

It is inversely proportional to \(r^3\) and blows up rapidly.

Again, we want to emphasize that the results are attained under the assumption of small radiation source. In reality, the separations between nuclei are much greater than the size of the nuclei themselves (the order of magnitude of the difference is more than \(10^3\)) so that the approximation is very well valid.

The above derivation is based on J. D. Jackson's Classical Electrodynamics (Ref. 5).
Appendix 2. The Self-Action and Renormalization Mass (Classical Model)

Newton's equation of motion:

\[
\frac{dp}{dt} = \mathbf{F}_{\text{ext}}, \quad (A2-1)
\]

where \(\frac{dp}{dt}\) can be expressed as,

\[
\frac{dp}{dt} = m_0 \dot{v} - \int (\rho E_s + \frac{1}{c} J x B_s) dx = m_0 \dot{v} + \frac{dp}{dt}, \quad (A2-2)
\]

where \(E_s\) and \(B_s\) are the self fields. Assume that (1) the particle is instantaneously at rest (2) the charge distribution is rigid and spherically symmetric. These two assumptions are approximately satisfied in our case since the motion of the particle is slow compared to that of the speed of light.

Under assumption 1, (A2-2) becomes:

\[
\frac{dp}{dt} = \int \rho(x,t)[\nabla \phi(x,t) + \frac{1}{c} \frac{\partial A(x,t)}{\partial t}] dx^3. \quad (A2-3)
\]

The 4-potential is given by:

\[
A^\alpha(x,t) = \frac{1}{c} \int \frac{[J^\alpha(x',t')]_{\text{rel}}}{R} dx'. \quad (A2-4)
\]

with \(J^\alpha=(\mathbf{c} \rho, \mathbf{J})\) and \(R=x-x'\).

Notice the retarded 4-current can be expanded around \(t'=t\), we have:

\[
\frac{dp}{dt} = \sum_{n=0}^{\infty} \frac{(-1)^n}{n! c^n} \int d^3 x \int d^3 x' \rho(x,t) \frac{\partial^n }{\partial t^n}[\rho(x',t) \nabla R^{n-1} + \frac{R^{n-1}}{c^2} \frac{\partial \mathbf{J}(x',t)}{\partial t}] \quad (A2-5)
\]

Consider the \(n=0\) and \(n=1\) terms in the scalar potential part (the first term in the square bracket) of the right hand side. For \(n=0\) the term is proportional to:
\[ \int d^3x \int d^3x' \rho(x,t) \rho(x',t) \nabla \left( \frac{1}{R} \right). \]  
(A2-6)

This is nothing but the electrostatic self force. For spherically symmetric charge distributions it vanishes. The \( n=1 \) term is also zero since it involves \( \nabla R^{n-1} \). So the summation now reads:

\[
\frac{dp}{dt} = \sum_{n=0}^{\infty} \frac{(-1)^n}{n! c^{n+2}} \int d^3x \int d^3x' \rho(x,t) R^{n-1} \frac{\partial^{n+1}}{\partial t^{n+1}} \nabla R^{n-1} \left\{ \right\} \\
\text{where} \\
\left\{ \right\} = J(x',t) + \frac{\partial \rho}{\partial t} (x',t) \frac{\nabla R^{n+1}}{(n+1)(n+2)R^{n-1}} 
\]  
(A2-7)

Considering the continuity equation for charge and current densities, the curly bracket can be written as,

\[
\left\{ \right\} = J(x',t) - \frac{R}{n+2} \nabla \cdot J(x',t). 
\]  
(A2-8)

In the integral over \( d^3x' \) we can integrate the second term by parts. We then have,

\[
-\int d^3x' \frac{R^{n-1}}{n+2} \nabla \cdot J = -\frac{1}{n+2} \int d^3x' (J \cdot \nabla) R^{n-1} R \\
= \frac{1}{n+2} \int d^3x' R^{n-1} (J + (n-1)J \cdot \frac{R}{R^2} R). 
\]  
(A2-9)

This means that (A2-8) is effectively equal to:

\[
\left\{ \right\} = \frac{n+1}{n+2} J(x',t) - \frac{n-1}{n+2} \frac{(J \cdot R) R}{R^2}. 
\]  
(A2-10)

For a rigid charge distribution the current is:

\[
J(x',t) = \rho (x',t) v(t). 
\]  
(A2-11)

If the charge distribution is spherically symmetric, the only relevant direction in the problem is that of \( v(t) \). Consequently in the integration over \( d^3x \) and \( d^3x' \), only the component of (A2-10) along the direction of \( v(t) \) survives. Hence (A2-10) is equivalent to:
\[ \{ \} = p(x', t)v(t) \left[ \frac{n+1}{n+2} - \frac{n-1}{n+2} \left( \frac{R \cdot v}{Rv} \right)^2 \right] \]  

(A2-12)

Furthermore all directions of \( R \) are equally probable. This means that the above term can be replaced by its average value of 1/3. That gives an even simpler form of (A2-12):

\[ \{ \} = \frac{2}{3} p(x', t)v(t) \]  

(A2-13)

With this, the self force becomes, neglecting non-linear terms in time derivatives of \( v \) which appears for \( n \geq 4 \):

\[ \frac{dp}{dt} = \sum_{n=0}^{\infty} \frac{2(-1)^n}{3n!} \varepsilon_{n+1}^{\alpha+2} \int d^3x \int d^3x' p(x)p(x')R^{n-1}p(x) \]  

(A2-14)

We consider the first few terms in Eq. (A2-14):

\[ \frac{dp}{dt}_0 = \frac{2}{3}v \int d^3x \int d^3x' p(x)p(x') \]  

(A2-15)

\[ \frac{dp}{dt}_1 = -\frac{2}{3}v \int d^3x \int d^3x' p(x)p(x') = -\frac{2e^2}{3c^3}v \]  

(A2-16)

\[ \frac{dp}{dt}_n = \frac{e^2}{n!c^{n+2}} \varepsilon_{n+1}^{\alpha+1} a^{n-1}. \]  

(A2-17)

For particles that we consider here, (A2-17) terms can be neglected due to the size(\( a \rightarrow 0 \)).

Term (A2-16) is the radiative reaction force. It is independent of the structure of the particle.

Term (A2-17) deserves special attention. We know that the self-energy

\[ U = \frac{1}{2} \int d^3x \int d^3x' p(x)p(x') \]  

(A2-18)

If we call the additional mass put in by this portion of self-energy \( m_e \), we can write the equation of motion as:

\[ (m_0 + \frac{4}{3} m_e)\ddot{v} - \frac{2q^2}{3c^3}v = F_{\text{ext}}. \]  

(A2-19)
The self-energy portion of the mass is melted into the total mass $m$. It is not distinguishable. The mass that we see in the experiment is the sum of the two parts of mass. For near simple harmonic oscillators, the first term equals: $m \frac{d^2x}{dt^2}$. The second term equals: $\frac{2e^2\omega^2}{3c^3} \frac{dx}{dt}$. In turn the equation of motion becomes:

$$m \frac{d^2x}{dt^2} + \frac{2e^2\omega^2}{3c^3} \frac{dx}{dt} + m\omega^2x = 0.$$  \hspace{1cm} (A2-20)

We see that self-force term is: $\frac{2e^2\omega^2}{3c^3} \frac{dx}{dt}$. From (A2-20) we easily reach the damping coefficient because of the existence of this term,

$$\Gamma_\gamma = \frac{2e^2\omega^2}{3mc^3}. \hspace{1cm} (A2-21)$$

We got this form in (A1-9) of Appendix 1. That is where the $\Gamma_\gamma$ expression in the thesis comes from. The derivation is based on J.D. Jackson's Classical Electrodynamics. (Ref. 5)
Appendix 3. Radiation From A Plain Layer

For an oscillating source of which the dipole moment can be expressed as,

\[ \mathbf{d} = \mathbf{d}^{(0)} \mathbf{f}(t) = \mathbf{d}^{(0)} \exp[-i\omega t] \]  
(A3-1)

we introduce for convenience the Hertz Vector, or the Hertz Polarization Potential:

\[ \Pi_0 = \frac{1}{r} \mathbf{d}^{(0)} \mathbf{f}(t - \frac{r}{c}) = \frac{1}{r} \mathbf{d}^{(0)} \exp[i(kr - \omega t)]. \]  
(A3-2)

The electric field is in the form of:

\[ \mathbf{E}_0 = \nabla \times \nabla \times \Pi_0 = \nabla \times \nabla \times \frac{1}{r} \mathbf{d}^{(0)} \exp[i(kr - \omega t)]. \]  
(A3-3)

In order to calculate the total electric field from a plane consisting of such oscillating dipoles, we first calculate the sum of all Hertz Vectors.

\[ \Pi = \sum_j \Pi_j = \mathbf{d}^{(0)} e^{-i\omega t} \sum_j e^{ik \cdot r_j} \frac{1}{|r - r_j|} e^{i(k \cdot r - \omega t)}. \]  
(A3-4)

We have relation:

\[ \frac{1}{|r - r_j|} e^{i(k \cdot r - \omega t)} = \frac{1}{2\pi^2} \int \frac{1}{q^2 - k^2 - i\varepsilon} e^{i(q \cdot r - \omega t)} d^3q. \]  
(A3-5)

Substitute Eq. (A3-5) into Eq. (A3-4), we have:

\[ \Pi = \frac{1}{2\pi^2} \frac{p_0 e^{-i\omega t}}{(2\pi)^2} \int d^3q \frac{1}{q^2 - k^2 - i\varepsilon} \sum_j \exp[i(k - q) \cdot r] \exp(iq \cdot r). \]  
(A3-6)

Noting that:

\[ \sum_j e^{iQ \cdot r_j} = \left( \frac{2\pi}{a} \right)^2 \sum_\mu \delta^{(2)}(Q + \tau^\mu_{xy}). \]  
(A3-7)

Substituting (A3-7) into (A3-6), we have:

\[ \Pi = \frac{1}{2\pi^2} \frac{p_0 e^{-i\omega t} (2\pi a)^2}{2} \sum_\mu \int d^3q \frac{\delta^{(2)}(k - q + \tau^\mu_{xy}) e^{i(q \cdot r)}}{q^2 - k^2 - i\varepsilon}. \]
\[
\frac{2}{a^2} p e^{-i\omega t} \sum_{\mu} \exp[i(k_{xy} + \tau_{xy}^\mu) \cdot r] \int dq_x e^{iq_x x} \frac{1}{q_z^2 - (k_{xy} + \tau_{xy}^\mu)^2 - i\epsilon}.
\]  
(A3-8)

The integration part can be evaluated using the contour integration:

\[
I = i\pi \frac{\exp\{i[k^2 - (k_{xy} + \tau_{xy}^\mu)^2]^{1/2} |z|\}}{[k^2 - (k_{xy} + \tau_{xy}^\mu)^2]^{1/2}}
\]  
(A3-9)

Substituting (A3-9) into (A3-8), we get:

\[
\Pi = \sum_j \Pi_j
\]

\[
= \frac{2\pi i}{a^2} p e^{-i\omega t} \sum_{\mu} \exp\{i[k^2 - (k_{xy} + \tau_{xy}^\mu)^2]^{1/2} |z| + i(\tau_{\mu} + k_{xy}) \cdot r\} \frac{1}{[k^2 - (k_{xy} + \tau_{xy}^\mu)^2]^{1/2}}.
\]  
(A3-10)

Therefore

\[
\vec{E} = \nabla \times \nabla \times \Pi =
\]

\[
-i \frac{2\pi e}{a^2} \vec{X}_q \exp(-i\omega t) \sum_{\mu} \exp[i\sqrt{k^2 - (\tau_{\mu} + k_{xy})^2} |z| + i(\tau_{\mu} + k_{xy}) \cdot r] \frac{k_{\mu} \times k_{\mu} \times x_0}{\sqrt{k^2 - (\tau_{\mu} + k_{xy})^2}}
\]  
(A3-11)

This is actually the same as Eq (5-1-2). The derivation is based on the work of Dr. Hannon's in his M.A. thesis. For more detail, refer to Ref. 6.
Appendix 4. Equation of Motion for 2-D Square Arrays

Here we give more details leading to the planar equation of motion $\ddot{X}^{(q)} = \omega X^{(q)}$ given by Eq. (5-1-14) starting with the equation of motion for the jth oscillator, Eq. (5-1-13),

$$m\dddot{X}_j^{(q)} + m\omega^2 X_j^{(q)} + m\gamma \ddot{X}_j^{(q)} = e \hat{\kappa} \lim_{r \to 0} [E_r^{(q)}(r) - E_{rj}^{(q)}(r) \text{ in phase}].$$  \hspace{1cm} (A4-1)

Note that as $r$ goes to zero, $\rho$ in $\sum_{\mu} E_{\mu}^{(q)}$ [given by Eq. (5-1-2)] becomes $R_j$ which satisfies $\gamma R_j = 2n\pi$ and therefore $\gamma$ is eliminated from $\sum_{\mu} E_{\mu}^{(q)}$. Meanwhile, we have $|z|$ going to zero. So we have on the right hand side of Eq. (A4-1),

RHS

$$= X_j^{(q)} \lim_{r \to 0} \left[ -i \frac{2\pi e^2}{a^2} \sum_{\mu} \frac{(\gamma + \alpha)^2 - k_0^2}{\gamma} \exp\{i[g_{\mu} r \hat{\kappa} + (\gamma + \alpha) r^2] + 2m\omega_0 S_0(r)\}, \right]$$  \hspace{1cm} (A4-2)

where

$$S_0(r) = -\frac{3}{4} \Gamma \left\{ \frac{1}{(k_0 r)^3} \left[ 3(n \cdot \hat{\kappa})^2 - 1 \right] + \frac{1}{2k_0^2} \left[ (n \cdot \hat{\kappa})^2 - 1 \right] \right\},$$  \hspace{1cm} (A4-3)

which is a real quantity and diverges as $r$ goes to zero. In the other term which involves the sum over $\mu$, we have used the result $\hat{\kappa} [k_{\mu}^{(+)} x(k_{\mu}^{(+)} \cdot \hat{\kappa})] = (\gamma + \alpha)^2 - k_0^2$. The exponential function that is left in it normally goes unity as $r$ goes to zero. But here, we have to leave it in so that we may take the final limit together with $S_0(r)$.

In Eq. (A4-2), the sum over all $\gamma$ can be broken into two parts. In the first part, we sum over all $\gamma$'s that satisfy $|\gamma + \alpha| < k_0$, which we denote as "$\gamma <". There are only finite number of those terms to be summed. Therefore we may first take the limit of $r$ going to zero and then do the sum.

We let
\[ \Gamma_\mu^{(q)} = \frac{2\pi e^2}{m\omega_0 a^2} \left[ \frac{k_0^2 - (\tau_{\mu x} + q_x)^2}{k_0 n(2) \chi^2} \right] \Gamma_\nu \quad (A4-4) \]

where \( n(2) = 1/a^2 \) is the planar number density of the array. Now the sum over all "\( \mu < \)" can be written as,

\[ -i \frac{2\pi e^2}{a^2} \sum_{\mu <} \frac{(\tau_{\mu x} + q_x)^2 - k_0^2}{g_\mu} \Gamma_\mu^{(q)} = -i \frac{1}{m\omega_0} \sum_{\mu <} \Gamma_\mu^{(q)}. \]

In the second part, we sum over all \( \tau_\mu \)'s that satisfy \( |\tau_\mu + q| > k_0 \), which we denote as "\( \mu > \)". This sum involves infinite number of terms. We let

\[ S^{(q)}(r) = \frac{\pi e^2}{m\omega_0 a^2} \sum_{\mu >} \frac{(\tau_{\mu x} + q_x)^2 - k_0^2}{|g_\mu|} \exp[-|g_\mu|l|r| \cdot \hat{2} + i (\tau_\mu + q) \cdot r] \]

\[ = 3\pi n(2) \chi^2 \frac{\Gamma_\mu^{(q)}}{2} \sum_{\mu >} \frac{(\tau_{\mu x} + q_x)^2 - k_0^2}{|g_\mu|} \exp[-|g_\mu|l|r| \cdot \hat{2} + i (\tau_\mu + q) \cdot r] \]

\[ (A4-5) \]

which diverges as \( r \) goes to zero.

Therefore the interaction between the \( j \)th oscillator and the rest of the system [RHS of Eq. (A4-1)] can be summarized as,

\[ \text{RHS} = e \delta \sum_i E_{ij}^{(q)} = 2m\omega_0 \left( \sum_{\mu <} \Gamma_\mu^{(q)} - \lim_{r \to 0} \left[ S^{(q)}(r) - S_0(r) \right] \right) X_j^{(q)}. \]

\[ (A4-6) \]

On the other hand, for the normal mode motion, the left handside of Eq. (A4-1) is equivalent to

\[ \text{LHS} = m(\omega_0^2 - \omega^2 - i\Gamma \alpha) X_j^{(q)} = 2m\omega_0 (\omega_0 - \omega - \frac{\Gamma \alpha}{2}) X_j^{(q)}. \]

\[ (A4-7) \]
We note that both the left and the right handside are proportional to $X_j^{(q)}$ which proves that the Bloch wave motion that we assumed earlier in Eq. (5-1-1) is indeed the normal mode of the system.

Substitute this and $S_0(r)$, $S^{(q,\mu\alpha)}(r)$, $\Gamma_\mu$ of right handside back into Eq. (A4-1), we get the new form of the equation of motion,

$$[\omega_0 + \delta \omega^{(q)} - i \frac{1}{2} (\Gamma_\alpha + \sum_{\mu<} \Gamma_\mu^{(q)})] X^{(q)} = \omega X^{(q)},$$  \hspace{1cm} (A4-8)

where

$$\delta \omega^{(q)} = \lim_{r \to 0} [S^{(q)}(r) - S_0(r)]$$  \hspace{1cm} (A4-9)

is the frequency shift for the Bloch mode $q$.

The normal mode equation of motion can be put into the "effective Hamiltonian" form,

$$\mathcal{H}^{(q)} X^{(q)} = \omega X^{(q)},$$  \hspace{1cm} (A4-10)

where

$$\mathcal{H}^{(q)} = \omega_0 + \delta \omega^{(q)} - i \frac{1}{2} (\Gamma_\alpha + \sum_{\mu<} \Gamma_\mu^{(q)})$$  \hspace{1cm} (A4-11)

with $\delta \omega^{(q)}$ defined by Eq. (A4-9) and $\Gamma_\mu^{(q)}$ defined by Eq. (A4-4).
Appendix IV. Mathematica$^\text{TM}$ Programs

In this appendix, we list the Mathematica$^\text{TM}$ programs that are used in the thesis to generate different figures. In particular, program 1 calculates the normal mode frequencies and decay rates for a multi-layer system under "two wave approximation"; program 2 calculates the radiation intensity for a multilayer system after a synchrotron pulse excitation.

Both programs listed here have actually been run with Mathematica$^\text{TM}$ (enhanced Macintosh version 1.2).
(********** Program for Figs. 21,22,26,27,28,29 **********)
(*****************************************************************)

(***** This program calculate s the normal mode *******
(***** frequency shifts and normal mode decay ******
(***** rates for system of multi-layers under ********
(***** " two wave approximation " ******************

(***** note that the parameter n and div has to be ****
(***** changed to fit each plot **********************)

ClearAll;

n=3; (*********** number of layers **************)
gmin=0; (***** the starting ga value to plot *******)
gmax=N[Pi]; (***** the end ga value to plot **********)
div=30; (***** the number of data points **********)
g[i_]:=gmin+(gmax-gmin)/div*(i-1);
q[z_\_g_\_] := If[(z*Exp[I*g])==0, 0,
                  I*(1/(1-z*Exp[I*g])-1/(1-z*Exp[-I*g]))];

(********** In the following calculation, there are three
often used indices, i, j, k. i is always used to denote
different g values; j is always used to denote different
modes; k is always used to denote different layers ****)

(********** Part One: SIN Solutions **********)

(********** Solutions to the Equations ***********)
zs=Table[Chop[z/.(N[Solve[
        z^(n+1)-Exp[I*g[i]]*z^n+(z*Exp[I*g[i]]-1)
        ==0,z]]),{i,1,div+1}];

OpenWrite["zs.dat"];     
Write["zs.dat",zs];
Close["zs.dat"];
Q(w)=w0-i*Gamma(w)  

checks=Table[Chop[Sum[((zs[[i]][[j]])^((k-1)-(zs[[i]][[j]])^((n-k))^2},{k,1,n}]],
{i,1,div+1},{j,1,n+1}];

qs=Table[If[checks[[i]][[j]]==0,
0,
N[q[zs[[i]][[j]],g[i]]],{i,1,div+1},{j,1,n+1}];

OpenWrite["qs.dat"];Write["qs.dat",qs];Close["qs.dat"];
Clear[qs,zs,checks];

****************** Part Two: COS Solutions ******************

****************** Solutions to the Equations ******************

zc=Table[Chop[z/.(N[NSolve[
   z^(n+1)-Exp[I*g[i]]*z^n-(z*Exp[I*g[i]]-1)
   ==0,z]]),{i,1,div+1}];

OpenWrite["zc.dat"];Write["zc.dat",zc];Close["zc.dat"];
(************** Part Three: Frequency Shifts **************)

(******************* SIN Solutions *******************)
qs = <<qs.dat;

(******************* Complex Frequency Shifts **************)
dws = -qs;
Clear[qs];

(******************* Real Frequency Shifts *******************)
rws = Table[{{g[i], Re[dws[[i]][[j]]]}, {i, 1, div+1}, {j, 1, n+1}}];
Clear[dws];

(******************* Prepare for the plots *******************)
rws = Flatten[rws];
rws = Partition[rws, 2];

(*** Plot for Real Frequency Shifts as function of g **)
(******************* SIN Solutions *******************)
prws = ListPlot[rws];
Clear[rws];

qs = <<qs.dat;

(******************* Complex Frequency Shifts **************)
dws = -qs;
Clear[qs];
(************** the Decay Rates: \( G = -2\text{Im}(w) \) **************)

defays=Table[{g[i],-Im[dws[[i]][[j]]]},
{i,1,div+1},{j,1,n+1}];
Clear[dws];

(************** Prepair for the plots **************)
defays=Flatten[defays];
defays=Partition[defays,2];

(****** Plot for Decay Rates as function of g ******)
(*********************** SIN Solutions ***********************)

pdecays=ListPlot[defays,PlotRange->{0,n+1}];
Clear[defays];

(*********************** COS Solutions ***********************)

cos=<<cos.dat;

(******************** Complex Frequency Shifts **************)
dwc = -cos;
Clear[cos];

(*********************** Real Frequency Shifts ***********************)

dwc=Table[{g[i],Re[dwc[[i]][[j]]]},
{i,1,div+1},{j,1,n+1}];
Clear[dwc];

(*********************** Prepair for the plots ***********************)
rwc=Flatten[rwc];
rwc=Partition[rwc,2];
(** Plot for Real Frequency Shifts as function of g **)  
(********************** COS Solutions **********************)

prwc=ListPlot[rwc];

Clear[rwc];

qc=<qc.dat;

(********************** Complex Frequency Shifts **********************)

dwc = -qc;

Clear[qc];

(********************** the Decay Rates: G = -2*Im(w) ***************)

(** The equation does not have the factor 2 because **)  
(** we have changed the unit ***********************)

decayc=Table[{{g[i], -Im[dwc[[i]][[j]]]},  
{i,1,div+1},{j,1,n+1}}];

Clear[dwc];

(********************** Prepair for the plots **********************)

decayc=Flatten[decayc];

decayc=Partition[decayc,2];

(******** Plot for Decay Rates as function of g ********)  
(********************** COS Solutions ***********************)

pdecayc=ListPlot[decayc,PlotRange->{0,n+1}];

Clear[decayc];

pdecay=Show[pdecayc, pdecays]

prw=Show[prwc,prws]
(**********************)
(** The program for Figs. 23, 24, 25, 30, 31, 32, 33 **)  
(**********************)

(******** this program calculates and then plots the ********)
(******** radiated intensity for multilayer system in both **)  
(******** the forward scattering and the Bragg reflection **)  
(******** directions. Comparisons with the Born approximation*)  
(******** results are made. ***********************

(n=3; (****** the number of layers *********)

q=N[Pi]; (****** g0a value ***********************

q[z_, g_] := If[(z*Exp[I*g]) == (z*Exp[-I*g]), 0, 
                 I*(1/(1-z*Exp[I*g])-1/(1-z*Exp[-I*g]))];

(******** In the following calculation, there are three 
often used indices, i, j, k. i is always used to denote 
different g values; j is always used to denote different 
mode; k is always used to denote different layers ****)

(*********** Part One: SIN Solutions ***********)

(*********** Solutions to the Equations ***********

zs=Table[Chop[z/.(N[Solve[
    z^(n+1)-Exp[I*g]*z^n+(z*Exp[I*g]-1)
    ==0, z])])];

norms=Table[Chop[
    Sqrt[Sum[((zs[[j]])^(k-1)-(zs[[[j]])^(n-k))^2, 
             {k,1,n}]),{j,1,n+1}];

xs=Table[If[norms[[j]]==0,
            0,
            Chop[((zs[[j]])^(k-1)-(zs[[[j]])^(n-k))/
                 norms[[j]]),{j,1,n+1},{k,1,n}];

(*************** Q(w)=w0-iGama-w **********************

qs=Table[N[q[zs[[j]],g]],{j,1,n+1}];
(*************** Part Two: COS Solutions ***************)

(******************* Solutions to the Equations *******************)

zc=Table[Chop[z/.(N[Solve[
z^n*(1-Exp[I*G]*z^n-(z*Exp[I*G]-1)
==0,z])])]];

normc=Table[Chop[
  Sqrt[Sum[((zc[[j]])^(k-1)+zc[[j]])^(n-k)]^2,
  {k,1,n}],[j,1,n+1]];

xc=Table[If[normc[[j]]==0,
  0,
  Chop[((zc[[j]])^(k-1)+zc[[j]])^(n-k)]/
  normc[[j]]],[j,1,n+1],[k,1,n]];

(*********************** Q(w)=w-i*G-w ***********************)

qc=Chop[Table[N[q[zc[[j]],g]],[j,1,n+1]]];

(*************** Part Three: Frequency Shifts ***************)

(******************* SIN Solutions *******************)

dws = -qs;

(******************* COS Solutions *******************)

dwc = -qc;

(******************* Initial Conditions *******************)

x0=Table[Exp[I*G*k],[k,0,n-1]];

(******************* Expansion Coefficients *******************)

as=Table[1/2*Sum[xs[[j]][[k]]]*x0[[k]],
{k,1,n}],[j,1,n+1]];

ac=Table[1/2*Sum[xc[[j]][[k]]]*x0[[k]],
{k,1,n}],[j,1,n+1]];

(******************* Radiated Intensity *******************)
(* +z direction *)
rp1[t_] := Sum[as[[j]]*qs[[j]]*xs[[j]][[n]]*Exp[-2*I*t*dws[[j]]], {j, 1, n+1}]

rp2[t_] := Sum[ac[[j]]*qc[[j]]*xc[[j]][[n]]*Exp[-2*I*t*dwc[[j]]], {j, 1, n+1}]

radp[t_] := (Abs[rp1[t] + rp2[t]])^2

(*********** radiated intensity in the \( +z \) direction *******)

prdp = Plot[radp[t], {t, 0, 1}, PlotRange -> {0, n^2+1}]

(********* reference curve according to Born approx. ****)
p2 = Plot[n^2*Exp[-(1/n^2*(Sin[n*g]/Sin[g])^2)*2*t]), {t, 0, 1}, PlotStyle -> Dashing[{0.03, 0.03}], PlotRange -> {0, n^2+1}];

(* put the reference curve and radiation curve together*)
Show[prdp, p2]

(* -z direction *)

rn1[t_] := Sum[as[[j]]*qs[[j]]*xs[[j]][[1]]*Exp[-2*I*t*dws[[j]]], {j, 1, n+1}]

rn2[t_] := Sum[ac[[j]]*qc[[j]]*xc[[j]][[1]]*Exp[-2*I*t*dwc[[j]]], {j, 1, n+1}]

(****** the radiated intensity in the \( -z \) direction ******)

radn[t_] := (Abs[rn1[t] + rn2[t]])^2

prdn = Plot[radn[t], {t, 0, 1}, PlotRange -> {0, n^2+1}];

(** put the reference curve and the \( \pm z \) direction ******)
(** radiation together **************************************)

Show[prdp, prdn, p2];

(******************************* Log plot ******************************)
(****** \( +z \) direction ******)

np = radp[0];

p1 = Plot[Log[10, radp[t]/np], {t, 0, 10}];

(*************** reference curve: Born approx. *******)
\[ p2 = \text{Plot}[\log_{10}\left(\exp\left(-\left(1/n^* (n^* n^* (\sin[n^* g]/\sin[g])^2)^2\right)\right)),\{t, 0, 10\}, \text{PlotStyle}\to\text{Dashing}[\{0.03, 0.03\}] ] \]

\text{Show}[p1, p2]

(* -z direction *)

\text{Plot}[\log_{10}(\text{radn}[t]/np),\{t, 0, 20\}]
References


(14) J. P Hannon, Rice University M. A. Thesis, Appendix (B).