Quantum Properties of Light Emitted by Dipole Nano-Laser

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QUANTUM PROPERTIES OF LIGHT EMITTED BY DIPOLE NANO-LASER

by

Talal Ghannam

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Submitted to the
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Dr. Alvin Rosenthal, Advisor

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QUANTUM PROPERTIES OF LIGHT EMITTED FROM NANO-LASER

Talal Ghannam, Ph.D.
Western Michigan University, 2007

Recent technological advances allow entire optical systems to be lithographically implanted on small silicon chips. These systems include tiny semiconductor lasers that function as light sources for digital optical signals. Future advances will rely on even smaller components. At the theoretical limit of this process, the smallest lasers will have an active medium consisting of a single atom (natural or artificial). Several suggestions for how this can be accomplished have already been published, such as nano-lasers based on photonic crystals and nano wires. In particular, the “dipole nanolaser” consists of a single quantum dot functioning as the active medium. It is optically coupled to a metal nanoparticles that form a resonant cavity. Laser light is generated from the near-field optical signal. The proposed work is a theoretical exploration of the nature of the resulting laser light.

The dynamics of the system will be studied and relevant time scales described. These will form the basis for a set of operator equations describing the quantum properties of the emitted light. The dynamics will be studied in both density matrix and quantum Langevin formulations, with attention directed to noise sources. The equations will be linearized and solved using standard techniques. The result of the study will be a set of predicted
noise spectra describing the statistics of the emitted light. The goal will be to identify the major noise contributions and suggest methods for suppressing them. This will be done by studying the probability of getting squeezed light from the nanoparticle for the certain scheme of parameters.
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At the beginning of this dissertation, I would like to thank God, for that virtually everything I have accomplished was due to his grace and mercy.

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Finally I wish to thank my committee members; Dr. Nikolaus Stolterfoht and Dr. Clement Burns and all the faculty members who taught me and the students who I worked with. It was a real joy working with everyone in the Department of Physics at WMU. Thank you all.

Talal Ghannam
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CHAPTER I

INTRODUCTION

Introduction

During the past few years the need for small optical elements to replace the current electronic ones has become more and more urgent, especially for communications and the new generations of optical computers. One essential element is a very small laser that can generate light efficiently and reliably. Many designs for making nano-scale lasers have been proposed in the literature, and some have been realized in the laboratory. Almost all of these nano-lasers produce low-noise laser light, the main difference between them being their sizes. They are all called nano-lasers but because almost all of them require an optical cavity their sizes are on order of the wave length of the light emitted, i.e. micron size for optical wavelengths. This would seem to be a basic limitation since the very concept of a laser is usually taken to be an optical amplifier plus a cavity.

The laser concept can be extended beyond the conventional amplifier + cavity model: boson fields other than the electromagnetic field can be made to "lase" and these can in turn radiate light. Several suggestions have been put forward in the literature. A "laser" that doesn't require a cavity, like the one we investigate in this work, would be a truly nanometer scale object and thus ideal for many applications.

In this paper we carry out a theoretical and computational investigation of a dipole nanolaser, (DNL), a device proposed by (I. E. Protsenko, et al, 2005). The device

1
under study consists of a quantum dot, whose electronic oscillations act, through their near fields, to pump the dipole plasma modes of a metallic nano-particle. Far-field radiation is generated by coherent contributions of both the quantum dot and plasma oscillations. We describe this device in detail in this work. It is probably the smallest device that can conceivably be produced with current or foreseeable technology that deserves the name “laser”.

The basis for the device is the set of so-called “electrostatic resonances” of a small metallic or semi-conducting particle. These are modes that occur when the particle is placed in a dielectric matrix and excited at a frequency for which the real part of the matrix dielectric constant is negative, i.e. working near an optical resonance (Isaak D. Mayergoyz et al 2005). Large amplitude plasma oscillations can be readily induced on the nanoparticle in this circumstance.

The quantum dot is an artificial atom consisting of one or more electrons confined to a fixed volume with the aid of external voltages (P. Hawrylak et al 2000). The dot (called a two-level system or TLS in this work) can be excited using simple methods, such as applying suitable ac voltages to electrodes surrounding the quantum dot. Changing the voltage difference across the quantum dot will change its energy, and hence excite or de-excite the dot. The oscillating electron(s) of the dot are electrostatically coupled to the resonant dipole plasma modes of the nanoparticle (NP). A build-up of dipole amplitude occurs, similar to the build-up of a cavity field in a laser. De-excitation of the nanoparticle can occur either Ohmically or by the emission of electromagnetic radiation.
The proposed nanolaser configuration is shown below;

![Diagram of dipole nanolaser DNL configuration](image)

Figure 1.1. The dipole nanolaser DNL configuration.

In the paper introducing the idea of the DNL, Protsenko et. al. showed that the equations describing the plasmon oscillations of the NP induced by the TLS map isomorphically onto the standard equations describing an electromagnetic field in a laser cavity produced by an active medium. The analog of the active medium is the TLS and the analog of the laser cavity field is the plasmon amplitude. This work examined a number of conditions necessary to produce plasmon "lasing" and found that most of these were at, or near, the limits of what is currently technologically feasible. Left uninvestigated were the properties of the light such a device would produce. Below we give a partial survey of several devices that have been called "nanolasers" and we compare them to the dipole nanolaser.
One technique for a small laser involves the usage of photonic crystals (M. Loncar, et al. 2002). In this kind of device, holes are drilled in a substrate producing a so-called photonic crystal and the lasing modes are confined to the region external to these holes. Each hole is about 200nm in radius and the entire device encompasses at least $10^5$ holes. Although sometimes called a nanolaser, this should be thought of as a macroscopic device with nanometer components. The quality factor of a photonic crystal cavity is on the order of $10^3$ implying an optical line width on the order of $10^5$ MHz, while our DNL will have a quality factor of 50-5000 depending on the regime of numbers we are working with, thus the line width of the dipole nanolaser is estimated to be about $10^5$-$10^8$ MHz when all important sources of damping are taken into consideration. This is explained in detail in chapter four. On the other hand, photonic crystal nanolasers can work at room temperature while the ideal working temperature for the dipole nanolaser considered in this work is less than 100K.

D.J. Bergman and M.I. Stockman (2003) proposed a nano laser (called a Spaser, short hand for surface-plasmon amplification by stimulated emission of radiation) which is very similar to the dipole nanolaser in the sense that it doesn’t require a cavity, and it works by stimulating surface plasmons. But while the sizes of the spaser are comparable to ours, the spaser generates only near field electromagnetic modes, while the dipole nanolaser generates coherent dipole radiation. Also the spectral width of the spaser will be around $10^{13}$-$10^{14}$ Hz which is comparable to our DNL. But in their paper they don’t mention anything about the possibility of decreasing the line width as we will later on show can be achieved in our system for certain numbers regime.
Nano lasers can also be achieved by employing only quantum dots (S. Hoogland et al 2006). Quantum dot lasers are significantly superior to conventional semiconductor lasers, in that they feature higher performance in such aspects as temperature-independent operation, low power consumption, long-distance transmission, and fast speeds. In this system one or more electrons are confined to a potential well established by nearby electrodes. By manipulating the voltage across the electrode one can excite the electron which in turn emits light. Many quantum dots acting coherently produce significant intensities. These devices can work at room temperature but are designed for high bit transmission rates rather than for generating low-noise light that will itself be further processed optically. The spectral width of this laser is about $10^{11}$ Hz. Not far from our line width produced using our special parameter regime.

Another technology is the nano-wire lasers (Peter J. Pauzauskie et al 2006). Here a wide band gap semiconductor nanostructures with near-cylindrical geometry and large dielectric constants exhibit two-dimensional ultraviolet and visible photonic confinement (i.e. waveguiding). The nano wire works as a Fabry-Perot Cavity and a gain medium for light amplification. The spectral width of this laser is on the order of $10^{11}-10^{14}$ which is comparable to our spectral width. The typical cross section of the nano wire is on the order of 100nm, this makes it much bigger than our system.
To compare the proposed TLS-NP system to a typical laser, consider first the cavity 
quality factor $Q$. This determines the efficiency of the cavity of the laser and is defined 
by the following:

$$Q = \frac{\omega_0}{\nu} \frac{\text{Stored Energy}}{\text{Power Loss}} \frac{\omega_0}{\nu}$$  

(1.1)

Where $\nu$ is the damping of the cavity field due mainly to mirror transmission. The 
corresponding quantity entering the equations of the TLS-NP model is also $\frac{\omega_0}{\Gamma}$ where 
$\omega_0$ is the optical frequency and $\Gamma$ is the decay rate of the polariton field due to Ohmic and 
radiative mechanisms. This damping rate is on the order, $\gamma = 10^{12} - 10^{15}$ Hz, depending on 
the parameter regime used, and at sufficiently low temperatures is completely dominated 
by radiative damping. So the "quality factor" is on the order of 10-1000. For a standard 
laser, the quality factor determines the photon residence time in the cavity and its inverse 
is therefore a measure of the laser line width and is on the order of $10^8$ and above for 
regular lasers. In our case too, the radiated spectrum will have line width proportional to 
$1/Q$.

Another convenient feature of the proposed DNL is the number of photons emitted from 
it. This can be roughly estimated by calculating the power of the emitted light produced 
from a dipole using the standard Larmor formula:

$$P = \frac{Z_0}{12\pi c^2} |\vec{p}|^2$$  

(1.2)

where $Z_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}} \approx 377 \Omega$ and $\vec{p} = |\vec{p}_0|\sqrt{N_0}$ where $N_0$ is the number of plasmons and $\vec{p}_0$ 
the induced dipole moment of the NP. The rate at which photons are emitted is $R = \frac{P}{\hbar \omega}$
and it will be shown in the subsequent chapters that for reasonable parameter values this rate is on the order of $10^{11}$ photon/sec. Considering the rate reduction arising from coupling to optical elements such as fiber optics, this rate is quite convenient for quantum optics, and single or multi photon measurements. We will see in our detailed quantum calculation that the emission rate agrees with the above estimate when the optical line width is small, but is an under-estimate for the more reasonable situation of a large line width.

Another characteristic feature of lasers is that the line width of the spectrum decreases with increasing the number of photons in the cavity. This, as it will be shown in chapter three, is exactly what we find for the line width of our system, along with other contributing factors. This feature is usually considered as a signature of lasing, which indicates that we are indeed getting lasing or coherent plasmon excitation from our system.

In chapter two of this thesis we set up Hamiltonian for the various elements and reservoirs. The equations of motion for the system are then formulated by using the Heisenberg equation of motion. We then calculate the drift and diffusion terms for the various operators to reach the final form of the equations of motion for the various operators.
**Surface Plasmon**

Plasmons are collective oscillations of the free electron gas density, often at optical frequencies. They can also couple with a photon to create what is called a plasma polariton.

Surface plasmons are those plasmons that are confined to surfaces and that interact strongly with light resulting in a polariton. They occur at the interface of a vacuum or material with a positive dielectric constant with that of a negative dielectric constant.

The properties of these plasmons/polaritons can be computed by using the Mie theory. The full theory is given in the appendix.

We consider a metallic sphere of permittivity $\varepsilon_1$ embedded in a medium of permittivity $\varepsilon_2$. The sphere is subjected to an incident field.

We can write the fields inside and outside the sphere in terms of spherical Bessel functions and vector spherical harmonics:

$$\vec{E}_{\text{out}} = \vec{E}_{\text{inc}} + \vec{E}_{\text{sc}}$$

Where $E_{\text{out}}$ is the outgoing field, $E_{\text{inc}}$ is the field incident on the nano-particle and $E_{\text{sc}}$ is the scattered field.

$$\vec{E}_{\text{inc}} = \vec{E}_0 e^{i\kappa z} = \sqrt{4\pi} \vec{E}_0 \sum_l i^l \sqrt{2l+1} j_l(\kappa r) Y_l^0(\hat{r})$$

$$\vec{H}_{\text{inc}} = \left(\frac{\hat{z} \times \vec{E}_0}{Z_2}\right) e^{i\kappa z} = \sqrt{4\pi} \left(\frac{\hat{z} \times \vec{E}_0}{Z_2}\right) \sum_l i^l \sqrt{2l+1} j_l(\kappa r) Y_l^0(\hat{r})$$

We take the general form $\vec{E}_0 = E_0(\cos \alpha \hat{x} + e^{i\theta} \sin \alpha \hat{y})$. Written in spherical form:

$$(E_0)_\theta = -\frac{1}{2} \left(\left(E_0\right)_z + i \left(E_0\right)_r\right) = -\frac{E_0}{\sqrt{2}} \left(\cos \alpha + ie^{i\theta} \sin \alpha\right)$$

$$(E_0)_\phi = \frac{1}{2} \left(\left(E_0\right)_z - i \left(E_0\right)_r\right) = \frac{E_0}{\sqrt{2}} \left(\cos \alpha - ie^{i\theta} \sin \alpha\right), \quad (E_0)_0 = (E_0)_z = 0$$
The internal and scattered fields are

\[
\mathbf{E}_{\text{int}} = Z_j \sum_{l,m} \left\{ \frac{i}{k} a_{lm}^{(E)} \nabla \times \left[ j_l(kr) \mathbf{X}_m^l \right] + a_{lm}^{(M)} j_l(kr) \mathbf{X}_m^l \right\}
\]

\[
\mathbf{H}_{\text{int}} = \sum_{l,m} \left\{ a_{lm}^{(E)} j_l(kr) \mathbf{X}_m^l - \frac{i}{k} a_{lm}^{(M)} \nabla \times \left[ j_l(kr) \mathbf{X}_m^l \right] \right\}
\]

\[
\mathbf{E}_{\text{sc}} = Z_j \sum_{l,m} \left\{ \frac{i}{\kappa} b_{lm}^{(E)} \nabla \times \left[ h_l^i(kr) \mathbf{X}_m^l \right] + b_{lm}^{(M)} h_l^i(kr) \mathbf{X}_m^l \right\}
\]

\[
\mathbf{H}_{\text{sc}} = \sum_{l,m} \left\{ b_{lm}^{(E)} h_l^i(kr) \mathbf{X}_m^l - \frac{i}{\kappa} b_{lm}^{(M)} \nabla \times \left[ h_l^i(kr) \mathbf{X}_m^l \right] \right\}
\]

Where \( Z_j = \sqrt{\frac{\mu_0}{\varepsilon_j}} \) is the wave impedance of the medium. All multipole amplitudes \( a_{lm} \) and \( b_{lm} \) have units of \( A/m \) and are given in the appendix. Where we used the (E) modes only (J.D. Jackson 1999).

Now by using the correct boundary conditions, and after lots of algebra, we can find a form for the electric field everywhere and hence the induced dipole moment of the sphere.

The dipole moment is given by the following equation:

\[
|\mathbf{d}| = \sqrt{\frac{8\pi}{3}} \frac{Z_j a^2}{k} j_1(ka) \sqrt{\sum_{M} \left| a_{1M}^{(E)} \right|^2 (\varepsilon_2 - \varepsilon_1)} \tag{1.3}
\]

In the long wavelength approximation we get the following form of the induced dipole moment:

\[
|\mathbf{d}| = 4\pi \varepsilon_2 \frac{\left| \varepsilon_2 - \varepsilon_1 \right| a^3}{(\varepsilon_1 + 2\varepsilon_2) E_0} \tag{1.4}
\]

The below curve shows the dipole moment for a silver nanoparticle in a silicon matrix, at a fixed radius of 7nm, as a function of frequency. The frequency range is \( 0.1 - 10.0 \times 10^{15} \) Hz. Low frequency behavior is determined the electro-static dipole moment.
structure at 2.4 \( pHz \) is the plasmon resonance for this particle-matrix system: the permittivity of silver and silicon are structureless in this frequency range.

\[ |d(\omega_c)| \]

\( \omega_c \)

Figure 1.2. The dipole moment \( d \) (C.m) of a 7 nm silver sphere versus frequency (Hz). The resonance is clearly shown on the graph.

The plasmon energy density is practically constant throughout a nanoparticle of radius < 20nm because of the centrifugal barrier. Therefore, the plasmon occupies the entire volume of the particle. In effect, the entire particle is a “surface” for the “surface plasmon”.

\[ r \]

Figure 1.3. Plasmon energy density (arbitrary units) vs. distance (in nm) from the center of a=7.0nm radius silver particle in a silicon matrix.
Light generated by radiating plasmons will have an angular distribution characteristic of dipole radiation. In the below graphs the Mie cross-sections are as shown for a 7nm silver particle in a silicon matrix at the plasmon resonance with outgoing polarization along the y-axis.

Figure 1.4. Dependence of differential cross section on polar angle ($\theta = 0..\pi$) at the azimuthal angle $\phi = 0$. $\theta$ is taken in respect to the z axis.

The below graph shows the dipole moment for particles of different radii as a function of volume at one particular incident frequency ($1\text{pHz}=10^{15}\text{Hz}$). The plot extends from $r = 0$ to 20nm. (The appearance of a slight curvature is deceptive, detailed analysis shows that the curve is a straight line).
Figure 1.5. Dependence of the dipole moment (C.m) on the volume of the particle $r^3$ (nm$^3$).

From the above we see that surface plasmon arises naturally and classically in any nanoparticle subjected to an electromagnetic field with the right frequency. These plasmons, together with the oscillating charge of the quantum dot, form the source of the radiation field we study in this work.

In chapter two we describe the Hamiltonian of the coupled quantum dot and nanoparticle subjected to an external heat bath and use this to derive the Langevin-type equations of motion. In chapter three we solve for the steady state solutions which enable us to find the threshold conditions for the lasing operation, the spectrum of the intensity of the light, the number of photon emitted and spectral line width. Then we linearize the equations by expanding each operator in terms of a constant large term and small fluctuation term. We then Fourier transform the linearized equations of motion: this enables us to solve them and find the various quadratures of the system and to find the squeezing spectrum. In chapter four we conclude our research by examine different parameter regimes and show
that there are two extreme types of DNL, one corresponding to polariton lifetimes very long compared to the quantum dot lifetime and the other to very short polariton lifetimes.
CHAPTER II

EQUATIONS OF MOTION

General Outline of the Calculations

We will carry out our analysis of the quantum optical properties of the dipole nanolaser using the Langevin formalism to model stochastic processes such as spontaneous emission and thermal fluctuation. In the Langevin formalism the equations of motion take the following form:

\[
\frac{d}{dt} \nu = -\Gamma_{\nu}(t) \nu(t) + F_{\nu}(t)
\]  

(2.1)

Where \( \nu \) is a system variable, \( \Gamma_{\nu}(t) \) is the some function of system variables, and \( F(t) \) represent a fluctuating noise term (physically identified with the coupling of the system...
to its larger environment) with zero ensemble average $\langle F_\nu(t) \rangle = 0$. We will be interested in the expectation value of the average bilinear product of this noise term (the correlation): $\langle F_\beta(t)F_\nu(t') \rangle$ where $\beta, \nu$ can refer to the same variable or to different ones.

This term represents the fluctuation of the external variables (vacuum electromagnetic field, thermal phonons, etc.) about their mean values. In the Markoffian approximation, the time for significant change of the noise term $F(t)$ is much smaller than other relevant time scales and we can write:

$$\langle F_\nu(t)F_\nu(t') \rangle = 2D_{\nu \nu} \delta(t-t') \quad (2.2)$$

Where $D_{\nu \nu}$ is a constant term that determines the magnitude of the fluctuating forces, and is called the diffusion coefficient. It can also be shown that $D_{\nu \nu}$ may be written in terms of the expectation value of the product of the variable and its noise term (M. Sargent et al 1974):

$$D_{\nu \nu} = \langle \nu(t)F_\nu(t) \rangle. \quad (2.3)$$

An entirely equivalent formalism uses the Fokker-Planck equation, which involves a drift term expressing the rate of change of system variables under the applied external forces and a diffusion term representing the fluctuations due to coupling to an external heat bath. These are related to the quantities $\langle \Gamma_\mu(t) \rangle, D_{\mu \nu}$ used in a Langevin approach. In this thesis we work with harmonic oscillator operators to model both the plasmon field of the nanoparticle and the external noise sources. Equations for the time development of the various operators can be obtained from the Heisenberg equation of motion:

$$\dot{a}_\nu(t) = -\frac{i}{\hbar} [H, a_\nu] \quad (2.4)$$

These are of the form of Eq. (2.1) when $H$ includes the coupling to the environment.
The drift term is calculated in lowest-order perturbation theory by the following relation:

\[ \langle D_{\rho}(t)B \rangle = -\frac{1}{\hbar^2} \int_0^\infty d\tau \left\langle \left[ V(t+\tau), [V(t),A_{\rho}(t)] \right] \right\rangle_B \]  

(2.5)

Where \( V(t) \) is the energy of interaction with the environment, and the average is taken over a reservoir or "bath" \( B \). The diffusion term can be calculated using Eq. (2.3). It is sometimes easier to calculate the drift term first and then find the diffusion from the generalized Einstein relation developed from the Langevin formalism, and given below (M. Sargent et al 1974):

\[ 2\left\langle D_{\rho\nu} \right\rangle_B = -\left\langle D_{\rho}(t)A_{\nu}(t) \right\rangle_B - \left\langle A_{\rho}(t)D_{\nu}(t) \right\rangle_B + \frac{d}{dt}\left\langle A_{\rho}(t)A_{\nu}(t) \right\rangle_B \]  

(2.6)

We model the electronic states of the quantum dot by two states (ground and excited) and represent them by the Pauli operators (\( \sigma \)'s). For brevity, this two-level system will be denoted TLS. The metallic nano-particle dipole amplitudes are represented by the harmonic oscillator's operators (\( \hat{a}, \hat{a}^\dagger \)), and their slowly varying counterparts. The TLS decays by spontaneous emission, a process represented by coupling to a reservoir "C". The plasmons decay into random thermal excitations of the NP as well as by emission of light: these decays are modeled by coupling to another reservoir "A". We take the radiation explicitly into account in the later stages of this work. The TLS and nano-particle interact with each other through a dipole-dipole interaction \( V(t) \).

Once we have the equations of motion for the system, we look for the drift and diffusion terms: to find the mean value, and fluctuation of the dipole moment. The radiated electric field is given by the coherent sum of TLS and NP contributions. The
formalism is useful for computing the spectral distribution of the emitted light, which depends on the temporal fluctuations via:

\[
I(\omega) = \int_{-\infty}^{\infty} dt \ e^{-i\omega t} \left\langle E(t) E(0) \right\rangle
\]  

(2.7)

To calculate the plasmon squeezing spectrum, we write the plasmon field “A” as a constant term plus a fluctuating term: \( \hat{A}(t) = (|A_0| + \delta \hat{A}(t)) \), in the same way our quadratures can be written as a constant term plus fluctuating one:

\[
\hat{X} = X + \delta \hat{X}, \quad \hat{Y} = Y + \delta \hat{Y}
\]  

(2.8)

From this we can get the squeezing spectrum after Fourier transforming:

\[
S_x(\omega) = \left\langle |\delta X(\omega) \delta X(-\omega)| \right\rangle, \quad S_y(\omega) = \left\langle |\delta Y(\omega) \delta Y(-\omega)| \right\rangle
\]  

(2.9)

and by changing some parameters of the configuration one can see whether a squeezing is achievable or not.

**The Hamiltonian**

Let \( a_H, \sigma_H \) denote Heisenberg operators for the nano-particle and the TLS respectively where we are using \( \sigma \)'s to denote the Pauli matrices, or rather, operators that, in the Heisenberg picture are equal to the Pauli matrices at time zero. We have

\[
[\sigma_-, \sigma_+] = 2\sigma_z, \quad [\sigma_-, \sigma_+] = -\sigma_z
\]

where \( \sigma_z \) are the raising and lowering operators that will take the quantum dot from the ground state to the excited state and vice versa, and \( \sigma_z \) is the population difference \( \sigma_z = |2\rangle \langle 2| - |1\rangle \langle 1| \)

The unperturbed system Hamiltonian is:

\[
H_{sys,0} = \hbar \omega_b a_H^\dagger a_H + \frac{\hbar \omega_s}{2} \sigma_{H,z}
\]  

(2.10)
In terms of slowly varying operators we have

\[ a_H = a e^{-i\omega_H t}, \quad \sigma_{H,-} = \sigma_- e^{-i\omega_H t}, \text{ etc.} \]

We follow Bergman and Stockman in writing the Coulomb electric field produced by the plasmon as (D.J. Bergman and M.I. Stockman 2003):

\[ E = -\nabla \phi, \quad \phi = \varphi(\vec{r}) \left( a(t) e^{-i\omega_H t} + a^\dagger(t) e^{i\omega_H t} \right) \]

(2.11)

where \( \varphi(\vec{r}) \) is a suitably normalized mode function that can be written as:

\[ \varphi(\vec{r}) = \frac{1}{4\pi\epsilon_0} \left( \frac{\vec{\mu}_0 \cdot \vec{r}}{r^3} \right), \] where \( \vec{\mu}_0 \) is the matrix element of the dipole moment for the nanoparticle. The TLS has a dipole moment operator \( \vec{\mu}_2(t) = \vec{\mu}_2 \left[ \sigma_-(t) e^{-i\omega_H t} + \sigma_+(t) e^{i\omega_H t} \right] \) and the dipole-dipole interaction energy is:

\[ V = -\vec{E} \cdot \vec{\mu}_2(t) \]
\[ = \frac{1}{4\pi\epsilon_0} \left( \frac{\vec{\mu}_0 \cdot \vec{r}}{r^3} \right) \left[ \sigma_-(t) e^{-i\omega_H t} + \sigma_+(t) e^{i\omega_H t} \right] \left[ a(t) e^{-i\omega_H t} + a^\dagger(t) e^{i\omega_H t} \right] \]

In the rotating wave approximation (RWA) we ignore high frequency components of this interaction to get

\[ V \approx \frac{1}{4\pi\epsilon_0} \left( \frac{\vec{\mu}_0 \cdot \vec{r}}{r^3} \right) \left[ \sigma_-(t) a^\dagger(t) e^{i(\omega_0 - \omega_H)t} + \sigma_+(t) a(t) e^{-i(\omega_0 - \omega_H)t} \right] \]

(2.12)

\[ = \hbar \Omega_{\text{res}} \left[ \sigma_-(t) a^\dagger(t) e^{-i\omega_2 t} + \sigma_+(t) a(t) e^{i\omega_2 t} \right] \]

The contributions of the reservoirs may be written as \( H_{\text{res}} = H_{\text{res},0} + H_{\text{irrev}} \) where

\[ H = H_0 + H_{\text{int}} \]
\[ H_0 = H_{\text{sys},0} + H_{\text{res},0} \]
\[ H_{\text{int}} = V + H_{\text{irrev}} \]

(2.13a)

\[ H_{\text{res},0} = \hbar \sum_j \omega_j \left\{ A^\dagger_j A_j + C^\dagger_j C_j \right\} \]

(2.13b)
In these equations, the $A$'s represent the combined effects of electromagnetic radiation and NP thermal phonons while the $C$'s represent the combined effects of electromagnetic radiation, thermal noise, and electronic pump noise contributing to decay and dephasing of the TLS. Detailed estimates of these effects are given in chapter 4. In Eq. (2.14) $\gamma$, $\gamma_2$ describe rates of dipole polariton damping and TLS spontaneous emission and damping due to other sources.

We work with the slowly varying counterparts of the system and reservoir operators. We seek operator equations of the form (2.1) for any system operator. The physics is completely determined by the reservoir averages of $D(t)$ and of $F(t)F(t')$.

**The Drift and Diffusion for the Nano-particle**

The equation of motion for the polariton amplitude is given by

$$\dot{a} = (i\hbar)^{-1} [a, H] = -i\Omega e^{i\omega_0 t} a - i \sum_j \sqrt{\gamma_j} A_j e^{-i(\omega_j - \omega_0) t}$$  

We therefore have $F_a(t) = -i \sum_j \sqrt{\gamma_j} A_j e^{-i(\omega_j - \omega_0) t}$. The drift term can be calculated by the following:

$$\frac{d}{dt} \langle a(t) \rangle_A = \langle D_a(t) \rangle_A = -\hbar^{-2} \int_0^\infty d\tau \langle [H(t + \tau), [H(t), a(t)]] \rangle_A$$

where $H(t)$ is the total Hamiltonian given by Eq. (2.13). The drift term then is given by $\langle D_a \rangle = -\gamma_2 \langle a(t) \rangle$. We therefore write, as an operator equation, where the extra term represents damping:

\[ H_{\text{res}} = \hbar \sum_j \sqrt{\gamma_j} \left( A_j e^{i(\omega_j - \omega_0) t} + A_j e^{-i(\omega_j - \omega_0) t} \right) + \sqrt{\gamma_{\text{det}}} \left( C_j e^{i(\omega_j - \omega_0) t} + C_j e^{-i(\omega_j - \omega_0) t} \right) \]  

(2.14)
\[
\frac{d}{dt} a = -\frac{\gamma}{2} a(t) - i\Omega_{im} e^{-\text{land}} \sigma_+ + F_a(t)
\]  
(2.16)

The formal solution is:

\[
a(t) = a(0) e^{-\gamma t/2} + \int_0^t dt' e^{-\gamma (t-t')/2} \left\{ -i\Omega_{im} e^{-\text{land}} \sigma_+ (t') + F_a(t') \right\}
\]  
(2.17)

The diffusion term can be calculated by the following:

\[
2 \text{Tr}_R \left[ D_{a,a} \right] \delta(t-t') = \text{Tr}_R \left[ F_a(t) F_a(t') \right]
\]  
(2.18)

Where

\[
\text{Tr}_R \left[ D_{a,a} \right] = \text{Tr}_R \left[ a^\dagger(t) F_a(t) \right]
\]
(2.19)

\[
= \text{Tr}_R \left\{ a^\dagger(0) F_a(t) e^{-\gamma t/2} + \int_0^t dt' e^{-\gamma (t-t')/2} \left\{ -i\Omega_{im} e^{i\omega t} \sigma_+ (t') + F_a(t') \right\} F_a(t) \right\}
\]

(2.19)

\[
= \text{Tr}_R \left\{ \int_0^t dt' e^{-\gamma (t-t')/2} F_a^\dagger(t') F_a(t) \right\} = \int_0^t dt' e^{-\gamma (t-t')/2} \text{Tr}_R \left[ F_a^\dagger(t') F_a(t) \right]
\]

(2.20)

\[
\text{Tr}_R \left[ F_a^\dagger(t') F_a(t) \right] = \sum_{j,k} \sqrt{\gamma_j \gamma_k} \text{Tr}_R \left[ A_j^\dagger(t') A_k(t) \right] e^{\frac{i}{\hbar} (\omega_j - \omega_k) t} e^{-i(\omega_j - \omega_k) t}
\]

(2.20)

\[
\text{Tr}_R \left( A_j^\dagger(t') A_k(t) \right) = \delta (t-t') \delta_{jk} \text{Tr}_R \left( A_j^\dagger(0) A_k(0) \right) = n^{\text{A}} \delta_{jk} \delta(t-t')
\]

(2.21)

so that

\[
\text{Tr}_R \left[ F_a^\dagger(t') F_a(t) \right] = \delta(t-t') \sum_j \gamma_j n^A_j \to \bar{n}^A \gamma \delta(t-t')
\]

(2.22)

where the final step, equivalent to the Markov approximation, is justified by the fact that the \( \gamma_j \) are nonzero only for \( |\omega_j - \omega_0|/\omega_0 \ll 1 \), and \( \bar{n}^A \) is the average number of quanta in the \( A \)-reservoir

\[
\text{Tr}_R \left[ D_{a,a} \right] = \int_0^t dt' e^{-\gamma (t-t')/2} \bar{n}^A \gamma \delta(t-t') = \bar{n}^A \gamma / 2
\]

(2.23)

Similarly,
It can be shown that for thermal reservoirs, the expectation value $[F_a(t')F_a(t)]$ is equal to zero.

**Drift Terms for the TLS**

Next, examine the equations of motion for TLS and its associated vacuum fields.

\[
\frac{d}{dt} \sigma_- = i\Omega_{\text{in}} \sigma_z e^{-i\omega_{\text{in}} t} + i \sum_j \left\{ \sqrt{\gamma_{j,2}} C_j \sigma_z e^{-i(\omega_j - \omega_2) t} \right\}
\]

\[
\equiv i\Omega_{\text{in}} \sigma_z e^{-i\omega_{\text{in}} t} + F_{C_-}
\]

Implementing Eq. (2.5) we find:

\[
\langle D_-(t) \rangle = -\hbar^{-2} \int_0^\infty d\tau \text{Tr}_R \left[ H(t+\tau), \left[ H(t), \sigma_- (t) \right] \right]
\]

\[
\left[ H(t), \sigma_- (t) \right] = -\hbar \sum_j \left\{ \sqrt{\gamma_{j,2}} C_j \sigma_z e^{-i(\omega_j - \omega_2) t} \right\}
\]

\[
\left[ H(t+\tau), \left[ H(t), \sigma_- (t) \right] \right] = -\hbar^2 \sum_{j,k} \left\{ \sqrt{\gamma_{j,2} \gamma_{k,2}} \left[ C_j(t+\tau) \sigma_z(t+\tau) e^{-i(\omega_j - \omega_2)(t+\tau)} + \sigma_- (t+\tau) C_k^\dagger(t+\tau) e^{i(\omega_k - \omega_2)(t+\tau)} \right] \right\}
\]

For a phase-incoherent reservoir, the trace of operators like $CC$ is zero. Therefore

\[
\left[ H(t+\tau), \left[ H(t), \sigma_- (t) \right] \right] = -\hbar^2 \sum_{j,k} \left\{ \sqrt{\gamma_{j,2} \gamma_{k,2}} \left[ \sigma_- (t+\tau) C_j^\dagger(t+\tau) e^{i(\omega_j - \omega_2)(t+\tau)}, C_k (t) \sigma_z (t) e^{-i(\omega_k - \omega_2) t} \right] \right\}
\]

The contributions of the $C$-dependent terms to the drift operator are:

\[
\text{Tr}_R \left[ D_{a_a^\dagger} (t) \right] = (\bar{n}^2 + 1) \frac{\gamma}{2}
\]

(2.24)
\[
\langle D_{c-}(t) \rangle = -\sum_{j,k} \sqrt{\gamma_{j,2} y_{k,2}} \int_{0}^{\infty} d\tau e^{-i(\omega_{j} - \omega_{k})\tau + 2\pi n c + \lambda} \left\{ \sigma_{-}(t + \tau) C_{j}^{\dagger}(t + \tau) C_{k}(t) \sigma_{-}(t) - \sigma_{-}(t) C_{j}^{\dagger}(t + \tau) C_{k}(t) \sigma_{-}(t + \tau) \right\}
\]

\[
= -\sum_{j,k} \sqrt{\gamma_{j,2} y_{k,2}} \int_{0}^{\infty} d\tau e^{-i(\omega_{j} - \omega_{k})\tau + 2\pi n c + \lambda} \left\{ \sigma_{-}(t + \tau) Tr_{p} \left[ C_{j}^{\dagger}(t + \tau) C_{k}(t) \right] \sigma_{-}(t) - \sigma_{-}(t) Tr_{p} \left[ C_{j}(t) C_{j}^{\dagger}(t + \tau) \right] \sigma_{-}(t + \tau) \right\}
\]

\[
= -\gamma_{j,2} \left\{ n_{j}^{c} \sigma_{-}(t + \tau) \sigma_{-}(t) - (n_{j}^{c} + 1) \sigma_{-}(t) \sigma_{-}(t + \tau) \right\} \delta(\tau)
\]  \hspace{1cm} (2.28)

\[
= -\frac{1}{2} \sum_{j} \gamma_{j,2} \left\{ n_{j}^{c} \left( \sigma_{-}(t), \sigma_{-}(t) \right) - \sigma_{-}(t) \sigma_{-}(t) \right\} = -\sum_{j} \gamma_{j,2} \left\{ 2n_{j}^{c} \sigma_{-}(t) + \sigma_{-}(t) \right\}
\]

\[
\rightarrow \frac{2}{\gamma_{j,2}} (2n_{j}^{c} + 1) \sigma_{-}(t)
\]

\[
\frac{d}{dt} \sigma_{-}(t) = i\Omega_{\text{int}} \sigma_{-} e^{-i\omega_{a} t} - \frac{\gamma_{j,2}}{2} (2n_{j}^{c} + 1) \sigma_{-}(t) + F_{C_{-}}(t)
\]  \hspace{1cm} (2.29)

where \( \gamma_{c} \equiv \gamma_{j} \left( 2n_{c}^{c} + 1 \right) \). The inversion satisfies an equation of motion determined by

\[
\left[ H(t), \sigma_{-}(t) \right] = \hbar \sum_{j} \left\{ 2\sqrt{\gamma_{j,2}} \left( -C_{j} \sigma_{-} e^{-i(\omega_{j} - \omega_{a})t} + \sigma_{-} C_{j}^{\dagger} e^{i(\omega_{j} - \omega_{a})t} \right) \right\}
\]  \hspace{1cm} (2.30)

\[
\left[ H(t + \tau), \left[ H(t), \sigma_{-}(t) \right] \right] =
\]

\[
2\hbar^{2} \sum_{j,k} \left\{ \sqrt{\gamma_{j,2} y_{k,2}} \left[ C_{j}(t + \tau) \sigma_{-}(t + \tau) e^{-i(\omega_{j} - \omega_{a})\tau} + \sigma_{-}(t + \tau) C_{j}^{\dagger}(t + \tau) e^{i(\omega_{j} - \omega_{a})\tau} \right] - C_{k} \sigma_{-} e^{-i(\omega_{k} - \omega_{a})\tau} + \sigma_{-} C_{k}^{\dagger} e^{i(\omega_{k} - \omega_{a})\tau} \right\}
\]

\[
= 2\hbar^{2} \sum_{j,k} \left\{ \sqrt{\gamma_{j,2} y_{k,2}} \left[ e^{-i(\omega_{j} - \omega_{a})\tau} e^{i(\omega_{k} - \omega_{a})\tau} \left[ C_{j}(t + \tau) \sigma_{-}(t + \tau), C_{j}(t + \tau) \right] \right] - e^{-i(\omega_{k} - \omega_{a})\tau} e^{i(\omega_{j} - \omega_{a})\tau} \left[ \sigma_{-}(t + \tau) C_{j}^{\dagger}(t + \tau), C_{k}(t) \sigma_{-}(t) \right] \right\}
\]
\[ Tr_R \left[ H(t+\tau), [H(t), \sigma_z(t)] \right] = 2\hbar^2 \delta(\tau) \cdot \sum_j \left\{ e^{-i(\alpha_j-\epsilon_0)t} \gamma_{j,2} \left[ (n_j^c + 1)\sigma_+(t)\sigma_-(t) - n_j^c \sigma_-(t)\sigma_+(t) \right] \right\} \]
\[ = 2\hbar^2 \delta(\tau) \cdot \sum_j \left\{ \gamma_{j,2} \left[ (n_j^c + 1)\left( \frac{1+\sigma_+}{2} \right) - n_j^c \left( \frac{1-\sigma_+}{2} \right) \right] \right\} \]
\[ = 2\hbar^2 \delta(\tau) \cdot \sum_j \left\{ \gamma_{j,2} \left[ (2n_j^c + 1)\sigma_z + 1 \right] \right\} \]

(2.31)

\[ \langle D_z(t) \rangle = -\hbar^2 \int_0^\infty d\tau Tr_R \left[ V(t+\tau), [V(t), \sigma_z(t)] \right] \]
\[ = -\sum_j \left\{ \gamma_{j,2} \left[ (2n_j^c + 1)\sigma_z + 1 \right] \right\} \]
\[ \rightarrow -\gamma_2 \left[ (2n_c^c + 1)\sigma_z + 1 \right] \]

(2.32)

\[ \frac{d}{dt} \sigma_z = -\gamma_2 \left[ (2n_c^c + 1)\sigma_z + 1 \right] \]
\[ + 2i\Omega_{\text{int}} \left[ \sigma_-(t)a^\dagger(t)e^{-i\omega_0 t} - \sigma_+(t)a(t)e^{i\omega_0 t} \right] + F_{C,z}(t) \]
\[ = 2i\Omega_{\text{int}} \left[ \sigma_-(t)a^\dagger(t)e^{-i\omega_0 t} - \sigma_+(t)a(t)e^{i\omega_0 t} \right] \]
\[ - \gamma_c \sigma_z - \gamma_2 + F_{C,z}(t) + \Lambda_p |1\rangle \langle 1| \]

(2.33)

Where \( |1\rangle \langle 1| \) is the population of the lower level and is given by the equation:

\[ |1\rangle \langle 1| = \frac{1}{2} \left[ 1 - \sigma_z(t) \right] \]

Thus the equation of the inversion will be:

\[ \frac{d}{dt} \sigma_z = -\gamma_p \sigma_z + 2i\Omega_{\text{int}} \left[ \sigma_-(t)a^\dagger(t)e^{-i\omega_0 t} - \sigma_+(t)a(t)e^{i\omega_0 t} \right] - \gamma_2 + \frac{\Lambda_p}{2} + F_{C,z}(t) \]

Where \( \gamma_p = \gamma_c + \frac{\Lambda_p}{2} \)
**Diffusion Terms for the TLS**

In calculating the diffusion terms for the TLS we use a different approach than the one for the NP where we used the double commutator. This is because in the case of the NP the double commutator will include terms involving the commutator of the NP operators $a, a^\dagger$ which is just unity, but in the case of the TLS the commutator of $\sigma_+, \sigma_-$ is not a c-number anymore but proportional to the operator $\sigma_z$, which introduces calculational complexities in the method previously employed. Instead we use the so called Einstein relation, which is given by the following form:

$$2\langle D_{\mu}(t) \rangle = -\langle D_+(t)\mu(t) \rangle - \langle \nu(t)D_+(t) \rangle + \frac{d}{dt} \langle \nu(t)\mu(t) \rangle$$  \hspace{1cm} (2.34)

Where $\langle D_{\mu}(t) \rangle$ is the diffusion term composed from the byproduct of the noise terms of the operators "$\mu, \nu$" and $D_+(t), D_-(t)$ are the drift terms of the corresponding operators.

$$2\langle D_+(t) \rangle = -\langle D_+(t)\sigma_-(t) \rangle - \langle \sigma_+(t)D_-(t) \rangle + \frac{d}{dt} \langle \sigma_+(t)\sigma_-(t) \rangle$$  \hspace{1cm} (2.35)

Where

$$\langle D_+(t)\sigma_-(t) \rangle = -\langle \frac{\gamma}{2} \sigma_+(t)\sigma_-(t) \rangle - i\Omega \langle \sigma_z(t)A(t)\sigma_z(t) \rangle$$  \hspace{1cm} (2.36)

$$\langle \sigma_+(t)D_-(t) \rangle = -\langle \frac{\gamma}{2} \sigma_+(t)\sigma_-(t) \rangle + i\Omega \langle \sigma_z(t)\sigma_z(t)A(t) \rangle$$

$$\frac{d}{dt} \langle \sigma_+(t)\sigma_-(t) \rangle = \frac{d}{dt} \langle |2\rangle \langle 2| \rangle = \frac{d}{dt} \left\langle \frac{1}{2} (1+\sigma_z) \right\rangle = \frac{1}{2} \langle \dot{\sigma}_z \rangle =$$

$$\frac{1}{2} \left\langle -\gamma_0 \sigma_z(t) + 2i\Omega \left[ \sigma_-(t)A(t) - \sigma_+(t)A(t) \right] - \gamma_1 \frac{\Lambda_0}{2} \right\rangle$$  \hspace{1cm} (2.37)

Finally the diffusion term is found to be:
\[ \langle D_{-}(t) \rangle = \frac{1}{2} \gamma_2 \langle \hat{n}^c + 1 \rangle + \frac{\Lambda_p}{8} [1 - \langle \sigma_z(t) \rangle] \]  

(2.38)

In calculating the above term we used the following relations:

\[ \sigma_z(t)\sigma_z(t) = U^\dagger(t)|1\rangle\langle1| + |2\rangle\langle2| U(t) = 1 \]

\[ \sigma_z(t)\sigma_z(t) = U^\dagger(t)|2\rangle\langle2| U(t) = \frac{1}{2} [1 - \sigma_z(t)] \]  

(2.39)

Where \( U \) is the unitary time evolution operator. Similarly one can find the other two diffusion terms to be:

\[ \langle D_{-}(t) \rangle = \frac{1}{2} \gamma_2 \langle \hat{n}^c + 1 \rangle + \frac{\Lambda_p}{8} [\langle \sigma_z(t) \rangle - 1] \]  

(2.40)

Eq's. (2.38) and (2.40) describe the decay of the dipole moment of the TLS due its coupling to the larger environment. This includes both damping and dephasing. It can be easily shown that expectation values \( \langle D_{-}(t) \rangle \) and \( \langle D_{+}(t) \rangle \) are identically zero just like the one for the NP \( \langle D_{n=2}(t) \rangle \).

The diffusion of the inversion is calculated from the following:

\[ 2\langle D_{n=2}(t) \rangle = -\langle \sigma_z(t)\sigma_z(t) \rangle - \langle \sigma_z(t)D_z(t) \rangle + \frac{d}{dt} \langle \sigma_z(t)\sigma_z(t) \rangle \]

\[ \langle D_z(t)\sigma_z(t) \rangle = -\langle \gamma_p \rangle \langle \sigma_z(t)\sigma_z(t) \rangle + 2i\Omega \langle \sigma_z(t)\sigma_z(t) A(t) \rangle^\dagger - \sigma_z(t)\sigma_z(t) A(t) \rangle + \langle \Lambda_p - \gamma_z \rangle \langle \sigma_z(t) \rangle \]

\[ \langle \sigma_z(t)D_{-}(t) \rangle = -\langle \gamma_p \rangle \langle \sigma_z(t)\sigma_z(t) \rangle + 2i\Omega \langle \sigma_z(t)\sigma_z(t) A(t) \rangle^\dagger - \sigma_z(t)\sigma_z(t) A(t) \rangle + \langle \Lambda_p - \gamma_z \rangle \langle \sigma_z(t) \rangle \]

\[ \frac{d}{dt} \langle \sigma_z(t)\sigma_z(t) \rangle = \frac{d}{dt} \langle |1\rangle\langle1| + |2\rangle\langle2| \rangle = \frac{d}{dt} (1) = 0 \]

\[ \Rightarrow \langle D_{n=2}(t) \rangle = \gamma_2 + (\gamma_2 - \frac{\Lambda_p}{2}) \langle \sigma_z(t) \rangle \]  

(2.41)
So in summary the various values of the diffusion terms will be as follow:

\[
\begin{align*}
\langle F_a(t)F_a(t') \rangle &= \frac{\bar{n}^2}{2} \delta(t-t') \\
\langle F_a(t)F_a^\dagger(t') \rangle &= \left(\frac{\bar{n}^2 + 1}{2}\right) \delta(t-t') \\
\langle F_\sigma(t)F_\sigma(t') \rangle &= \left(\frac{1}{2} \gamma_2 \bar{n}^C + \frac{\Lambda_p}{8} [1 - \langle \sigma_2(t) \rangle] \right) \delta(t-t') \\
\langle F_\sigma(t)F_\sigma^\dagger(t') \rangle &= \left(\frac{1}{2} \gamma_2 (\bar{n}^C + 1) + \frac{\Lambda_p}{8} [\langle \sigma_2(t) \rangle - 1] \right) \delta(t-t') \\
\langle F_z(t)F_z(t') \rangle &= \left(\gamma_p + \gamma_2 - \frac{\Lambda_p}{2} \langle \sigma_2(t) \rangle \right) \delta(t-t')
\end{align*}
\]

(2.42)

From the above we equations of motion will take the following form:

\[
\begin{align*}
\frac{d}{dt} a &= -\frac{\gamma}{2} \hat{a}(t) - i \Omega_{\text{int}} e^{-i\omega_0 t} \hat{\sigma}_- (t) + F_a (t) \\
\frac{d}{dt} \hat{\sigma}_- (t) &= -\frac{\gamma_2}{2} \hat{\sigma}_- (t) + i \Omega_{\text{int}} \hat{\sigma}_2 (t) \hat{a}(t) e^{-i\omega_0 t} + F_{\sigma^-} (t) \\
\frac{d}{dt} \hat{\sigma}_2 (t) &= -\gamma_p \hat{\sigma}_2 + 2 i \Omega_{\text{int}} \left[ \hat{\sigma}_- (t) \hat{a}^\dagger (t) e^{-i\omega_0 t} - \hat{\sigma}_+ (t) \hat{a} (t) e^{i\omega_0 t} \right] - \gamma_2 + \frac{\Lambda_p}{2} + F_{\sigma^+} (t)
\end{align*}
\]

(2.43)

In the next chapter we solve for the steady state solutions, the spectral width and for the intensity and squeezing spectrum.
CHAPTER III

SOLUTIONS OF THE EQUATIONS OF MOTION

Steady State

We change the slowly varying operators so they are all evaluated relative to the same (circular) frequency \( \nu \), undefined for the moment. That is, we define:

\[
\alpha_{H}(\nu) = \alpha(t)e^{-i\omega t} = A(t)e^{-i\nu t}, \quad \sigma_{H,\nu} = \sigma_{-}e^{-i\omega t} = \Sigma_{-}e^{-i\nu t},
\]

This leads to:

\[
\frac{d}{dt} \alpha = -\left[ \frac{\gamma}{2} + i(\omega - \nu) \right] A(t) - i\Omega_{\text{int}} \Sigma_{-} + F_{\alpha}(t)
\]  

where \( F_{\alpha}(t) = F_{a}(t)e^{i(\nu - m_{0})t} \) and:

\[
\frac{d}{dt} \Sigma_{-}(t) = -\left[ \frac{\gamma_{c}}{2} + i(\nu - \omega_{2}) \right] \Sigma_{-}(t) + i\Omega_{\text{int}} \Sigma_{z} A + e^{i(\nu - m_{0})t} F_{C,-}(t)
\]  

\[
\frac{d}{dt} \Sigma_{z} = -\gamma_{p} \Sigma_{z} + 2i\Omega_{\text{int}} \left[ \Sigma_{-} A^{\dagger} - \Sigma_{+} A \right]
\]

\[
-\gamma_{2} + F_{C,z}(t) + \frac{\Lambda_{p}}{2}
\]

Define \( G = \frac{2\Omega_{\text{int}}^{2}}{\gamma_{c}} \) and \( N = A^{\dagger}A = A A^{\dagger} - 1 \) and of course \( AA^{\dagger} + A^{\dagger}A = 2N + 1 \) where \( N \) is the polariton number. If we make the semi-classical approximation of factorizing expectation values, where we assume that we are working with coherent states we have:

\[
\frac{d}{dt} \langle A \rangle = -\left[ \frac{\gamma}{2} + i(\omega_{0} - \nu) \right]\langle A(t) \rangle + G \langle \Sigma_{z} \rangle \langle A \rangle
\]  

27

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\[ \frac{d}{dt} \langle \Sigma \rangle = -\gamma_p \langle \Sigma \rangle - 4G \langle \Sigma \rangle \langle N \rangle + \left[ -\gamma_2 + \frac{\Lambda_p}{2} \right] \] (3.5)

In the steady state, this has the solution:

\[ \langle \Sigma \rangle = \frac{\left[ \frac{\Lambda_p}{2} - \gamma_2 \right]}{\gamma_p + 4G \langle A' \rangle \langle A \rangle} \] (3.6)

\[ \left[ \frac{\gamma}{2} + i(\omega_b - \nu) \right] \langle A \rangle = +G \langle \Sigma \rangle \langle A \rangle \] (3.7)

This equation seems peculiar, since the LHS multiplies \( A \) by a complex number and the RHS multiplies it by a real number. We will take \( \nu = \omega_b \) in the following. Eq. (3.7) has the trivial solution \( \langle A \rangle = 0 \) as well as a nontrivial solution given by:

\[ \langle N \rangle = \langle A' \rangle \langle A \rangle = \left\{ \frac{1}{2\gamma} \left[ \frac{\Lambda_p}{2} - \gamma_2 \right] - \frac{\gamma_p}{4G} \right\} \] (3.8)

The right side must be positive, requiring

\[ \Lambda_p > \frac{2 \gamma \gamma_c + 4G \gamma_2}{2G - \gamma} \] (3.9)

Which is the threshold condition for polariton “lasing” in the parameter regime of interest.

The semi-classical solutions can be written

\[ A_0 = \sqrt{\left\{ \frac{1}{2\gamma} \left[ \frac{\Lambda_p}{2} - \gamma_2 \right] - \frac{\gamma_p}{4G} \right\}} \] (3.10)

\[ \Sigma_{z0} = \frac{\left[ \frac{\Lambda_p}{2} - \gamma_2 \right]}{\gamma_p + 4GN_0} = \frac{\gamma}{2G} \] (3.11)
\[ \Sigma_{-0} = i \frac{2\Omega_{\text{int}}}{\gamma_c} \Sigma_{z0} A_0 \]  

(3.12)

Where \( N_0 = A_0^* A_0 \). These equations represent the steady state average fields well above threshold.

We can easily show that requiring the threshold value of \( \Sigma_{z0} \) to be less than one implies that \( \Omega^2 > \frac{\gamma_c \gamma}{4} \).

**Intensity Spectrum**

In determining the intensity and squeezing spectrum we will take two approaches: in the first we follow the usual laser model in which the phase decay of the TLS coherence is large: \( \gamma_2 \gg \gamma \). In the second approach, corresponding more closely to conventional materials and technology, we assume the opposite condition \( \gamma \gg \gamma_2 \).

**First case:** When \( \gamma_2 \gg \gamma \), both \( \Sigma_\pm \) and \( \Sigma_z \) decay rapidly compared to the polariton field and can be assumed to follow this field on time scales long compared to \( \gamma_2^{-1} \) and we can make an adiabatic approximation in Eq.’s (3.2) and (3.3), setting \( \dot{\Sigma}_\pm(t) = \dot{\Sigma}_z(t) = 0 \):

\[
\Sigma_- = \frac{2i\Omega}{\gamma_c} \Sigma_z A + \frac{2}{\gamma_c} F_{c+}(t), \quad \Sigma_+ = -\frac{2i\Omega}{\gamma_c} \Sigma_z A^\dagger + \frac{2}{\gamma_c} F_{c+}(t) \tag{3.13}
\]

\[
\Sigma_z = \frac{2i\Omega}{\gamma_p} (\Sigma_- A^\dagger - \Sigma_+ A) + \frac{\lambda}{\gamma_p} + F_z(t), \quad \text{where} \quad \lambda = \frac{\Lambda_p}{2} - \gamma_2 \tag{3.14}
\]

From (3.13) we find:
\[ \Sigma_z (1 + \frac{4\Omega^2}{\gamma_c \gamma_p} A^\dagger A) = \frac{2i\Omega}{\gamma_p} (\Sigma - A^\dagger) - \frac{i4\Omega}{\gamma_c \gamma_p} AF_{e+}^\dagger(t) + \frac{\lambda + F_z^\dagger(t)}{\gamma_p}, \text{ where } M = 1 + \frac{4\Omega^2}{\gamma_c \gamma_p} A^\dagger A \]

\[ \Rightarrow \Sigma_z = \frac{2i\Omega}{\gamma_p M} (\Sigma - A^\dagger) - \frac{i4\Omega}{\gamma_c \gamma_p M} AF_{e+}^\dagger(t) + \frac{\lambda + F_z^\dagger(t)}{\gamma_p M}, \text{ where } M = 1 + \frac{4\Omega^2}{\gamma_c \gamma_p} A^\dagger A \]

Thus the equation of the polariton amplitude of the NP becomes:

\[ \frac{dA}{dt} = -\frac{\gamma}{2} A + 2\Omega^2 (\lambda + F_z^\dagger(t)) A + \frac{2i\Omega F_{e+}(t)}{\gamma_p \gamma_c} A A - \frac{2i\Omega F_{e-}(t) + F_a(t)}{K\gamma_c} \]

This is a non-linear equation familiar from standard laser theory. To calculate the intensity spectrum we assume that we are far above threshold, that we can neglect the fluctuations in the amplitude inasmuch as this quantity is constrained to fluctuate about its steady state value, while phase can change freely. Pictorially, the tip of the complex field diffuses around a circle in the complex plane while its amplitude remains relatively constant Fig. (1.2). We therefore consider only the time dependence in the phase. The plasmon field operator \( A(t) \) can be written classically as follow:

\[ A(t) = A_0 e^{-i\theta} \] (3.17)

Where \( A_0 \) is the steady state value of \( A \) given in Eq. (3.10). The intensity is then given by the following expression:
\[ \langle A(t)\dagger A(0) \rangle = N_0 e^{-i[B(t)\cdot \phi(0)]} = N_0 e^{-\frac{i}{2} B(t)^2} \]  

(3.18)

Where the last expression is derived in the appendix, and we took the arbitrary phase at zero time to be zero. The phase in the exponential can be computed from the phase diffusion given by the following:

\[ 2i\dot{\phi}(t') = \frac{1}{A(t)} A^\dagger(t) - \frac{1}{A(t)} A(t) \]  

(3.20)

This leads to

\[ 2i\dot{\phi}(t') = \frac{2i\Omega}{K\gamma_c \sqrt{N_0}} \left( F_{\text{c},-}(t) e^{-i\phi} - F_{\text{c},+}(t) e^{i\phi} \right) + \frac{1}{\sqrt{N_0}} \left( \frac{1}{N_0} \left( F_{\text{a},+}(t) e^{-i\phi} - F_{\text{a},-}(t) e^{i\phi} \right) \right) \]

Putting it all in Eq. (3.19) and by using the expectation values of the byproduct of the noise terms (2.43), we reach the following relation for the phase diffusion coefficient:

\[ \langle D(\phi) \rangle = \frac{1}{4N_0} \left[ \gamma (\bar{n} + \frac{1}{2}) \left( \frac{\Omega^2}{\gamma_c} \right) \right] \]

(3.21)

The polariton operator \( A(t) \) then can be written as follow:

\[ \langle A(t)\dagger A(0) \rangle = N_0 e^{-(\phi(t))} = N_0 e^{-\frac{i}{2} B(t)^2} \]

(3.22)

Where \[ \Gamma = \frac{1}{4N_0} \left[ \gamma (\bar{n} + \frac{1}{2}) + \frac{\Omega^2}{\gamma_c} \right] \]

(3.23)

We see that the spectral width of the intensity spectrum \( \Gamma \) for the NP is inversely proportional to the polariton number \( N_0 \) as known from standard laser theory, and
contains a term inversely proportional to $\gamma_c$, Eq. (3.23). This latter term represents the decrease of NP lifetime due its coupling to a decaying TLS. This means that as the decay rate from the excited states of the TLS increases the spectral width of the NP becomes narrower, which is counter-intuitive. To understand this behavior we go back to the equations of motion (2.43). The width of the spectrum of the NP arises from the spontaneous decay of the NP. This decay can happen in many different ways. The NP polaritons are coupled to the NP reservoir and can spontaneously decay into it, or the decay may be due to the coupling to the TLS: the polaritons can send a virtual photon to the TLS which in turn decays to the surrounding reservoir, or the TLS could return the virtual photon back to the NP which in turn decays back into its reservoir and so on. These processes lead to the broadening of the NP spectrum. This process is represented pictorially below:

$$\Gamma = \gamma \quad + \quad \gamma_c \quad + \quad \gamma \quad + \quad \gamma_c$$

NP \quad NP \quad TLS \quad NP \quad TLS \quad NP \quad TLS

Figure 3.1. A pictorial representation showing the process of spontaneous emission of the NP.

Looking into the equation of motions, we find that the NP operator $A(t)$ is proportional to the dipole moment of the TLS $\Sigma$:

$$\frac{d}{dt} A = -\frac{\gamma}{2} A(t) - i \Omega_{\text{int}} \Sigma - + F_A(t)$$
But in the adiabatic approximation $\Sigma$, itself is proportional to the inverse of $\gamma_c$ (larger $\gamma_c$ implies more population in the ground state and therefore decreased transition dipole moment), so when $\gamma_c$ increases the dipole moment of the TLS $\Sigma$ decreases, which in turn reduces the coupling between the TLS and the NP which in turn decreases the line width of the NP. This feature is peculiar to our system, as it is not present in regular lasers.

**Second Case:** When $\gamma >> \gamma_2$ we cannot eliminate the time derivatives of the $\Sigma$'s, but we can eliminate the time derivative of the plasmon field amplitude $A(t)$. This results in a set of nonlinear equations which can be solved numerically but not analytically. To increase the insight into the properties of the solution, we suppose below that the population inversion of the TLS $\Sigma_z$ is constant. This can be justified by requiring a fast pumping for the TLS so the population inversion can be fixed to a certain value. Fixing the inversion allows us to solve for the spectrum exactly using the same phase diffusion technique as above.

We start by eliminating the time derivative of the plasmon field $A(t)$:

$$A(t) = \frac{2}{\gamma} (-i \Omega \Sigma_+ + F_A(t))$$  \hspace{1cm} (3.24)

By setting $\Sigma_z$ fixed, the equation of the decay $\Sigma$ is given by the following:

$$\frac{d}{dt} \Sigma_-(t) = -\frac{\gamma_c}{2} \Sigma_-(t) + \frac{2}{\gamma} i \Omega \Sigma_-(t) + F_A(t) + F_{C,-}(t)$$

$$= \left[ -\frac{\gamma_c}{2} + \frac{2}{\gamma} \Omega^2 \Sigma_+ \right] \Sigma_-(t) + \frac{2i \Omega}{\gamma} F_A(t) + F_{C,-}(t)$$  \hspace{1cm} (3.25)

Eq. (3.25) can be solved to give the following:
\[
\Sigma_\pm(t) = e^{-\pi t} \Sigma_\pm(0) + \int_0^t e^{-i(t-t')} \left[ \frac{2i \Omega}{\gamma} F_A(t') + F_{C\pm}(t') \right] dt' 
\]

(3.26)

Where \( r = \left[ \frac{T_c}{2} - \frac{2 \Omega^2 \Sigma_\pm}{\gamma} \right] \). But from Eq. (3.11) \( \Sigma_{\pm 0} = \frac{\gamma}{2G} \) so \( r \) is identically zero.

We substitute Eq. (3.26) into the full time equation of motion of the plasmon amplitude Eq. (3.1):

\[
\frac{d}{dt} A = -\frac{\gamma}{2} A(t) - i\Omega \left\{ \Sigma_\pm(0) + \int_0^t \frac{2i \Omega}{\gamma} F_A(t') + F_{C\pm}(t') \right\} + F_A(t) 
\]

(3.27)

Now by using Eq. (3.20) we get the following expression for \( \phi(t') \) Eq. (3.28):

\[
2i\phi(t') = \frac{1}{A^*} A^\dagger A - \frac{1}{A} A = \\
\frac{i \Omega}{\sqrt{N_0}} \left( \Sigma_\pm(0) e^{-i\phi} + \Sigma_\pm(0) e^{i\phi} \right) + \frac{1}{\sqrt{N_0}} \left( F_A^\dagger(t') e^{-i\phi} - F_A(t') e^{i\phi} \right) + \\
\frac{2 \Omega^2 \Sigma_\pm(0)}{\gamma \sqrt{N_0}} \int_0^t \left( F_A^\dagger(t') e^{-i\phi} - F_A(t') e^{i\phi} \right) dt' + \frac{i \Omega}{\sqrt{N_0}} \int_0^t \left( F_{C\pm}^\dagger(t') e^{-i\phi} - F_{C\pm}(t') e^{i\phi} \right) dt' 
\]

By taking the expectation value of the double integral in Eq. (3.19) we realize that the self product of the first term, and its product with all other terms contribute nothing. This is also true for the self product of the second and fourth terms. This we explain in more details below:

The product of the first term \( \frac{i \Omega}{\sqrt{N_0}} e^{\pi t} \left( \Sigma_\pm(0) e^{-i\phi} + \Sigma_\pm(0) e^{i\phi} \right) \) with any other term will give zero because the expectation value of single noise term is zero, Markovian noise, \( \langle F_A(t) \rangle = 0 \). The self product of this term is also zero because when doing the double integral we will have term like \( \Delta t^2/\Delta t \) and when \( \Delta t \) goes to zero the whole term vanishes.
The integral is zero for the self product of the 3\textsuperscript{rd} term, and the 4\textsuperscript{th} term for the same reason, so we only show the calculation for one of them:

\[
\int_{t}^{t+\Delta t} dt_{1} \int_{t}^{t+\Delta t} dt_{2} \int_{0}^{t_{1}} \left(F_{a}^{\dagger}(t')e^{-i\phi} - F_{a}(t'e^{i\phi})\right) dt' \int_{0}^{t_{2}} \left(F_{a}^{\dagger}(t'')e^{-i\phi} - F_{a}(t''e^{i\phi})\right) dt''
\]

\[= -\gamma \left(2\bar{n}^{4} + 1\right) \int_{t}^{t+\Delta t} dt_{1} \int_{t}^{t+\Delta t} dt_{2} \int_{0}^{m} dt'
\]

Where \(m\) is the minimum of \((t_{1}, t_{2})\) and we used the fact that:

\[
\langle F_{a}^{\dagger}(t')F_{a}(t'') \rangle = \bar{n}^{4} \frac{\gamma}{2} \delta(t' - t'')
\]

\[
\langle F_{a}(t')F_{a}^{\dagger}(t'') \rangle = \left(\bar{n}^{4} + 1\right) \frac{\gamma}{2} \delta(t' - t'')\]

Now \(\int_{0}^{m} dt' = m\)

\[
\Rightarrow \int_{t}^{t+\Delta t} dt_{1} \int_{t}^{t+\Delta t} dt_{2} m = \int_{t}^{t+\Delta t} dt_{1} \left\{ \int_{t}^{t+\Delta t} dt_{2} t_{2} + \int_{t}^{t+\Delta t} dt_{1} \int_{t}^{t+\Delta t} dt_{1} \right\}
\]

\[= \int_{t}^{t+\Delta t} dt_{1} \left\{ \frac{t_{2}^{2} - t^{2}}{2} + t_{1}(t + \Delta t - t_{1}) \right\}
\]

The integral can evaluated as follow:

\[
\frac{(t + \Delta t)^{3} - t^{3}}{6} - \frac{t^{2}}{2}\Delta t + (t + \Delta t) \left(\frac{(t + \Delta t)^{2} - t^{2}}{2}\right) - \frac{(t + \Delta t)^{3} - t^{3}}{3} = 0
\]

So we get nothing form the \(\langle D(\phi) \rangle_{33}\) and \(\langle D(\phi) \rangle_{44}\)

Now we calculate the integral of the product of the 2\textsuperscript{nd} and 3\textsuperscript{rd} terms:

\[
\frac{2\Omega^{2} \Sigma_{z}(0)^{t+\Delta t}}{\gamma N_{0}\Delta t} \int_{t}^{t+\Delta t} dt_{1} \int_{t}^{t+\Delta t} dt_{2} \int_{0}^{t_{1}} \left(F_{a}^{\dagger}(t')e^{-i\phi} - F_{a}(t'e^{i\phi})\right) dt' \left(F_{a}^{\dagger}(t_{2})e^{-i\phi} - F_{a}(t_{2}e^{i\phi})\right)
\]

\[= -\gamma \left(2\bar{n}^{4} + 1\right) \frac{2\Omega^{2} \Sigma_{z}(0)^{t+\Delta t}}{\gamma N_{0}\Delta t} \int_{t}^{t+\Delta t} dt_{1} \int_{t}^{t+\Delta t} dt_{2} \int_{0}^{m} \delta(t' - t_{2}) dt'
\]

The integral is then:

\[35\]

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\[
\int_{t}^{t+\Delta t} \int_{t}^{t+\Delta t} dt_1 \, dt_2 = \Delta t^2
\]

So \( \langle D(\phi) \rangle_{23} \) also goes to zero.

In the same way it is easy to show that phase diffusion of the self product of the 2\textsuperscript{nd} term is given by:

\[
\langle D(\phi) \rangle_{22} = \frac{(2\bar{n}_a + 1)}{8N_0} \gamma
\]

which is the only term left. The spectral width will wound up to be:

\[
\Gamma = \frac{(2\bar{n}_a + 1)}{8N_0} \gamma \tag{3.29}
\]

By comparing to (3.23) we find that the first terms in both cases are the same, but while in case one there are contribution from noise term coming from the coupling to the TLS, in this case this other term is missing because the polariton lifetime is so much shorter than the TLS lifetime.

**The Radiated Light**

The total system radiated electric field is given by the following equation:

\[
E_{\text{rad}}(r,t) = E_{\text{vac}}(r,t) + E_{NP}(r,t) + E_{TLS}(r,t)
\]

Where \( E_{\text{vac}} \) is the vacuum electric field, \( E_{NP} \) and \( E_{TLS} \) are the electric fields of the nanoparticle and the two-level system:

\[
E_{NP}^{+}(r,t) = -\frac{\omega_0^2}{4\pi\varepsilon_0\varepsilon_r c^2 r} \left( \mu_x \times \frac{\vec{r}}{r} \right) \frac{A(t - \frac{r}{c})}{r}
\]

\[
E_{TLS}^{+}(r,t) = -\frac{\omega_0^2}{4\pi\varepsilon_0\varepsilon_r c^2 r} \left( \mu_x \times \frac{\vec{r}}{r} \right) \frac{\Sigma_{-}(t - \frac{r}{c})}{r}
\]

\[\text{(3.31)}\]
Where $\varepsilon_2$ is the permittivity of the medium in which the system is embedded, and $c$ is the speed of light.

We write back the operators in term of the fast varying forms:

$$A(t) = a(t) e^{-i\omega_b t} = A_0 e^{-i\omega \cdot r}, \quad \Sigma_-(t) = \sigma_-(t) e^{-i\omega_b t} = iL A_0 e^{-i\omega \cdot r}$$

and so for their complex conjugates, and

$$L = \frac{2\Omega \lambda}{\gamma c N_p + 8\Omega^2 N_0} = \frac{2\gamma}{\Omega}$$

We first calculate the first order correlation function:

$$G^1(r,t) = \langle E_{\text{tot}}^-(r,t)E_{\text{tot}}^+(r,t) \rangle$$

To do so we define the following:

$$I_{NP} = \frac{\omega_0^2}{4\pi \varepsilon_0 \varepsilon_2 c^2 r} \left( \mu_0 \varepsilon_0 \varepsilon_2 c^2 r \right)$$

$$I_{TLS} = \frac{\omega_0^2}{4\pi \varepsilon_0 \varepsilon_2 c^2 r} \left( \mu_0 \varepsilon_0 \varepsilon_2 c^2 r \right)$$

Where $r$ is the distance of the detector from the system and is taken to be on the order of few nano meters.

This will give the following values for the various correlations (3.32):

$$\langle E_{\text{TLS}}^- (r,t) E_{\text{NP}}^+ (r,t) \rangle = -i \sqrt{I_{TLS} I_{NP}} A_0^2 e^{i\omega_b (t-t') - \Gamma (t+t')}$$

$$\langle E_{\text{TLS}}^- (r,t) E_{\text{TLS}}^+ (r,t) \rangle = i \sqrt{I_{TLS} I_{NP}} A_0^2 e^{i\omega_b (t-t') - \Gamma (t+t')}$$

$$\langle E_{\text{TLS}}^- (r,t) E_{\text{TLS}}^+ (r,t) \rangle = I_{TLS} L^2 A_0^2 e^{i\omega_b (t-t') - \Gamma (t+t')}$$

$$\langle E_{\text{NP}}^- (r,t) E_{\text{NP}}^+ (r,t) \rangle = I_{NP} A_0^2 e^{i\omega_b (t-t') - \Gamma (t+t')}$$

The correlation function will take the form:

$$G^1(r,t) = A_0^2 (I_{NP} + I_{TLS} L^2) e^{i\omega_b (t-t') - \Gamma (t+t')}$$

(3.33)

The spectrum is given by:
\[ S_1(\omega, r, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 e^{i(\omega t - rt)} G^1(rt_1, rt_2) \]

\[ \Rightarrow S_1(\omega, r, t) = \frac{1}{2\pi} A_0^2 (I_{NP} + I_{TLS} L^2) \left[ \frac{1 + e^{-2i\tau} - 2e^{-2i\tau} \cos[(\omega - \omega_0)T]}{\left(\omega - \omega_0\right)^2 + \Gamma^2} \right] \]

Now for \( \Gamma < 0 \) and \( T \to \infty \) the spectrum will take the final form:

\[ \Rightarrow S_1(\omega, r, t) = \frac{A_0^2}{2\pi} (I_{NP} + I_{TLS} L^2) \left[ \frac{1}{\left(\omega - \omega_0\right)^2 + \Gamma^2} \right] \quad (3.34) \]

The second order coherence function can also be found to be:

\[ G^2(r, t) = \left\langle E_{tot}^-(r, t) E_{tot}^-(r, t') E_{tot}^+(r, t') E_{tot}^+(r, t) \right\rangle = [A_0^2 (I_{NP} + I_{TLS} L^2)]^2 e^{-2i(\tau + \tau')} \]

\[ \Rightarrow S_2(\omega, r) = \frac{A_0^4}{2\pi} (I_{NP} + I_{TLS} L^2)^2 \left[ \frac{1}{\left(\omega - \omega_0\right)^2 + 4\Gamma^2} \right] \quad (3.35) \]

Second order coherence can be used to find the statistical characters of intensity fluctuations. A wider spectrum means more fluctuation between intensities measured at different times. If \( G^2(0) \geq G^2(\tau) \) is satisfied the light is said to be bunched “classical”.

On the other hand if \( G^2(0) \leq G^2(\tau) \) light is said to be anti-bunched and will exhibit quantum effects.

**Squeezing Spectrum**

Any kind of light will have some noise in its amplitude or phase or both. No matter how careful one can be, you can never reduce the noise below a certain level, called the standard quantum limit (SQL). But it can be shown that in certain situations one can reduce the noise in one quadrature of the field at the expense of increasing it in the other...
quadrature. Quadratures can be represented as expectation values of the two Hermitian combinations of the Bose amplitudes:

\[ X = \frac{1}{2} (\hat{a}^\dagger + \hat{a}) \quad Y = \frac{i}{2} (\hat{a}^\dagger - \hat{a}) \]  \hspace{1cm} (3.36)

Squeezed light has a smaller value for the variance of one quadrature and an increased variance for the other. This must be the case as the uncertainty product for these quadratures is:

\[ (\Delta X)^2 \ (\Delta Y)^2 \geq 1 \]  \hspace{1cm} (3.37)

The noise of the light is defined by the equation:

\[ N = \langle (\chi + iY)^2 \rangle - \langle (\chi + iY) \rangle^2 \]  \hspace{1cm} (3.38)

where \( \chi = ax - kz + \phi \). This noise cannot go below certain value, defined as the SQL:

\[ N \geq 1. \]

For example in coherent light the quadrature variances equal each other and have the common value 1, the minimum value allowed by the uncertainty principle. The noise for the coherent light also takes its minimum value \( N = 1 \). These properties can be illustrated by Fig. (3.1) where the uncertainty principle restricts any measurement of the light field to lie somewhere inside the shaded circle:
Figure 3.2. The mean and the fluctuation of a coherent state field.

As we see from the figure above, the circular disk represents the uncertainty in phase and amplitude in the coherent state field. The circular shape comes from the fact that the variances of the quadratures are equal.

In squeezed coherent state, the circle in Fig. (3.1) will have an ellipsoidal shape, whose major and minor axes represent either the amplitude or phase variance:

Figure 3.3. An amplitude squeezed laser light.
As an example, the squeezing operator is defined by the following equation:

\[
\hat{S}(\zeta) = \exp\left(\frac{1}{2} \zeta \hat{a}^2 - \frac{1}{2} \zeta (\hat{a}^\dagger)^2\right)
\] (3.39)

To get squeezed laser light, first we let the squeezing operator operate on the vacuum state \(|0\rangle\) changing it to a squeezed "vacuum" state: \(\hat{S}(\zeta)|0\rangle = |\zeta\rangle\).

Squeezed coherent light results from applying the displacement coherent operator

\[
\hat{D}(\alpha) = \exp(\alpha \hat{a}^\dagger - \alpha^* \hat{a})
\]
to the squeezed vacuum. Squeezed vacuum light is typically produced by parametric down conversion (PDC) where an incident photon gives rise to two quantum-correlated photons in a non-linear crystal, while squeezed coherent light can today be produced from certain lasers.

Squeezing is a peculiarly quantum property of light and has a number of uses. It has been suggested that squeezed light can act as a component of a high quality detector of mechanical vibrations (for detecting gravitational waves) and for quantum communication involving the transfer of quantum-correlated but spatially separated photon pairs as in the EPR effect. All known sources of squeezed light result from highly macroscopic systems. The dipole nanolaser may produce squeezed light in a device that can fit onto a computer chip.

To understand the squeezing mechanism, we notice that the squeezing operator operates with the square of the Bose operators \((\hat{a}^2, \hat{a}^\dagger^2)\), which means that squeezing produces photons in pairs, entangled photon pairs.

But why would creating correlated photon pairs leads to reducing the quantum noise? The answer lies in the statistical distribution of the detection of photons. Let us
look, for fixed interval of time, at the distribution of photons detected for the two cases below:

i. When there is no squeezing: the detector will detect random numbers of photon in each interval: e.g. one in the first, three in the second, two in the third, etc. this distribution is called Poisson distribution.

ii. When there is squeezing: in this case, once the detector detects a photon, there is a very high probability that it will detect its correlated photon. This means that the randomness associated with the Poissonian distribution is diminished now, the Poisson distribution will have a diminished probability of detecting odd number of photons, and this will lead to smaller variance in the number of photons detected.

In our work, we will examine the spectrum, spatial distribution, line width, and squeezing spectrum of the light emitted from the nano-particle.

We write the slowly varying operators in terms of a constant term and fast varying term:

\[ A(t) = A_0 + \delta A(t) , \quad \Sigma_z(t) = \Sigma_{z0} + \delta \Sigma_z(t) , \quad \Sigma_- (t) = \Sigma_{-0} + \delta \Sigma_-(t) \quad (3.40) \]

Here the quantum fluctuations are assumed to have zero mean and to be small in magnitude. We now substitute (3.40) into Eq. (3.1-3).

\[
\begin{align*}
\frac{d}{dt} \delta A(t) & = -\frac{\gamma}{2}[A_0 + \delta A(t)] - i \Omega_{\text{int}} \left[ \Sigma_{z0} + \delta \Sigma_z(t) \right] + F_A \\
& = \left\{ -\frac{\gamma}{2} A_0 - i \Omega_{\text{int}} \Sigma_{z0} \right\} - \frac{\gamma}{2} \delta A(t) - i \Omega_{\text{int}} \delta \Sigma_z(t) + F_A \\
& = -\frac{\gamma}{2} \delta A(t) - i \Omega_{\text{int}} \delta \Sigma_z(t) + F_A
\end{align*}
\]

The term in braces on the second line above is zero.
Summarizing,

\[
\frac{d}{dt} \delta A(t) = -\frac{\gamma}{2} \delta A(t) - i \Omega_{\text{int}} \delta \Sigma_-(t) + F_t
\]  

(3.41)

\[
\frac{d}{dt} \delta \Sigma_-(t) = -\frac{\gamma c}{2} \left[ \Sigma_{-0} + \delta \Sigma_-(t) \right] + i \Omega_{\text{int}} \left[ \Sigma_{z0} + \delta \Sigma_+(t) \right] A_0 + \delta A(t) + e^{i(\nu \omega t)} \left[ F_{C,-}(t) \right]
\]

(3.42)

Finally,

\[
\frac{d}{dt} \delta \Sigma_+(t) = -\gamma_p \left[ \Sigma_{z0} + \delta \Sigma_+(t) \right] + 2i \Omega_{\text{int}} \left[ \left\{ \Sigma_{z0} A_0 - \Sigma_{-0} A_0 \right\} + \left\{ \Sigma_{z0} \delta \Sigma_+ + \delta \Sigma_+ \right\} A_0 \right] - \left[ \gamma_2 - \frac{\Lambda_p}{2} \right] + F_{C,+}(t)
\]  

(3.43)

We can form the quadrature operators:

\[
\delta X = \delta A + \delta A', \quad \delta Y = i \left( \delta A' - \delta A \right)
\]

(3.44)

so that
\[
\frac{d}{dt} X(t) = -\frac{\gamma}{2} X(t) + i \Omega_{\text{int}} \left[ \delta \Sigma_+ - \delta \Sigma_- \right] + F_A + F_A^\dagger
\]
\[
\frac{d}{dt} Y(t) = -\frac{\gamma}{2} Y(t) - \Omega_{\text{int}} \left[ \delta \Sigma_+ + \delta \Sigma_- \right] + i \left( F_A^\dagger - F_A \right)
\]

\[
\frac{d}{dt} \delta \Sigma_z(t) = 2i \Omega_{\text{int}} \left\{ \Sigma_{z0} \frac{1}{2} (\delta \Sigma_+ - i \delta \Sigma_-) + \delta \Sigma_- A_0 - \Sigma_{z0} \frac{1}{2} (\delta \Sigma_+ - i \delta \Sigma_-) - \delta \Sigma_+ A_0 \right\} + F_{C,z}(t) - \gamma_p \delta \Sigma_z(t)
\]

\[
\frac{d}{dt} \delta \Sigma_z(t) = -\gamma_p \delta \Sigma_z(t) + i \Omega_{\text{int}} (\Sigma_{z0} - \Sigma_{z0}) \delta \Sigma_+ + \Omega_{\text{int}} (\Sigma_{z0} + \Sigma_{z0}) \delta \Sigma_-
\]

\[
= -\gamma_p \delta \Sigma_z(t) + i \Omega_{\text{int}} \left( \Sigma_{z0} - \Sigma_{z0} \right) \delta \Sigma_+ + \Omega_{\text{int}} \left( \Sigma_{z0} + \Sigma_{z0} \right) \delta \Sigma_-
\]

We write this in matrix form:

\[
\begin{pmatrix}
\delta X(t) \\
\delta Y(t) \\
\delta \Sigma_+(t) \\
\delta \Sigma_-(t) \\
\delta \Sigma_z(t)
\end{pmatrix}
= J \begin{pmatrix}
\delta X(t) \\
\delta Y(t) \\
\delta \Sigma_+(t) \\
\delta \Sigma_-(t) \\
\delta \Sigma_z(t)
\end{pmatrix} + \begin{pmatrix}
F_A + F_A^\dagger \\
i \left( F_A^\dagger - F_A \right) \\
e^{-i(r-v_0)t} \left[ F_{C,z} \right] \\
e^{i(r-v_0)t} \left[ F_{C,z} \right] \\
F_{C,z}(t)
\end{pmatrix}
\]

where

\[
J = \begin{pmatrix}
-\frac{\gamma}{2} & 0 & i \Omega_{\text{int}} & -i \Omega_{\text{int}} & 0 \\
0 & -\frac{\gamma}{2} & -\Omega_{\text{int}} & -\Omega_{\text{int}} & 0 \\
-\frac{i \Omega_{\text{int}}}{2} \Sigma_{z0} & -\frac{i \Omega_{\text{int}}}{2} \Sigma_{z0} & \frac{\gamma_p}{2} & 0 & -i \Omega_{\text{int}} A_0^* \\
i \Omega_{\text{int}} \Sigma_{z0} & -i \Omega_{\text{int}} \Sigma_{z0} & 0 & \frac{\gamma_p}{2} & i \Omega_{\text{int}} A_0 \\
i \Omega_{\text{int}} \left( \Sigma_{z0} - \Sigma_{z0} \right) & \Omega_{\text{int}} \left( \Sigma_{z0} + \Sigma_{z0} \right) & -2i \Omega_{\text{int}} A_0 & 2i \Omega_{\text{int}} A_0^* & -\gamma_p
\end{pmatrix}
\]

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\[
\begin{align*}
\delta \Xi &= \begin{pmatrix}
\delta X(t) \\
\delta Y(t) \\
\delta \Sigma_+(t) \\
\delta \Sigma_-(t) \\
\delta \Sigma_z(t)
\end{pmatrix}, \quad F_\Xi = \begin{pmatrix}
F_\Delta(t) + F_\Delta^+(t) \\
i(F_\Delta^+(t) - F_\Delta(t)) \\
e^{-i(v_\omega)\tau}F_{C_+}^+(t) \\
e^{i(v_\omega)\tau}F_{C_-}(t) \\
F_{C_z}(t)
\end{pmatrix}
\text{so that } \frac{d}{dt} \delta \Xi = J \delta \Xi + F_\Xi.
\end{align*}
\]

\[
\delta \Xi(t) = \exp(Jt) \Xi(0) + \int_0^t dt' \exp\left[J(t-t')\right] F_\Xi(t')
\]

We require \(S(\omega) = \int dt \ e^{-ist} \langle \delta X(t), \delta X(0) \rangle \).

Now \(\delta X(t) = \left[\exp(Jt)\right]_t \Xi(0) + \int_0^t dt' \exp\left[J(t-t')\right]_t F_\Xi(t')\) where sums on repeated indices are understood.

**Solving the Differential Equations**

We solve the above set equations by taking their Fourier transform, and solve the coupled set of linear equations using MathCAD software.

We define our Fourier transformations with the following definitions:

\[
F(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{ist} f(t) dt \quad \text{and} \quad F^\dagger(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{ist} f(t)^\dagger dt \quad (3.48)
\]

But note that \(F(\omega)^\dagger\) is not the conjugate of \(F(\omega)\), it is the conjugate of \(F(-\omega)\).

\[
F^\dagger(\omega) = [F(-\omega)]^\dagger \quad (3.49)
\]

The Fourier transformed set of equations takes the following form Eq. (3.50):
By using MathCAD in solving the above equation, we get very long and complicated solutions. For case one in section 3.2 we can, instead of solving the above five equations directly, use the adiabatic approximation to eliminate $\Sigma_\pm(\omega)$, reducing the five equations to three: (3.51)

$$\left( -i\omega + \frac{2\Omega^2}{\gamma_c} \right) \delta X(\omega) - i\Omega \left( \delta \Sigma_+(\omega) - \delta \Sigma_-(\omega) \right) - \left( F_\Delta(\omega) + F_\Delta^*(\omega) \right) = 0$$

$$\left( -i\omega + \frac{2\Omega^2}{\gamma_c} \right) \delta Y(\omega) + \Omega \left( \delta \Sigma_+ + \delta \Sigma_- \right) - i \left( F_\Delta^*(\omega) - F(\omega)_A \right) = 0$$

$$\left( -i\omega + \gamma_p \right) \delta \Sigma_\pm(\omega) - i\Omega \left( \Sigma_\pm - \Sigma_0 \right) \delta X(\omega) - \Omega \left( \Sigma_\pm + \Sigma_0 \right) \delta (\omega) Y + 2i\Omega A_0 \left( \delta \Sigma_+ - \delta \Sigma_- \right) - F_\Sigma(\omega) = 0$$

The solutions of Eq.'s (3.50) and (3.51) are given by the following expressions Eq. (3.52):

$$\delta X(\omega) = \frac{1}{2} \left[ S_{\Delta\pm}(\omega) F_{\Delta\pm}(\omega) + S_{\Delta\pm}(\omega) F_{\Delta\pm}^*(\omega) + S_{\Delta\mp}(\omega) F_{\Delta\mp}(\omega) + S_{\Delta\mp}(\omega) F_{\Delta\mp}^*(\omega) \right] \frac{D_\pm(\omega)}{D_y(\omega)}$$

$$\delta Y(\omega) = \frac{S_{\Delta\pm}(\omega) [F_{\Delta\pm}^*(\omega) - F_{\Delta\pm}(\omega)] + S_{\Delta\mp}(\omega) [F_{\Delta\mp}(\omega) + F_{\Delta\mp}^*(\omega)]}{D_y(\omega)}$$

Where the various $S$'s coefficients are different for both cases and are given in the appendix.
The corresponding quadratures of the TLS can be easily found from the equations of motion:

\[
\begin{align*}
\delta X_{\pm}(\omega) &= \delta \Sigma_{\pm}(\omega) + \frac{i[F_a^+(\omega) - F_a(\omega)] + (i\omega - \frac{\gamma}{2})\delta Y(\omega)}{\Omega} \\
\delta Y_{\pm}(\omega) &= \delta \Sigma_{\pm}(\omega) - \frac{[F_a^+(\omega) + F_a(\omega)] - (i\omega - \frac{\gamma}{2})\delta X(\omega)}{\Omega}
\end{align*}
\]

(3.53)

The amplitude and phase squeezing of the total light emitted from the system are defined as follows:

Amplitude spectrum = \[\langle \delta X_{\text{out}}(\omega)^\dagger \delta X_{\text{out}}(\omega) \rangle = \langle \delta X_{\text{out}}(-\omega)\delta X_{\text{out}}(\omega) \rangle + 1 \] (3.54.a)

Phase Spectrum = \[\langle \delta Y_{\text{out}}(\omega)^\dagger \delta Y_{\text{out}}(\omega) \rangle = \langle \delta Y_{\text{out}}(-\omega)\delta Y_{\text{out}}(\omega) \rangle + 1 \] (3.54.b)

Where the +1 come from the contribution of the vacuum, and:

\[
\begin{align*}
\delta X_{\text{out}}(\omega) &= \delta X(\omega) + \delta X_{s}(\omega) \\
\delta Y_{\text{out}}(\omega) &= \delta Y(\omega) + \delta Y_{s}(\omega)
\end{align*}
\]

(3.55.a, b)

In chapter four we discuss the time scales of the various processes we have analyzed in general in this chapter.
CHAPTER IV

RESULTS AND DISCUSSION

In this chapter we describe the parameters of our model, use these to compute the properties of the emitted light, and discuss the ways in which the photon flux emitted by the DNL compares with that of an ordinary (but much larger) laser.

Dipole Moments and Time Scales

To calculate and plot the various results we have reached in the proceeding chapters we need first to set the values of the various variables we are working with. This we do now:

We start with the dipole moment of the TLS, $\mu_2$ this can be calculated by taking the TLS to be consisting of an electron oscillating in a cavity of sizes of the order of few nanometers. The dipole moment is then given by the charge of the electron multiplied by the size of the cavity: this is on the order of $\mu_2 = e.d = 1.6 \cdot 10^{-19} \cdot 10 \cdot 10^{-9} \approx 10^{-27} \text{ C.m}$

The induced electrostatic dipole moment of the nano particle $\mu_0$ can be calculated classically and is found to be, in dipole approximation (see appendix):

$$\mu_0 = -2\mu_2 \left( \frac{a}{R_2} \right)^3 \left[ \frac{\epsilon_r - \epsilon_m}{\epsilon_r + 2\epsilon_m} \right]$$

(4.1)

Where $a$ is the radius of the NP, $R_2$ is the distance between the TLS and NP, $\epsilon_r$ and $\epsilon_m$ are the real parts of the permittivity of the NP and the surrounding material respectively.
If we take the NP to be made of silver, the surrounding material to be made of silicon, and for radius of the NP about 7 nm, and $R_q$ on the order of 50 nm, the induced dipole moment of the NP can be made as large as $\mu_0 = 10^{-26} - 10^{-27} \text{C} \cdot \text{m}$ for frequencies near the plasmon resonance. This resonance happens when the term $\epsilon_P = \frac{\epsilon_r - \epsilon_m}{(\epsilon_r + 2\epsilon_m)}$ diverges. We can see this by plotting $\epsilon_P$ vs. frequency. We show this below for silver where the resonance frequency is found to be around $\omega_c = 4.27 \cdot 10^{15} - 4.28 \cdot 10^{15}$ which is very close to the resonant found in chapter 1.4 using full Mie theory:

![Figure 4.1. EP term plotted vs. frequency (Hz). The spike shows where the plasmon resonance occurs.](image)

Expression (4.1) is computed in dipole approximation: $\lambda / a << 1$. Corrections to the dipole approximation are also resonantly enhanced. They contribute with the same sign leading to even larger values of $\mu_0$ when the NP diameter becomes comparable to the inter-particle separation. Eq. (4.1) should be thought of as a lower bound on the induced dipole moment. We will use it in this work so as to be as conservative as possible in our conclusions.
The dipole nanolaser requires that the near field of the TLS be able to produce a strong polarization of the NP. This means that the inter-particle separation must be small compared to a critical value: \( R_q \leq r_{cr} = \left(4|\alpha_0||\alpha_2|\right)^{\frac{1}{6}} \) (I. E. Protsenko, et al, 2005)

where \( 4\pi \varepsilon_0 \alpha_0, 4\pi \varepsilon_0 \alpha_2 \) are the polarizabilities of the NP and the TLS respectively. This condition will guarantee that the TLS and NP interact through their near fields.

If \( R_q \geq r_{cr} \), higher moments \( \propto 1/R_q^4 \) should be added to the interaction energy term. Moreover, \( r_{cr} \) should be bigger than the sum of the sizes of the TLS and NP, i.e. \( r_{cr} > a + r_2 \), since we are not interested in the case where the two particles physically overlap one another. Also for the dipole approximation to be valid, the distance \( R_q \) between the two elements should be much less than the wavelength of the light emitted: \( r_2, a \ll R_q \ll k^{-1} \), taking all the above in consideration we find that the following condition should be valid:

\[
    r_{cr} - (a + r_2) >> \max(2r_2, 2a)
\]

This condition is satisfied by our choices of parameters.
The spontaneous decay of the TLS can be calculated using the following equations:

\[ \gamma_2 = \frac{\omega^2 \mu^2}{3\pi e_0 \hbar c^3} + \left(7.6 \times 10^9 \text{s}^{-1} \text{K}^{-1}\right)T \]  

(4.2)

The first term is due to radiative loses computed from Wigner-Weisskopf theory and the second term is due to the excitonic lifetime of a quantum dot (M. Bayer and A. Forchel, 2002) where \( T \) is the working temperature of the system. In this work, \( T \) lies in the range of 30-100 K so that radiative damping of the TLS dominates non-radiative damping. \( \varepsilon_0 \) is the permittivity of free space, and \( \hbar \omega \) is the energy separation of the two states of the TLS. We calculated a value of \( \gamma \) on the order of \( 10^{11} \text{s}^{-1} \).

The NP decay time can be taken to be around \( 10^{14} \text{s}^{-1} \) (Stietz, F et al. 2000), and it can be approximated by the following equation:

\[ \gamma = \frac{2 \mu^2}{3 \varepsilon_0 \hbar c^3} + \frac{\mu^2}{\hbar \alpha_{r_0}} \]  

(4.3)

Here again the first term is due to radiative loses, and the second term to resistive losses computed in a Drude model. \( \alpha_{r_0} \) is the polarizability of the nano particle and is given by the following (L. D. Landau and E. M. Lifshitz, 2001):

\[ \alpha_{r_0} = \frac{4}{3} \pi a^3 \left( \varepsilon_p + \frac{(\varepsilon_m - 1)^2}{\varepsilon_p} \right) \]

where \( \varepsilon_p = \frac{\varepsilon_i}{\varepsilon_m} \) and \( \varepsilon_m = \frac{\varepsilon_i}{\varepsilon_m} \) and \( \varepsilon_i \) is the imaginary part of permittivity of the NP material, and \( k \) is the wave vector of the surrounding material:

\[ k = k_1 + ik_2 \]

\[ k_1 = \omega \sqrt{\frac{\mu_0}{2}} \left( \sqrt{\varepsilon_{im}^2 + \varepsilon_m^2 + \varepsilon_m} \right) \]

\[ k_2 = \omega \sqrt{\frac{\mu_0}{2}} \left( \sqrt{\varepsilon_{im}^2 + \varepsilon_m^2 - \varepsilon_m} \right) \]
Here $e_{im}$ is the imaginary part of the permittivity of the surrounding material (taken to be silicon in this work). The values of the permittivities are calculated by interpolating data taken from tables in the CRC Handbook of Chemistry and Physics 48th edition.

The decay constant for the TLS and the NP given by the above equations have the following values: $\gamma \approx 10^{14} \text{ s}^{-1}$ and $\gamma_2 \approx 10^{11} \text{ s}^{-1}$ and it is clear that the condition for adiabatic approximation $\gamma_2 >> \gamma$ used in case one of chapter 3.2, the standard laser theory, is not valid for silver particles in a silicon matrix.

The threshold pumping is given by Eq. (3.9) $\Lambda_p \geq \frac{2\gamma \gamma_c + 4G \gamma_2}{2G - \gamma}$ where $G = \frac{2 \Omega^2}{\gamma_c}$.

The coupling $\Omega$ is given by $\Omega = \frac{\mu_0 H_0}{4\pi \epsilon_0 h R_q^3} \approx 10^{13} \text{ s}^{-1}$ using the above numbers.

Care must be exercised to be certain that one is operating in a physically allowed region of parameter space. When choosing our numbers we have to make sure that all the below condition are met simultaneously:

1. The pump rate $\Lambda_p$ should always be positive. Some values of $\gamma_c$, can lead to a negative pumping as shown in Fig. (4.3), where it is clear that the pumping rate swings negative for $\gamma_c$ bigger than a certain value, $\gamma_c > 3.3 \cdot 10^{13} \text{ s}^{-1}$ in this case.
Figure 4.3. The pumping rate (s⁻¹) vs. γc (s⁻¹): the effective decay of the TLS.

2. As mentioned at the end of chapter 3.1 the lasing condition should always be satisfied: \( \Omega^2 > \frac{\gamma c \Gamma}{4} \).

3. The spectral width should always be positive: \( \Gamma > 0 \).

Once we have chosen a material, e.g. silver, the resonance frequency \( \omega_c \) will be fixed and we can work in a relatively narrow region around it, say of width \( 10^{12} \) Hz. The only free parameters available for us to change are: the average noise quanta of the reservoirs: \( \bar{n}_s \), and \( \bar{n}_c \), the size of the nano particle: \( a \), the distance between the TLS and the NP: \( R_g \), and the temperature \( T \). We find that \( T \) doesn’t have a significant effect in the temperature regime we are working in and we will not investigate it further, other than to note the following:

A low temperature regime 30-100 K is necessary because for higher temperatures, the condition \( r_{cr} - (a + r_z) >> \max(2r_z, 2a) \) does not hold anymore. This happens because as temperature rises, the average induced dipole moment of the NP decreases due to random thermal agitation and the corresponding interaction \( r_{cr} \) radius decreases.
Intensity Spectrum

Using the parameters above, we can find the intensity spectrum given by Eq. (3.34):

\[ S_1(\omega) = \frac{A_\omega^2}{2\pi} \left( I_{NP}(\omega) + I_{TLS}(\omega) \right) \left( \frac{1}{(\omega - \omega_0)^2 + \Gamma^2} \right) \]

Where \( \Gamma \) is the width of the spectrum given by Eq. (3.23) for case one:

\[ \Gamma = \frac{1}{4N_0} \left( \gamma \left( \bar{n}_a + \frac{1}{2} \right) + \frac{\Omega^2}{\gamma_c} \right) \]

And by Eq. (3.29) for case two:

\[ \Gamma = \frac{(2\bar{n}_a + 1)}{8N_0} \gamma \]

The spectral width is one of the most important characteristics of the laser field. It is defined as the FWHM of the intensity spectrum. A large value of \( \Gamma \) corresponds to a spectrally impure radiated field. Lasers, of course are justly famous for their spectral purity, which decrease with increasing photon number, proportional to \( N_0 \) in our case. For traditional lasers, \( \Gamma \) arises most commonly from phase drift due to different noise-contributing factors, such as noise from the mirrors of the cavity e.g. vibrations and imperfections, broadening due to lifetime of the gain medium, noise coming from the unstable pumping mechanism etc.

The spectral width of the lasers known to science varies from 10’s of GHz for multimode lasers, MHz for semiconductor lasers, to kHz for free running solid state lasers, even below 1 Hz for highly stabilized lasers. A narrow spectral width is required for many laser applications such as spectroscopic measurements. We note a critical difference
between ordinary laser light and the light radiated by the DNL: the spectral width is much larger for the latter system, since $\gamma$ is so much larger. This is not hard to understand: the typical line width for an empty laser cavity is on the order of $c/2L$ where $L$ is the cavity length, i.e. on the order of $10^8$ s$^{-1}$. Using this expression and scaling the lengths from 50cm to 50nm results in a width of $10^{15}$ s$^{-1}$. Of course, our DNL does not consist of light confined to a cavity of this size but the computed radiative width of the NP is very similar.

The closest resemblance to a typical laser would have parameters appropriate to case one: $\gamma_2 \gg \gamma$. While this does not correspond to metal NP’s in a silicon matrix, it might conceivably be engineered by using different kind of materials than those we have been considering. We choose some arbitrary numbers for $\gamma$ and $\gamma_2$. For acceptable pumping rates and for $\bar{n}_c = 0$ we plot the spectrum in Fig. (4.4) for case one with the following parameters: $\bar{n}_a = 0$, and $\bar{n}_c = 0$, $\Omega = 10^{13}$ s$^{-1}$, $R_q = 50$ nm, $\Lambda_p = 10^{13}$ s$^{-1}$, $a = 7$ nm, $\gamma_2 = 10^{13}$ s$^{-1}$, and $\gamma = 10^{12}$ s$^{-1}$.

![Graph](image-url)

**Figure 4.4.** Spectrum of light emitted from the TLS-NP system. Vertical axis in units of $\left(\frac{V S}{m}\right)^2$, horizontal axis in Hz, and the electric field are evaluated one micron from the TLS-NP system.
Here the number of plasmons excited $N_0$ is 5, and the light emitted from NP is much bigger than that from the TLS, $I_{NP} \gg I_{TLS} L^2$ in agreement with the work of Protsenko, et. al. The line width is about the same as for the NP $\Gamma \approx \gamma = 10^{12}$. The factor $1/N_0$ tends to decrease the width while the factor $\Omega^2/\gamma_c$ tends to increase it. The net result is little change.

Increasing the pumping of the TLS, we see a reduction in the line width. Also, as pointed earlier in chapter three, increasing the noise quanta of the TLS will reduce the line width even more. For example increasing the number of noise quanta in around the TLs to $\bar{\pi}_c = 12$, will reduce the width almost a hundred times $\Gamma = 10^{10}$ as shown below:

![Figure 4.5](image)

Figure 4.5. The reduced spectral width of the light emitted by the system for case one when $n_c= 12$. Units are same as Fig.(4.4).

In this case the number of plasmon excited is about 45 which help reducing the width along with $\gamma_c$. The number of photon emitted can be calculated by the following relation:
\[ \Gamma \cdot N_0 = 10^{10} \cdot 45 \approx 10^{12} \text{ photons} \cdot \text{s}^{-1}. \] We note that corrections to the dipole approximation cannot bring us into this regime because those corrections not only increase \( \mu_0 \) but also greatly increase \( \gamma \propto \mu_0^2 \).

The differences between case one and case two have to do with two effects: the increased value of the plasmon width \( \gamma \) (almost entirely radiative) and the corresponding decrease of polariton number, as shown in Fig. (4.6):

![Figure 4.6](image)

Figure 4.6. The relation of the plasmon number \( N_0 \) on the life time of the NP in unit of inverse seconds.

The pumping also depends sensitively on \( \gamma \) as seen below:

![Figure 4.7](image)

Figure 4.7. The dependence of pumping (s\(^{-1}\)) on the decays of the NP \( \gamma \) (s\(^{-1}\)).
For the second case, using parameters suitable to metal nano-particles like silver and gold in a silicon matrix, we obtain the following typical distribution:

\[
\begin{array}{ccc}
14 & 14 & 15 \\
1 & 5 & 0 \\
5 & 1 & 0 \\
110 & -1 & -1 & 0 \\
-5 & 10 & \\
\end{array}
\]

Figure 4.8. The spectral width of the light emitted by the system for regime of the following numbers \( \bar{n}_a = 0 \), and \( \bar{n}_c = 0 \), \( \mu_0 = 10^{-26} \), \( R_q = 50 \) nm, \( \Lambda_p = 10^{13} \), \( a = 7 \) nm and \( \gamma = 10^{14} \) s\(^{-1} \). Here the spectral width actually increases above \( \gamma \): \( \Gamma \approx 4 \cdot 10^{14} \) s\(^{-1} \).

As in case one, the width can be reduced by increasing the pumping. But to reduce the width a hundred times below \( \gamma = 10^{14} \) s\(^{-1} \) the pumping needs to be increased greatly, into the optical range \( \Lambda_p = 10^{15} \) s\(^{-1} \) or higher, and the stability of the system at such high pump values would need to be addressed, even if these values could be technologically attained.

Again, as for the first case, increasing \( \gamma_c \) will decrease the line width. This is not obvious from Eq. (3.29) as \( \gamma_c \) is not explicitly present there, but it is still there implicitly in \( N_0 \), as seen below:

Figure 4.9. Dependence of the mean plasmon number on effective decay of the TLS \( \gamma_c \). \( \bar{n}_a = 0 \), and \( \bar{n}_c = 12 \), \( \mu_0 = 10^{-26} \) C.m, \( R_q = 50 \) nm, \( a = 7 \) nm and \( \gamma = 10^{14} \) s\(^{-1} \).
An increased value of $\gamma_c$ requires, of course, an increased pump rate to exceed the threshold condition. We plot below a typical spectrum for $\gamma_c \approx 10^{12} \text{s}^{-1}$.

![Spectrum](image)

Figure 4.10. The spectral width of the light emitted by the system. Here the spectral width decreases below $\gamma$: $\Gamma \approx 10^{13} \text{s}^{-1}$, $n_a = 0$, and $n_c = 12$, $\mu_0 = 10^{-26} \text{C.m}$, $R_q = 50 \text{nm}$, $A_p = 5 \cdot 10^{15} \text{s}^{-1}$, $a = 7 \text{nm}$ and $\gamma = 10^{14} \text{s}^{-1}$.

In case two the number of plasmons is much smaller than for case one, on the order 0.6. Additionally, the light emitted comes roughly equally from the TLS and NP,

$$I_{NP} \approx I_{TLS} L^2$$

disagreeing with Protsenko, et. al.

We can also calculate the rate of photons emitted: $\Gamma \cdot N_0 = 10^{13} \cdot 0.6 \approx 10^{13}$ photons $\cdot s^{-1}$.

This is a factor of $10^2$ above the estimate made in the introduction.

We also realize that the maximum of $S_1$ of case two is on the order of $10^{-26} \left( \frac{V_s}{m} \right)^2$,

which is $10^7$ time smaller than the intensity produced in the first case. This is due mainly to the fact that in case one the number of plasmons excited, $N_0$, is about 100 times bigger in case one than in case two.
Increasing the pumping more will decrease the width even more as this increases the number of polaritons excited \( N_0 \), which in turn, as mentioned before, decreases the line width. This is shown below in:

![Graph](image)

Figure 4.11. The relation of the plasmon number \( N_0 \) on pumping of the TLS (s\(^{-1}\)).

The second order coherence spectrum \( S_2(\omega, r) \) given by Eq. (3.35) can be plot as shown below:

![Graph](image)

Figure 4.12. Second order coherence for the situation. \( \Gamma \approx 10^{13} \text{s}^{-1} \), \( \mu_0 = 10^{26} \text{C.m} \), \( R_q = 50 \text{nm} \), \( A_p = 5 \times 10^{15} \text{s}^{-1} \), \( a = 7 \text{ nm} \) and \( \gamma = 10^{14} \text{s}^{-1} \).

As mentioned before the second order degree of coherence measures the degree of fluctuation in intensities, of the sort that would be measured in a Hanbury-Brown-Twiss type experiment. The narrower the spectrum the fewer fluctuations we have. As it is
obvious from Eq. (3.35) the first order and second order coherence share almost the same Lorentzian line width, reduction in the spectral width in the intensity will automatically leads to reduction in the width of second order coherence.

The second order coherence $G^2(t)$ is a strictly decreasing function of time and implies classical-type light.

**Squeezing Spectrum**

We do not expect significant squeezing from the continuously pumped, randomly emitting light source described by our model of the dipole nanolaser. In laser systems with highly stabilized pumping (so-called “regular pumping”) it is possible to obtain amplitude-squeezed light (T. C. Ralph, et al 1995). Had we investigated the dynamics of the pumping, including appropriate control parameters, we no doubt would have found a similar sort of phenomenon here. Despite the wide variety of parameters open to our theoretical investigation, however, squeezing does not appear to be an option for the continuously pumped dipole nanolaser. We report our investigations below.

We plot the squeezing spectrum using Eq.’s (3.54-3.55):

Amplitude $\langle \delta X_{\text{out}}(-\omega) \delta X_{\text{out}}(\omega) \rangle + 1$

Phase $\langle \delta Y_{\text{out}}(-\omega) \delta Y_{\text{out}}(\omega) \rangle + 1$

To do so we use the expectation values of the by product of the various noise terms calculated in chapter two Eq. (2.42) after we take their Fourier transformation:

$\langle F^\dagger(\omega)F(\omega') \rangle = \bar{n}^4 \frac{\nu}{2} \delta(\omega + \omega')$

$\langle F(\omega)F^\dagger(\omega') \rangle = (\bar{n}^4 + 1) \frac{\nu}{2} \delta(\omega + \omega')$
\[ \langle F_+ (\omega) F_+ (\omega') \rangle = \left( \frac{1}{2} \gamma_2 \bar{n}^C + \frac{\Lambda}{8} [1 - \Sigma_{z_0}] \right) \delta(\omega + \omega') \]  

\[ \langle F_- (t\omega) F_+ (\omega') \rangle = \left( \frac{1}{2} \gamma_2 (\bar{n}^C + 1) + \frac{\Lambda}{8} [\Sigma_{z_0} - 1] \right) \delta(\omega + \omega') \]  

\[ \langle F_+ (\omega) F_+ (\omega') \rangle = \left( \gamma_p + (\gamma_2 - \frac{\Lambda}{2}) \Sigma_{z_0} \right) \delta(\omega + \omega') \]  

In our work, we set the reservoirs quantas \( \bar{n}_q \) to zero. These quantas represent the amplitude of the ambient noise that affects the NP. This is justified by the fact that we are working in a low temperature regime \( T=30-100 \, k \), so the existence of ambient optical photons is very small.

As mentioned in chapter one, the product of the variances of the quadratures should always maintain the uncertainty principle: \( \Delta X \Delta Y \geq 1 \). Now for a squeezing to happen, one of the quadratures will have less than 1 value on the expense of increasing in the value of the other. As in most lasers, the phase quadratures always diverge due to phase drift. This would give the possibility for the amplitude quadrature to go below one while still satisfying the uncertainty principle.

By taking the above in consideration and by using the parameters calculated at the beginning of this chapter for silver, the plots of amplitude quadrature takes the following shapes:
Figure 4.13. Amplitude quadrature plotted against frequency around the central frequency $2.48 \times 10^{15}$ Hz.

Our phase quadrature diverges expectedly, but the amplitude quadrature doesn't go below one, as obvious from Fig. (4.13). Even when we use the other numbers regime which reduces the line width, we find that no significant squeezing is present. However, note that the amplitude squeezing does not exceed unity and so the light emitted by the DNL is no more chaotic than thermal radiation.

We also investigated the more complicated system of a single NP situated in the vicinity of two TLS’s oscillating in a quantum-mechanically coherent mode (so called atomic squeezing). While we do not report these investigations here, we found that there was no noticeable squeezing for this system either.

**Coherence Length**

The coherence length, is the length in which the laser field keeps its coherence, and is defined by: $l_c = \frac{\lambda^2}{\Delta \lambda}$, where $\Delta \lambda = \frac{\lambda^2 \Gamma}{c}$, Where $\Gamma$ is the line width. For our silver
-in-silicon parameters \( \Delta \lambda = \frac{(10^{-6})^2}{3 \cdot 10^8} \approx 10^{-6} \Rightarrow l_e \approx 10^{-6} m = 1 \mu m \). Since photons are being emitted in bursts at the rate of \( 10^{11} \) photons s\(^{-1} \) and the duration of emission is on average only \( 10^{-14} \) s, the average separation between photons in different bursts is about 3mm. We conclude that there is no coherence from one burst to another.

It is clear that the dipole nanolaser does not produce anything like ordinary laser light. In fact, what it produces are single photons that can lie anywhere in a broad spectral region. These photons are weakly correlated with one another (correlation length \( \approx 1 \mu m \)) and are emitted in a two-lobed dipole pattern. The advantage of using the dipole nanolaser over simply a pumped TLS is the strong mutual interaction of TLS and NP which greatly increases the radiant efficiency of the system \( L^2 \approx 10 \) for case two). The price one pays for this enhanced efficiency (at least when operating with metal NP’s in a silicon matrix) is a broad spectrum. The photons are emitted in short time intervals, the average duration of a radiation process being \( 1/\Gamma \), and the interval between successive emissions is about \( \frac{\hbar \omega}{P} \approx 1/\Gamma \).

Another result of this work is the prediction of a narrow-\( \Gamma \) regime of parameters as in case one. Should these parameters be realizable by existing or new materials and technology, the dipole nanolaser would produce light of a width similar to that of conventional multi-mode lasers.

A spectral width of \( 10^{12} \) s\(^{-1} \) hardly resembles ordinary laser light and a spectral width of \( 10^{14} \) s\(^{-1} \) even less so. In the latter case we are not speaking about a continuously emitted beam but rather a series of single-photon pulses similar to that emitted by the TLS acting...
alone. This is reminiscent of the device described by D.J. Bergman and M.I. Stockman (2003), in which modes confined to a metallic surface are generated as femtosecond pulses. Even the lower limit of $10^{12} \text{s}^{-1}$ implies significant temporal structure in the emitted light. We cannot predict the nature of this structure without going beyond the factorization approximation of chapter three.
CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

The miniaturization of optical components has proceeded dramatically in recent years. Today we can process information using millimeter lasers and photon waveguides etched onto a chip. The trend toward further miniaturization will no doubt continue well into the future. Information processing by nanometer and even atomic sized components has been suggested for quantum computing. It is probably impossible to construct a light source smaller than a few nanometers and the device we have been investigating theoretically in this work, the dipole nanolaser, is at the outer limits of physically attainable miniaturized technology. Our work is intended to inform experimenters of the likely optical properties of this device.

The DNL is not a laser in the conventional sense of the term. Electronic oscillations of a nearby quantum dot stimulate resonant plasma oscillations of a metallic nanoparticle. The equations describing the coupling of quantum dot and nanoparticle are isomorphic to the equations describing laser oscillations, with the DNL’s quantum dot corresponding to the standard laser’s population inverted active medium and the plasmon amplitude corresponding to standard laser’s the cavity field. Thus, it is the plasmons which are “lasing”. Both the plasma oscillations and the quantum dot radiate and this work has reported a study of the nature of this radiation.
visualization of the plasmon field. We solved the equations of motion for the coupled plasmon and quantum dot, found the threshold conditions for plasmon lasing and computed the inversion, TLS dipole moment, and plasmon number in mean field approximation. The coupling of plasmons and quantum dot to the radiation field was taken to be of standard form and the resulting radiation was studied in detail by the methods of quantum optics.

We have learned that there are two regimes of operation. In the first case the plasmon lifetime is much longer than the quantum dot lifetime. This is similar to standard laser theory where the cavity field lifetime is much longer than the atomic lifetimes. We showed that the line width can be made smaller than the natural line width of the nanoparticle. This is not surprising since ordinary lasers show a decreased line width with increasing cavity field. In this regime the number of excited polaritons is relatively large and most of the light is emitted from the NP. This regime, however, requires materials of a novel sort.

In the second case, corresponding to conventional materials, the plasmon lifetime is much shorter than the quantum dot lifetime, a highly non-standard situation in laser theory. We found that the loss of plasmon amplitude to the radiation field so limits the plasmon number that no significant line width shortening such as found in table top lasers occurs. The plasmons and their associated radiation are quite broad. In fact their spectral width is practically in the optical range. This implies a significant temporal structure to the resulting radiation. The light is emitted in extremely short bursts rather than continuously. Each burst corresponds to a single photon. There is no natural tendency of this radiation to form correlated bursts: The computed statistics such as the squeezing
spectrum are entirely consistent with the conclusion that the emitted photons occupy a broad range of frequencies and are emitted in a Gaussian random fashion. The expectation that light might be emitted in something like a number state is unfounded. Whether this limits the utility of the DNL or not depends on the application.

The work presented here can be improved in a number of ways. At the classical level, the TLS-NP coupling should be described by the results of a true dynamical calculation rather than by using the electrostatic induced dipole moment of a conductor in the presence of an external charge. Such a calculation would include the effects of retardation. At the quantum level, the uncontrolled approximation of using the factorization approximation to find mean field quantities is known to be accurate only when describing something like a quantum mechanical coherent state.

An assessment of corrections to this approximation would be useful in advancing understanding beyond this work, particularly given the small polariton number implied by our calculations. The use of Markoffian white noise for the noise operators needs to be investigated for systems operating with such large decay constants as those that characterize the case 2.

In conclusion our work showed us that the DNL nano-laser is efficient in producing very fast pulses of light. This light in general has a very wide frequency range, but in principle, and for specific materials, the line width can be reduced 100 below the natural line width of the NP. Experiments are eagerly awaited.
APPENDIX

A1. The amplitude and phase quadratures of the nano-particle are given by the following;

\[
\delta X(\omega) = \frac{1}{2} S_{x,z}(\omega) F_{c,z}(\omega) + S_{x,ca}(\omega) F^*(\omega) + S_{x,a}(\omega) F_a(\omega) + S_{x,-}(\omega) F_{c,-}(\omega) + S_{x,+}(\omega) F_{c,+}(\omega) \]

\[
\delta Y(\omega) = \frac{S_{y,a}(\omega)[F^*(\omega) - F_a(\omega)] + S_{y,c}(\omega)[F_{c,-}(\omega) + F_{c,+}(\omega)]}{D_y(\omega)}
\]

Where the various coefficients are given below;

Case one: \( \gamma_2 >> \gamma \)

\[
D_x = 2\omega^2 \gamma_c + i \omega \gamma_c \gamma - 4i \omega \Omega \Sigma_{z0} + 2i \gamma_p \omega \gamma - \gamma_p \gamma_c \gamma + 4 \gamma_p \Omega^2 \Sigma_{z0} + i 16 \Omega^2 A_0^2 \omega
\]

\[
-8 \Omega^2 A_0^2 \gamma + i 16 \Omega^3 A_0 \Sigma_{z0}
\]

\[
S_{x,z} = -4 \Omega^2 A_0
\]

\[
S_{x,ca} = S_{x,ca} = i \omega \gamma_c - \gamma_p \gamma_c - 8 \Omega^2 A_0^2
\]

\[
S_{x,-} = -S_{x,+} = 2 \omega \Omega + 2i \gamma_p \Omega
\]

\[
D_y = -2i \omega \gamma_c + \gamma_c \gamma - 4 \Omega^2 \Sigma_{z0}
\]

\[
S_{y,c} = -4 \Omega
\]

\[
S_{y,a} = 2i \gamma_c
\]

And the various noise terms, the \( F \)'s, are given in chapter II.

Case two: \( \gamma >> \gamma_2 \)

\[
D_x = (-4 \omega) 3 \gamma^2 2 \gamma - 64i A0^2 \gamma \Omega 2 \omega 2 - 4i \omega \gamma_c \gamma + 16 A0^2 \gamma 2 \Omega 2 \omega
\]

\[
-16i \omega^3 \Sigma m0 A0\gamma_c \omega + 16i \gamma_p \omega^4 - 16 \omega^3 \gamma_c \gamma - 4i \omega^2 \gamma_c \gamma - 4 \omega^3 \gamma_c \gamma - 16i \gamma_p \omega^2 \gamma_c \gamma - 8i \gamma_p \gamma_c \gamma \Omega^2 \Sigma z0
\]

\[
\omega \gamma_c \gamma 2 \omega + 16i \omega^2 \Sigma z0 \Omega 2 \gamma_c + 16i \omega^2 \Sigma z0 \Omega 2 \gamma - 4i \gamma_p \gamma_c \gamma 2 \omega + i \gamma_p \gamma_c \gamma 2 \omega + 4 \gamma_p \gamma_c \gamma 2 \omega
\]

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\[ 16i\Omega^3 \cdot \Sigma p0A0\cdot \gamma c\cdot \omega + 16i\cdot \Omega^3 \cdot \Sigma p0A0\cdot \omega \cdot \gamma - 8\Omega^3 \cdot \Sigma p0A0\cdot \gamma c\cdot \gamma + 8\Omega^3 \cdot \Sigma m0A0\cdot \gamma c\cdot \gamma - 16\gamma p\cdot \Sigma z0\Omega^2 \cdot \omega \cdot \gamma \]

\[ 32i\cdot \gamma p\cdot \Sigma z0\Omega^2 \cdot \omega^2 - 16i\cdot \gamma p\cdot \gamma c^2 \cdot \omega^2 + 16i\cdot \omega^4 \cdot \gamma + 32\omega^3 \cdot \Sigma z0\Omega^2 - 64A0^2\cdot \gamma c^2 \cdot \Omega^2 - 16\gamma p\cdot \gamma c^3 + 16i\cdot \gamma p\cdot \Sigma z0^2\cdot \Omega^4 \]

\[ 16\omega^5 + 16\omega^2 \cdot \Sigma z0^2 \cdot \Omega^4 - 32\omega^5 \cdot \Sigma m0A0\cdot \Sigma z0 - 32\Omega^3 \cdot \Sigma m0A0\cdot \omega^2 + 32\omega^3 \cdot \Sigma p0A0\cdot \Sigma z0 + 32\Omega^3 \cdot \Sigma p0A0\cdot \omega^2 \]

\[-64A0^2\cdot \omega^2 \cdot \Omega^4 \cdot \Sigma z0 - 16\gamma p\cdot \Sigma z0\Omega^2 \cdot \gamma c^2 \cdot \omega + 32A0^2\cdot \gamma c^2 \cdot \Omega^2 \cdot \gamma c - 16\gamma p\cdot \omega^3 \cdot \gamma + 16i\cdot \omega^4 \cdot \gamma \]

\[ S_{\chi c0}(\omega) := 0 \]

\[ S_{\chi,2} = (-32i\cdot A0(\omega)\cdot \Omega(\omega)^4 \cdot \Sigma z(\omega) - 32i\cdot A0(\omega)\cdot \Omega(\omega)^2 \cdot \omega^2 + 16A0(\omega)\cdot \Omega(\omega)^2 \cdot \omega \cdot \gamma(\omega)) + 16A0(\omega)\cdot \Omega(\omega)^2 \cdot \gamma c(\omega) + 8iA0(\omega)\cdot \Omega(\omega)^2 \cdot \gamma c(\omega) \]

\[ S_{\chi,3} = (-16i\cdot \gamma p\cdot \omega^2 \cdot \Sigma z0 + 2i\cdot \gamma p\cdot \gamma c^2 \cdot \gamma + 16i\cdot \omega^4 - 16\omega^3 \cdot \gamma c - 16\gamma p\cdot \omega^3 + 8\gamma p\cdot \gamma c^2 \cdot \omega + 32A0^2 \cdot \Omega^2 \cdot \gamma c\cdot \omega - 8\omega^3 \cdot \gamma + 16i\cdot \omega^2 \cdot \Omega^2 \cdot \Sigma z0 + 4i\cdot \gamma p\cdot \gamma c^2 \cdot \omega - 8\omega^3 \cdot \gamma c - \omega^2 \cdot \gamma c^2 - 8i\cdot \omega^2 \cdot \gamma c^2 + 64iA0^2 \cdot \Omega^2 \cdot \omega^2 + 16iA0^2 \cdot \Omega^2 \cdot \gamma c^2 + 8i\cdot \gamma p\cdot \gamma c \cdot \omega - 32i\cdot \gamma p\cdot \omega^2 \cdot \gamma c - 8i\cdot \gamma p\cdot \omega^2 \cdot \gamma^2 + 32A0^2 \cdot \Omega^2 \cdot \omega^2 \cdot \gamma - 64iA0^2 \cdot \Omega^2 \cdot \Sigma z \]

\[ S_{\chi,4} = (-32i\cdot \Omega^4 \cdot \Sigma m(\omega) \cdot A0 + 4i\cdot \gamma p\cdot \Omega \cdot \gamma c \cdot \gamma - 4i\cdot \omega \cdot \Omega \cdot \gamma c \cdot \gamma + 16i\cdot \omega \cdot \Omega^3 \cdot \Sigma z0 - 8i\cdot \gamma p\cdot \Omega \cdot \omega \cdot \gamma - 8i\cdot \gamma p\cdot \gamma c - 32i\cdot \omega^3 \cdot \Sigma p0A0 + 16i\cdot \omega^3 \cdot \Omega^2 - 16\gamma p\cdot \Omega^2 \cdot \omega^2 - 8\omega^2 \cdot \Omega \cdot \gamma c - 8\omega^2 \cdot \gamma \cdot \Omega - 16\gamma p\cdot \Omega^2 \cdot \Sigma z \]

\[ S_{\chi,5} = (-4i\cdot \gamma p(\omega) \cdot \Omega(\omega) \cdot \gamma c(\omega) \cdot \gamma(\omega) - 16i\cdot \omega^3 \cdot \Omega(\omega) - 16i\cdot \omega \cdot \Omega(\omega)^3 \cdot \Sigma z(\omega) + 8\omega^2 \cdot \Omega(\omega) \cdot \gamma c(\omega)) - 32i\cdot \Omega(\omega)^4 \cdot \Sigma p0(\omega) \cdot A0(\omega) - 32i\cdot \Omega(\omega)^4 \cdot \Sigma m(\omega) \cdot A0(\omega) + 8\omega^2 \cdot \gamma(\omega) \cdot \Omega(\omega) + 16\gamma p(\omega) \cdot \Omega(\omega)^3 \cdot \Sigma z(\omega) + 16\gamma p(\omega) \cdot \Omega(\omega) \cdot \omega^2 + 8i\gamma p(\omega) \cdot \Omega(\omega) \cdot \gamma c(\omega) \cdot \omega + 8i\gamma p(\omega) \cdot \Omega(\omega) \cdot \gamma c(\omega) \cdot \gamma + 4i\cdot \omega \cdot \Omega(\omega) \cdot \gamma c(\omega) \cdot \gamma(\omega) \]

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\[
\begin{align*}
Dy & := 4 \cdot \omega^2 + 2 \cdot i \cdot \omega \cdot \gamma + 2 \cdot i \cdot \gamma \cdot \omega - \gamma \cdot \gamma + 4 \cdot \Omega^2 \cdot \Sigma c \\
S_{\gamma c} & := 4 \cdot \Omega \\
S_{\gamma a} & := -(4 \cdot \omega + i \cdot 2 \cdot \gamma c)
\end{align*}
\]

**A2. Proof of** \( \langle A(t) A(0) \rangle = N_0 e^{-i[\phi(t) - \phi(0)]} = N_0 e^{-\frac{1}{2}(\phi(t))^2} \)

We start from the following; \( \langle e^{-i\Delta \phi(t)} \rangle = \sum_n \left( \frac{1}{n!} \Delta \phi^n \right) \)

But \( \Delta \phi \) involves single noise terms \( F_v \) and the expectation value of any odd product of the \( F \)'s will vanish as long as the noises are Markoffian's, so \( n \) can only be even \( n = 0,2,4 \). thus the sum will be as follow;

\[
\sum_{n=0,2,4} \left( \frac{-1}{n!} \Delta \phi^n \right) = \sum_{p} \left( \frac{-1}{2p!} \Delta \phi^{2p} \right) \quad \text{where} \quad p = 0,1,2,3,...
\]

Now each product of \( \Delta \phi \)'s can be written as a product of combination of double products as follow;

\[
\langle \Delta \phi(t_1) \Delta \phi(t_2) \rangle = \langle \Delta \phi(t_2) \Delta \phi(t_1) \rangle \\
\langle \Delta \phi(t_1) \Delta \phi(t_2) \Delta \phi(t_3) \Delta \phi(t_4) \rangle = \langle \Delta \phi(t_3) \Delta \phi(t_4) \rangle \langle \Delta \phi(t_1) \Delta \phi(t_2) \rangle \]

\[
= \langle \Delta \phi(t_2) \Delta \phi(t_1) \rangle \langle \Delta \phi(t_3) \Delta \phi(t_4) \rangle = \langle \Delta \phi(t_4) \Delta \phi(t_3) \rangle \langle \Delta \phi(t_2) \Delta \phi(t_1) \rangle \quad \text{etc...}
\]

It is clear then that each product is repeated \( \frac{2p!}{2^p p!} \) time. Thus, and after setting \( \phi(0) = 0 \) the sum will finally reduce to;

\[
\sum_{p} \left( \frac{-1}{2p!} \Delta \phi^{2p} \right) = \sum_{p} \left( \frac{-1}{2p!} \Delta \phi^{2p} \right) = \sum_{p} \left( \frac{-1}{2p!} \Delta \phi^{2p} \right) = e^{-\frac{1}{2}(\Delta \phi)^2}
\]
A3. Mie Theory

We start with a spherical nano particle subject to plane electromagnetic field.

\[ \mathbf{E}_{\text{out}} = \mathbf{E}_{\text{inc}} + \mathbf{E}_{\text{sc}} \]

where \( \mathbf{E}_{\text{out}} \) is the incoming field, \( \mathbf{E}_{\text{inc}} \) is the enclosed field in the nano-
particle, and \( \mathbf{E}_{\text{sc}} \) is the scattered field.

\[ \mathbf{E}_{\text{inc}} = \mathbf{E}_0 \, e^{i \kappa z} = \sqrt{4 \pi} \mathbf{E}_0 \sum_l j_l \sqrt{2l+1} j_l (\kappa r) Y_l^0 (\hat{r}) \]

\[ \mathbf{H}_{\text{inc}} = \left( \frac{\mathbf{\hat{z}} \times \mathbf{E}_0}{Z_2} \right) e^{i \kappa z} = \sqrt{4 \pi} \left( \frac{\mathbf{\hat{z}} \times \mathbf{E}_0}{Z_2} \right) \sum_l j_l \sqrt{2l+1} j_l (\kappa r) Y_l^0 (\hat{r}) \]

We take the general form \( \mathbf{E}_0 = E_0 \left( \cos \alpha \mathbf{\hat{x}} + e^{i \beta} \sin \alpha \mathbf{\hat{y}} \right) \). Written in spherical form,

\( (E_0)_x^l = -\frac{1}{\sqrt{2}} \left( (E_0)_0 - i (E_0)_y \right) = -\frac{E_0}{\sqrt{2}} \left( \cos \alpha + ie^{i \beta} \sin \alpha \right) \)

\( (E_0)_y^l = \frac{1}{\sqrt{2}} \left( (E_0)_0 + i (E_0)_y \right) = \frac{E_0}{\sqrt{2}} \left( \cos \alpha - ie^{i \beta} \sin \alpha \right), \quad (E_0)_0 = (E_0)_z = 0 \)

The internal and scattered fields are;

\[ \mathbf{E}_{\text{int}} = Z_1 \sum_{l,m} \left\{ \frac{i}{k} a_{lm}^{(E)} \nabla \times \left[ j_l (kr) \mathbf{\hat{X}}_m^l \right] + a_{lm}^{(M)} j_l (kr) \mathbf{\hat{X}}_m^l \right\} \]

\[ \mathbf{H}_{\text{int}} = \sum_{l,m} \left\{ a_{lm}^{(E)} j_l (kr) \mathbf{\hat{X}}_m^l - \frac{i}{k} a_{lm}^{(M)} \nabla \times \left[ j_l (kr) \mathbf{\hat{X}}_m^l \right] \right\} \]

\[ \mathbf{E}_{\text{sc}} = Z_2 \sum_{l,m} \left\{ \frac{i}{\kappa} b_{lm}^{(E)} \nabla \times \left[ h_l^0 (kr) \mathbf{\hat{X}}_m^l \right] + b_{lm}^{(M)} h_l^0 (kr) \mathbf{\hat{X}}_m^l \right\} \]

\[ \mathbf{H}_{\text{sc}} = \sum_{l,m} \left\{ b_{lm}^{(E)} h_l^0 (kr) \mathbf{\hat{X}}_m^l - \frac{i}{\kappa} b_{lm}^{(M)} \nabla \times \left[ h_l^0 (kr) \mathbf{\hat{X}}_m^l \right] \right\} \]

Where \( Z_j = \frac{\mu_0}{\varepsilon_j} \) is the wave impedance of the medium. All multipole amplitudes \( a_{lm} \).

\( b_{lm} \) have units of A/m.
I. Continuity of \( E_i \)

\[
\mathbf{\hat{r}} \times (\mathbf{\hat{E}}_{\text{inc}} + \mathbf{\hat{E}}_{\text{sc}}) = \mathbf{\hat{r}} \times \mathbf{\hat{E}}_{\text{int}} \quad \text{at} \quad r = a.
\]

\[
\mathbf{\hat{r}} \times \mathbf{\hat{E}}_{\text{inc}} = (\mathbf{\hat{r}} \times \mathbf{\hat{E}}_0) \sqrt{4\pi} \sum_l i^l \sqrt{2l+1} j_l(kr) Y_l^0(\mathbf{\hat{r}})
\]

\[
\mathbf{\hat{r}} \times \nabla \left[ z_i(qr) \mathbf{\hat{x}}_m^l \right] = \frac{q}{2l+1} \left[ l z_{i+1}(qr) - (l+1) z_{i-1}(qr) \right] \mathbf{\hat{x}}_m^l
\]

\[
\mathbf{\hat{r}} \times \mathbf{\hat{x}}_m^l = \frac{i}{\sqrt{2l+1}} \left[ \sqrt{l+1} \mathbf{\hat{x}}_{m+1}^{l+1} + \sqrt{l} \mathbf{\hat{x}}_{m}^{l+1} \right]
\]

\[
\mathbf{\hat{r}} \times (\mathbf{\hat{E}}_{\text{inc}} + \mathbf{\hat{E}}_{\text{sc}}) \bigg|_{r=a} = \mathbf{\hat{r}} \times \mathbf{\hat{E}}_{\text{int}} \bigg|_{r=a} \quad \Rightarrow
\]

\[
(\mathbf{\hat{r}} \times \mathbf{\hat{E}}_0) \sqrt{4\pi} \sum_l i^l \sqrt{2l+1} j_l(kr) Y_l^0(\mathbf{\hat{r}})
\]

\[
+ Z_2 \sum_{l,m} \left\{ \frac{i}{\kappa} b_{lm}^{(E)} \left[ \frac{\kappa}{2l+1} \left[ l h_{i+1}^l(\kappa a) - (l+1) h_{i-1}^l(\kappa a) \right] \mathbf{\hat{x}}_m^l \right] \right\}
\]

\[
+ \frac{i}{\sqrt{2l+1}} b_{lm}^{(M)} h_1^l(\kappa a) \left[ \sqrt{l+1} \mathbf{\hat{x}}_{m+1}^{l+1} + \sqrt{l} \mathbf{\hat{x}}_{m}^{l+1} \right]
\]

\[
= Z_1 \sum_{l,m} \left\{ \frac{i}{\kappa} a_{lm}^{(E)} \left[ \frac{k}{2l+1} \left[ l j_{i+1}^l(\kappa a) - (l+1) j_{i-1}^l(\kappa a) \right] \mathbf{\hat{x}}_m^l \right] \right\}
\]

\[
+ \frac{i}{\sqrt{2l+1}} a_{lm}^{(M)} j_1^l(\kappa a) \left[ \sqrt{l+1} \mathbf{\hat{x}}_{m+1}^{l+1} + \sqrt{l} \mathbf{\hat{x}}_{m}^{l+1} \right]
\]

Now

\[
\mathbf{\hat{r}} \times \mathbf{\hat{E}}_0 = i\sqrt{6} \left\{ \left[ r^i E_i^l \right] \mathbf{\hat{e}}_l^l \right\}^0 = i\sqrt{8\pi} \left\{ \left[ r^i E_i^l \right] \mathbf{\hat{e}}_l^l \right\}^0 = -i\sqrt{8\pi} \left\{ \left[ E_i^l r^{i-l} \right] \mathbf{\hat{e}}_l^l \right\}^0
\]

\[
= -i\sqrt{8\pi} \sum_{i,j} \sqrt{3} J^r \mathcal{W}(1,1,0,1;1,J') \left\{ E_i^l \left[ Y_i^j \mathbf{\hat{e}}_l^l \right] \right\}^0
\]

\[
\mathcal{W}(1,1,0,1;1,J') = \frac{1}{3} \delta_{J'}^r \quad \Rightarrow \quad \mathbf{\hat{r}} \times \mathbf{\hat{E}}_0 = -i\sqrt{8\pi} \left\{ E_i^l \left[ Y_i^j \mathbf{\hat{e}}_l^l \right] \right\}^0
\]

\[
\mathbf{\hat{r}} \times \mathbf{\hat{E}}_0 = -i\sqrt{8\pi} \sum_{\sigma} C_{-\sigma}^{1,10} E_{-\sigma}^l \mathbf{\hat{x}}_l^l = i \sqrt{8\pi} \sum_{\sigma} (-)^\sigma E_{-\sigma}^l \mathbf{\hat{x}}_l^l
\]
\[
\int d\hat{r} \hat{X}_M^{ij} \cdot (\hat{r} \times \hat{E}_0) \hat{Y}_0' (\hat{r}) = i\sqrt{\frac{8\pi}{3}} \sum_{\sigma} (-)^{\sigma} E_{-\sigma} \int d\hat{r} \hat{X}_M^{ij} \cdot \hat{X}_\sigma \hat{Y}_0' (\hat{r})
\]

\[
\hat{X}_\sigma \hat{Y}_0' (\hat{r}) = 3 \sqrt{\frac{i}{4\pi}} \sum_{LJ} C_0^{1J} \sum_{\sigma M} C_0^{1L} W(l_1, j_1, l_1, L_1) \hat{X}_M^{LJ}
\]

\[
= 3 \sqrt{\frac{i}{4\pi}} \sum_{\sigma M} C_0^{1J} \sum_{000} C_0^{1L} W(l_1, j_1, l_1, L_1) \hat{X}_M^{LJ}
\]

\[
\int d\hat{r} \hat{X}_M^{ij} \cdot (\hat{r} \times \hat{E}_0) \hat{Y}_0' (\hat{r})
\]

\[
= i \sqrt{\frac{8\pi}{3}} \sqrt{\frac{i}{4\pi}} \sum_{\sigma M} (-)^{\sigma} C_0^{1J} \sum_{000} C_0^{1L} W(l_1, j_1, l_1, L_1) E_{-\sigma} \int d\hat{r} \hat{X}_M^{ij} \cdot \hat{X}_\sigma
\]

\[
= i\sqrt{6i} (-)^{\nu} C_0^{1J} \sum_{000} C_0^{1L} W(l_1, j_1, l_1, L_1) E_{-M}
\]

Therefore, (1.1) implies

\[
(-)^{\nu} \sqrt{24\pi} \sum_{l} \delta_{l} (2l+1) j_{l} (\kappa a) C_0^{1J} \sum_{000} C_0^{1L} W(l_1, j_1, l_1, L_1) E_{-M}
\]

\[
+ Z \sum_{l} \left[ \frac{i}{k} \delta_{l} (2l+1) \left\{ \frac{\kappa}{2l+1} \left[ l h_{l+1} (\kappa a) - (l+1) h_{l-1} (\kappa a) \right] \right\} \right]
\]

\[
+ \frac{i}{\sqrt{2l+1}} b^{(M)} (\kappa a) \left[ \sqrt{l+1} \delta_{l} \delta_{l} + \sqrt{l} \delta_{l} \delta_{l} \right]
\]

\[
= Z \sum_{l} \left[ \frac{i}{k} \delta_{l} (2l+1) \left\{ \frac{k}{2l+1} \left[ l j_{l+1} (\kappa a) - (l+1) j_{l-1} (\kappa a) \right] \right\} \right]
\]

\[
+ \frac{i}{\sqrt{2l+1}} a^{(M)} (\kappa a) \left[ \sqrt{l+1} \delta_{l} \delta_{l} + \sqrt{l} \delta_{l} \delta_{l} \right]
\]

In the special case that \( J = L \), (1.2) becomes

\[
(-)^{\nu} \sqrt{24\pi} \sum_{l} \delta_{l} (2l+1) j_{l} (\kappa a) C_0^{1J} \sum_{000} C_0^{1L} W(l_1, j_1, l_1, L_1) E_{-M}
\]

\[
+ iZ \frac{b^{(E)} (\kappa a)}{2L+1} \left[ L h_{l+1} (\kappa a) - (L+1) h_{L-1} (\kappa a) \right]
\]

\[
= iZ \frac{a^{(E)} (\kappa a)}{2L+1} \left[ L j_{l+1} (\kappa a) - (L+1) j_{L-1} (\kappa a) \right]
\]

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II. Continuity of $H_t$

\[
\hat{r} \times \vec{H}_{mm} = \hat{r} \times \left( \hat{z} \times \vec{E}_0 \right) \left( \frac{\sqrt{4\pi}}{Z_2} \right) \sum_i {\sqrt{2l+1} j_i(\kappa r) Y_l^m(\hat{r})}
\]

\[
\hat{r} \times \left( \hat{z} \times \vec{E}_0 \right) = \hat{z} (\hat{r} \cdot \vec{E}_0) - \vec{E}_0 (\hat{r} \cdot \hat{z}) = E_0 \sin \theta \hat{z} \left( (\cos \phi \hat{x} + \sin \phi \hat{y}) \cdot (\cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y}) \right)
- E_0 (\cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y}) \cos \theta
\]

\[
= E_0 \sin \theta \left( \cos \phi \cos \alpha + e^{i\beta} \sin \phi \sin \alpha \right) \hat{z} - E_0 \cos \theta \left( \cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y} \right)
\]

\[
\therefore \hat{r} \times \vec{H}_{mm} = E_0 \left( \sin \theta \left( \cos \phi \cos \alpha + e^{i\beta} \sin \phi \sin \alpha \right) \hat{z} - \cos \theta \left( \cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y} \right) \right)
\]

\[
\left( \frac{\sqrt{4\pi}}{Z_2} \right) \sum_i {\sqrt{2l+1} j_i(\kappa r) Y_l^m(\hat{r})}
\]

I leave this expression in the above form for the present time. For the other terms, we have

\[
\hat{r} \times \vec{H}_{mm} = \sum_{l,m} \left\{ a_{lm}^{(E)} j_l(kr) \left( \hat{r} \times \vec{X}_m \right) - \frac{i}{k} a_{lm}^{(M)} \hat{r} \times \nabla \times \left[ j_l(kr) \vec{X}_m \right] \right\}
\]

\[
= \sum_{l,m} \left\{ a_{lm}^{(E)} j_l(kr) \left[ \frac{i}{\sqrt{2l+1}} \left[ \sqrt{l+1} \vec{X}_{m-1,l} + \sqrt{l} \vec{X}_{m+1,l} \right] \right] \right\}
\]

\[
\hat{r} \times \vec{H}_{mm} = \sum_{l,m} \left\{ b_{lm}^{(E)} h_l(kr) \hat{r} \times \vec{X}_m \right\}
\]

\[
= \sum_{l,m} \left\{ b_{lm}^{(E)} h_l(kr) \left[ \frac{i}{\sqrt{2l+1}} \left[ \sqrt{l+1} \vec{X}_{m-1,l} + \sqrt{l} \vec{X}_{m+1,l} \right] \right] \right\}
\]

Therefore
\[ E_0 \left\{ \sin \theta \left( \cos \phi \cos \alpha + e^{i\beta} \sin \phi \sin \alpha \right) \hat{z} - \cos \theta \left( \cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y} \right) \right\} \]

\[
\left( \frac{\sqrt{4\pi}}{Z_2} \right) \sum_l i^l \sqrt{2l+1} j_l (\kappa \alpha) Y_0^l (\hat{r})
\]

\[
\left[ \frac{b_{lm}^{(E)}}{h_{lm}^l (\kappa \alpha)} \frac{i}{\sqrt{2l+1}} \left[ \sqrt{l+1} \tilde{X}_{m+1}^{l+1} + \sqrt{l} \tilde{X}_{m-1}^{l+1} \right] \right] + \sum_{l,m} \left\{ \frac{-i}{k} b_{lm}^{(M)} \left[ \frac{\kappa}{2l+1} \left[ l h_{l+1}^l (\kappa \alpha) - (l+1) h_{l-1}^l (\kappa \alpha) \right] \tilde{X}_m^l \right] \right\}
\]

\[
\left[ \frac{a_{lm}^{(E)} j_l (\kappa \alpha)}{\sqrt{2l+1}} \left[ \sqrt{l+1} \tilde{X}_{m+1}^{l+1} + \sqrt{l} \tilde{X}_{m-1}^{l+1} \right] \right] + \sum_{l,m} \left\{ \frac{-i}{k} a_{lm}^{(M)} \left[ \frac{k}{2l+1} \left[ l j_{l+1} (\kappa \alpha) - (l+1) j_{l-1} (\kappa \alpha) \right] \tilde{X}_m^l \right] \right\}
\]

Now the trick is to do the angular projections of the source term with the least amount of labor. We have

\[
\sin \theta \left( \cos \phi \cos \alpha + e^{i\beta} \sin \phi \sin \alpha \right) \hat{z} - \cos \theta \left( \cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y} \right)
\]

\[
= \sqrt{\frac{2\pi}{3}} \left[ \cos \alpha (Y_{-1}^1 - Y_{1}^1) + ie^{i\beta} (Y_{-1}^1 + Y_{1}^1) \sin \alpha \right] \hat{z} - \sqrt{\frac{4\pi}{3}} Y_0^1 (\cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y})
\]

\[
= \sqrt{\frac{2\pi}{3}} \left[ \left( \cos \alpha + ie^{i\beta} \sin \alpha \right) Y_{-1}^1 + \left( -\cos \alpha + ie^{i\beta} \sin \alpha \right) Y_{1}^1 \right] \hat{e}_0^1
\]

\[
- \sqrt{\frac{2\pi}{3}} Y_0^1 \left( \cos \alpha \left( \hat{e}_{1}^1 - \hat{e}_{-1}^1 \right) + ie^{i\beta} \sin \alpha \left( \hat{e}_{1}^1 + \hat{e}_{-1}^1 \right) \right)
\]

\[
Y_{n+p}^1 \hat{e}_{n+p}^1 = \sum_l C_{n+p}^{11} L \tilde{X}_{n+p}^{1L} \Rightarrow \sin \theta \left( \cos \phi \cos \alpha + e^{i\beta} \sin \phi \sin \alpha \right) \hat{z} - \cos \theta \left( \cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y} \right)
\]

\[
= \sqrt{\frac{2\pi}{3}} \sum_l \left[ \left( \cos \alpha + ie^{i\beta} \sin \alpha \right) C_{-1-0}^{-1-0} L \tilde{X}_{-1}^{-1L} + \left( -\cos \alpha + ie^{i\beta} \sin \alpha \right) C_{1-0}^{1-0} L \tilde{X}_{1}^{1L} \right] - \cos \alpha \left( C_{0-1}^{01} L \tilde{X}_{0}^{0L} - C_{01}^{10} L \tilde{X}_{0}^{1L} \right) - ie^{i\beta} \sin \alpha \left( C_{0-1}^{01} L \tilde{X}_{0}^{0L} + C_{01}^{10} L \tilde{X}_{0}^{1L} \right)
\]

\[
= \sqrt{\frac{2\pi}{3}} \sum_l \left[ \left( \cos \alpha + ie^{i\beta} \sin \alpha \right) C_{-1-0}^{-1-0} L - \cos \alpha C_{0-1}^{01} L - ie^{i\beta} \sin \alpha C_{01}^{10} L \tilde{X}_{1}^{1L} \right] + \left[ \left( -\cos \alpha + ie^{i\beta} \sin \alpha \right) C_{1-0}^{1-0} L + \cos \alpha C_{0-1}^{01} L - ie^{i\beta} \sin \alpha C_{01}^{10} L \tilde{X}_{1}^{1L} \right]
\]

\[
C_{-1-0}^{-1-0} = (-)^L C_{0-1}^{11} L \Rightarrow
\]
\begin{align*}
\sin \theta (\cos \phi \cos \alpha + e^{i\beta} \sin \phi \sin \alpha) \hat{z} - \cos \theta (\cos \alpha \hat{x} + e^{i\beta} \sin \alpha \hat{y}) &= \sqrt{\frac{2\pi}{3}} \sum_{L} \left\{ C_{-10}^{lL} \left[ \left( \cos \alpha + i e^{i\beta} \sin \alpha \right) - (-)^L \cos \alpha - (-)^L i e^{i\beta} \sin \alpha \right] \tilde{X}_{-1}^{lL} \right\} + C_{10}^{lL} \left[ \left( \cos \alpha + i e^{i\beta} \sin \alpha \right) + (-)^L \cos \alpha - (-)^L i e^{i\beta} \sin \alpha \right] \tilde{X}_{1}^{lL} \\
&= \sqrt{\frac{2\pi}{3}} \sum_{L} \left\{ C_{-10}^{lL} \left[ 1 - (-)^L \right] \left( \cos \alpha + i e^{i\beta} \sin \alpha \right) \tilde{X}_{-1}^{lL} \right\} + C_{10}^{lL} \left[ 1 + (-)^L \right] \left( \cos \alpha + i e^{i\beta} \sin \alpha \right) \tilde{X}_{1}^{lL} \right\}
\end{align*}

Eq. (2.1) becomes

\begin{align*}
\frac{2\pi E_0}{Z_2} \sqrt{\frac{2}{3}} \sum_{L} \left( 1 - (-)^L \right) \left\{ C_{-10}^{lL} \left( \cos \alpha + i e^{i\beta} \sin \alpha \right) \tilde{X}_{-1}^{lL} \right\} + C_{10}^{lL} \left( \cos \alpha + i e^{i\beta} \sin \alpha \right) \tilde{X}_{1}^{lL} \right\} \sum_{l'} \sqrt{2l+1} j_{l'}(\kappa a) Y_{l'}(\hat{r}) \\
+ \sum_{t,m} \left\{ \frac{i}{k} b_{t}^{(l)} \left( \kappa a \right) \frac{1}{\sqrt{2l+1}} \left[ \sqrt{l+1} \tilde{X}_{m}^{l+1,l} + \sqrt{l} \tilde{X}_{m}^{l+1,l} \right] \right\} \\
= \sum_{t,m} \left\{ \frac{i}{k} a_{t}^{(l)} \left( \kappa a \right) \frac{1}{\sqrt{2l+1}} \left[ \sqrt{l+1} \tilde{X}_{m}^{l+1,l} + \sqrt{l} \tilde{X}_{m}^{l+1,l} \right] \right\}
\end{align*}

Replace $L \to L'$, $l \to l'$, $m \to m'$
\[
\frac{2\pi E_0}{Z_2} \sqrt{\frac{2}{3} \sum_{l'} \left( 1 - (-1)^{l'} \right)} \left\{ C_{-1,0}^{1,1} \left( \cos \alpha + ie^{i\theta} \sin \alpha \right) \hat{X}_{m}^{L',L} + C_{1,0}^{1,1} \left( -\cos \alpha + ie^{i\theta} \sin \alpha \right) \hat{X}_{m}^{L',L} \right\} \sum_{l} i^{l' + 1} f_{l} (\kappa a) Y_{l'} (\hat{r})
\]

\[
+ \sum_{l,m} \left\{ \frac{i b_{l,m}}{k^{(M)}} \left[ \frac{k}{2^{l' + 1}} \left[ l' h_{l+1}^{(M)} (\kappa a) - (l' + 1) h_{l}^{(M)} (\kappa a) \right] \hat{X}_{m}^{l'} \right] \right\}
\]

\[
= \sum_{l,m} \left\{ \frac{d_{l,m}}{k^{(M)}} \left[ \frac{k}{2^{l' + 1}} \left[ l' j_{l+1} (\kappa a) - (l' + 1) j_{l} (\kappa a) \right] \hat{X}_{m}^{l'} \right] \right\}
\]

Project out \( \hat{X}_{m}^{l',l} \). Again:

\[
\hat{X}_{m}^{l',l} \cdot \hat{X}_{\mu}^{L',L} = (-)^{l'-j} \frac{3(2l'+1)(2j+1)}{4\pi} \sum_{l,M} W (J,l,l',1;1,L) C_{000}^{nL} C_{nM \mu}^{jL'} Y_{M}^{L} \Rightarrow
\]

\[
\left( \hat{X}_{m}^{l',l} \right)^{*} \cdot \hat{X}_{\mu}^{L',L} = (-)^{l'+j+m} \hat{X}_{m}^{l',l} \cdot \hat{X}_{\mu}^{L',L}
\]

\[
= -(-)^{n} \frac{3(2l+1)(2j+1)}{4\pi} \sum_{l,M} (-)^{M} W (J,l,l',1;1,L) C_{000}^{nL} C_{nM \mu}^{jL'} \left( Y_{M}^{L} \right)^{*} \Rightarrow
\]

\[
\int d\Omega \left( \hat{X}_{m}^{l',l} \right)^{*} \cdot \hat{X}_{\mu}^{L',L} Y_{0}^{L} (\Omega)
\]

\[
= -(-)^{n} \frac{3(2l+1)(2j+1)}{4\pi} C_{000}^{nL} C_{nM \mu}^{jL'} W (J,l,l',1;1,l') \equiv Q (l,J,l',l';m) \delta_{m}
\]

So that
Therefore, for every \( J, l, m \), we have Eq. (2.3);

\[
\frac{2\pi E_0}{Z_2} \sqrt{2} \sum_{\ell, l'} \left( 1 - (-1)^{\ell} \right) i^{\ell} \sqrt{2l'+1} j_r(\kappa a) \left\{ C_{-10-1}^{L+1} \left( \cos \alpha + ie^{i\beta} \sin \alpha \right) Q(l, J, L', l'; -1) \delta_{\ell l'}^{m}
+ C_{10-1}^{L+1} \left( -\cos \alpha + ie^{i\beta} \sin \alpha \right) Q(l, J, L', l'; +1) \delta_{\ell l'}^{m} \right\}
\]

\[
+ \sum_{l', m'} \left\{ b_{l'm'}^{(E)} h_{l}^{(E)}(\kappa a) \frac{i}{\sqrt{2l'+1}} \left[ \sqrt{l+1} \delta_{l',l-1}^{m'} \delta_{l'}^{m} + \sqrt{l} \delta_{l',l}^{m} \delta_{l'}^{m} \right]
- \frac{i}{k} b_{l'm'}^{(M)} \delta_{l'}^{m} \delta_{l'}^{m} \left( \frac{\kappa}{2l'+1} \left[ l' h_{l'-1}^{(E)}(\kappa a) - (l'+1) h_{l'-1}^{(E)}(\kappa a) \right] \right) \right\}
\]

\[
= \sum_{l', m'} \left\{ a_{l'm'}^{(E)} j_r(\kappa a) \frac{i}{\sqrt{2l'+1}} \left[ \sqrt{l+1} \delta_{l',l-1}^{m'} \delta_{l}^{m} + \sqrt{l} \delta_{l',l}^{m} \delta_{l}^{m} \right]
- \frac{i}{k} a_{l'm'}^{(M)} \delta_{l}^{m} \delta_{l}^{m} \left[ l' j_{l'-1}(\kappa a) - (l'+1) j_{l'-1}(\kappa a) \right] \right\}
\]

In particular, for \( J = l \), we get Eq.(2.4).

\[
\frac{2\pi E_0}{Z_2} \sqrt{2} \sum_{\ell, l'} \left( 1 - (-1)^{\ell} \right) i^{\ell} \sqrt{2l'+1} j_r(\kappa a) \left\{ C_{-10-1}^{L} \left( \cos \alpha + ie^{i\beta} \sin \alpha \right) Q(l, J, L', l'; -1) \delta_{\ell l'}^{m}
+ C_{10-1}^{L} \left( -\cos \alpha + ie^{i\beta} \sin \alpha \right) Q(l, J, L', l'; +1) \delta_{\ell l'}^{m} \right\}
\]

\[
- \frac{i}{k} b_{l}^{(M)} \delta_{l}^{m} \delta_{l}^{m} \left[ l h_{l-1}(\kappa a) - (l+1) h_{l-1}(\kappa a) \right]
= - \frac{i}{k} a_{l}^{(M)} \delta_{l}^{m} \delta_{l}^{m} \left[ l j_{l-1}(\kappa a) - (l+1) j_{l-1}(\kappa a) \right]
\]
III. Continuity of $D_n$

$$\hat{r} \cdot \hat{X}_m^t = 0 \Rightarrow$$

$$\hat{r} \cdot \tilde{E}_{\text{inc}} = \sqrt{4\pi} (\hat{r} \cdot \tilde{E}_o) \sum_{l,m} i^l \sqrt{2l+1} j_l (kr) Y^t_l (\hat{r})$$

$$\hat{r} \cdot \tilde{E}_{\text{int}} = Z_i \sum_{l,m} i^l d^{(E)}_{l,m} \hat{r} \cdot \nabla \left[ j_l (kr) \tilde{X}_m^t \right], \quad \hat{r} \cdot \tilde{E}_{\text{sc}} = Z_2 \sum_{l,m} i^l b^{(E)}_{l,m} \hat{r} \cdot \nabla \left[ h_l^1 (\kappa r) \tilde{X}_m^t \right]$$

$$\hat{r} \cdot \nabla \left[ z_l (kr) \tilde{X}_m^t \right] = \frac{i}{r} \sqrt{l(l+1)} z_l (kr) Y^t_l (\hat{r}) \Rightarrow$$

$$\hat{r} \cdot \tilde{E}_{\text{int}} = -\frac{Z_i}{kr} \sum_{l,m} i^l d^{(E)}_{l,m} \sqrt{l(l+1)} j_l (kr) Y^t_l (\hat{r}), \quad \hat{r} \cdot \tilde{E}_{\text{sc}} = -\frac{Z_2}{\kappa r} \sum_{l,m} i^l b^{(E)}_{l,m} \sqrt{l(l+1)} h_l^1 (\kappa r) Y^t_l (\hat{r})$$

i.e.

$$\frac{Z_i}{ka} \sum_{l,m} a^{(E)}_{l,m} \sqrt{l(l+1)} j_l (ka) Y^t_l (\hat{r}) = \sqrt{4\pi} e_{1} (\hat{r} \cdot \tilde{E}_o) \sum_{l} i^l \sqrt{2l+1} j_l (\kappa a) Y^t_l (\hat{r})$$

$$\frac{Z_2}{\kappa a} \sum_{l,m} b^{(E)}_{l,m} \sqrt{l(l+1)} h_l^1 (\kappa a) Y^t_l (\hat{r})$$

Project out $Y^t_M$:

$$\frac{Z_i}{ka} a^{(E)}_{LM} \sqrt{L(L+1)} j_z (ka) = \sqrt{4\pi} e_{2} \sum_{l} i^l \sqrt{2l+1} j_l (\kappa a) \int d\hat{r} Y^t_M (\hat{r} \cdot \tilde{E}_o) Y^t_l (\hat{r})$$

$$\frac{Z_2}{\kappa a} b^{(E)}_{LM} \sqrt{L(L+1)} h_L^1 (\kappa a)$$

Now,

$$\int d\hat{r} (\hat{r} \cdot \tilde{E}_o) Y^t_M (\hat{r}) Y^t_l (\hat{r})$$

can be computed from:

$$Y^t_M (\hat{r}) Y^t_l (\hat{r}) = (-)^M \sum_{J} C_{-M,0-M}^{L,0-M} \left[ Y^t L | Y^t j \right] = (-)^M \sum_{J} C_{-M,0-M}^{L,0-M} \sqrt{\frac{\hat{t}}{4\pi J}} C_{000}^{L,0} Y^t_M$$

$$\int d\hat{r} (\hat{r} \cdot \tilde{E}_o) Y^t_M (\hat{r}) Y^t_l (\hat{r}) = (-)^M \sum_{J} C_{-M,0-M}^{L,0-M} \sqrt{\frac{\hat{t}}{4\pi J}} C_{000}^{L,0} \int d\hat{r} (\hat{r} \cdot \tilde{E}_o) Y^t_M$$

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The integral is given by
\[ \int d\hat{r} \left( \hat{r} \cdot \hat{E}_0 \right) Y_{M}^{J\l} = \sqrt{\frac{4\pi}{3}} (E_0)^{\l}_{-M} \delta^{J\l}_1 \] so that
\[ \int d\hat{r} \left( \hat{r} \cdot \hat{E}_0 \right) Y_{M}^{L\l} (\hat{r}) Y_{0}^{l} (\hat{r}) = (-)^{M} \sum_{J} C_{M0-M}^{L1-J} \sqrt{\frac{\mathcal{L}}{4\pi J}} C_{000}^{L1J} \sqrt{\frac{4\pi}{3}} (E_0)^{l}_{-M} \delta_{1}^{J} \]
\[ = (-)^{M} \frac{\mathcal{L}}{3} C_{M0-M}^{L1-J} C_{000}^{L1J} (E_0)^{l}_{-M} \]
This can be simplified slightly using \( C_{M0-M}^{L1-J} = (-)^{L-1} \frac{3}{L} C_{0M-M}^{L1L} \) to get
\[ \int d\hat{r} \left( \hat{r} \cdot \hat{E}_0 \right) Y_{M}^{L\l} (\hat{r}) Y_{0}^{l} (\hat{r}) = (-)^{M+L-1} \frac{\mathcal{L}}{3} C_{0M-M}^{L1L} C_{000}^{L1J} (E_0)^{l}_{-M} \]
So that
\[ \frac{-Z_{1} \epsilon_{1}}{k\alpha} a_{L,M}^{(E)} \sqrt{L(L+1)} j_{L}(k\alpha) = \sqrt{\frac{4\pi}{3}} \sum_{l} (-)^{L+M-1} (2L+1) C_{0M-M}^{L1L} C_{000}^{L1J} j_{l}(k\alpha) (E_0)^{l}_{-M} \] \hspace{1cm} (3.1)
\[ \frac{-Z_{2} \epsilon_{2}}{k\alpha} b_{L,M}^{(E)} \sqrt{L(L+1)} h_{L}(k\alpha) \]
Note that (3.1) and (1.3) together determine the electric multipoles.

IV. Continuity of \( B_{n} \)

Since the media are nonmagnetic \( (\mu_{j} = \mu_{0}) \) continuity of \( B_{n} \) is equivalent to continuity of \( H_{n} \)

\[ \hat{r} \cdot \hat{H}_{inc} = \sqrt{4\pi} \left( \frac{\hat{r} \cdot \hat{z} \times \hat{E}_0}{Z_{2}} \right) \sum_{l} i^{l} \sqrt{2l+1} j_{l}(k\alpha) Y_{l}^{l}(\hat{r}) \]
\[ \hat{r} \cdot \hat{H}_{int} = \frac{1}{kr} \sum_{l,m} a_{l,m}^{(M)} \sqrt{l(l+1)} j_{l}(k\alpha) Y_{l}^{l}(\hat{r}) \]
\[ \hat{r} \cdot \hat{H}_{sc} = \frac{1}{kr} \sum_{l,m} b_{l,m}^{(M)} \sqrt{l(l+1)} h_{l}^{l}(k\alpha) Y_{l}^{l}(\hat{r}) \]
\[
\frac{1}{\kappa a} a_{LM}^{(M)} \sqrt{L(L+1)} j_L(\kappa a) \\
= \frac{\sqrt{4\pi}}{Z_2} \sum_l i^{l+2l+1} j_L(\kappa a) \int d\hat{r} \left( \hat{r} \cdot \hat{z} \times \vec{E}_0 \right) Y_M^{L*}(\hat{r}) Y_0^L(\hat{r}) + \frac{1}{\kappa a} b_{LM}^{(M)} \sqrt{L(L+1)} h_L^0(\kappa a) 
\]  \hspace{1cm} (4.1)

Now

\[
\hat{r} \cdot \hat{z} \times \vec{E}_0 = -\hat{z} \cdot \hat{r} \times \vec{E}_0 = -i\sqrt{6} \left( \left[ \begin{array}{c} r^i E_1^1 \end{array} \right] \right)_0 = -i\sqrt{6} \sum_{m_1, m_2} C_{m_1 m_2 0}^{(1) \ 0} \left[ \begin{array}{c} r^1 E^1_1 \end{array} \right]_m \left( \hat{z} \cdot e_m^i \right) \\
= -i\sqrt{6} C_{0 \ 0 \ 0}^{(1) \ 0} \left[ r^1 E^1_1 \right]_0 = -i\sqrt{6} \left( -\frac{1}{\sqrt{3}} \right) \left[ r^1 E^1_1 \right]_0 = i\sqrt{2} \left[ \begin{array}{c} r^1 E^1_1 \end{array} \right]_0 = i\sqrt{2} \sqrt{\frac{4\pi}{3}} \left[ Y^1 E^1 \right]_0 \\
= i\sqrt{\frac{8\pi}{3}} \left[ Y^1 (\hat{r}) E^1 \right]_0
\]

\[
\int d\hat{r} \left( \hat{r} \cdot \hat{z} \times \vec{E}_0 \right) Y_M^{L*}(\hat{r}) Y_0^L(\hat{r}) \\
= (-)^M \sum_j \sqrt{\frac{L_i}{4\pi J}} C_{L_i}^{L M 0} C_{0000}^{J} \int d\hat{r} \left( \hat{r} \cdot \hat{z} \times \vec{E}_0 \right) Y_M^J(\hat{r}) \\
= (-)^M \sqrt{\frac{2}{3}} \sum_j \sqrt{\frac{L_i}{J}} C_{L_i}^{L M 0} C_{0000}^{J} \int d\hat{r} \left[ Y^J (\hat{r}) E^J \right]_0 Y_M^J(\hat{r}) \\
= (-)^{M+1} \sqrt{\frac{2}{3}} \sum_j \sqrt{\frac{L_i}{J}} C_{L_i}^{L M 0} C_{0000}^{J} \int d\hat{r} \left[ E^J Y^J (\hat{r}) \right]_0 Y_M^J(\hat{r})
\]
\[
\begin{align*}
&\left[ E^I Y^J (\hat{r}) \right]_0^M Y^J_M (\hat{r}) = \sum_{L'} C_{0-M-M}^{1} \left[ E^I Y^J (\hat{r}) \right] Y^{J'} (\hat{r}) \\
&= \sum_{J', L'} C_{0-M-M}^{1} \frac{3J}{4\pi J'} \sqrt{3J} W (1,1,L',J;1,J') \left[ E^I \left[ Y^{J'} \right] \right]_{-M}^{J'} \\
&= \sum_{J', L', m_1, m_2} C_{0-M-M}^{1} \frac{3J}{4\pi J'} \sqrt{3J} W (1,1,L',J;1,J') \sqrt{\frac{3J}{4\pi J'}} C_{m_1, m_2}^{1} \left[ E^I Y^{J'} \right]_{-M}^{J'} \\
&= 3 \sum_{J', L', m_1, m_2} C_{0-M-M}^{1} \frac{3J}{4\pi J'} \sqrt{3J} W (1,1,L',J;1,J') \sqrt{\frac{3J}{4\pi J'}} C_{m_1, m_2}^{1} \left[ E^I Y^{J'} \right]_{-M}^{J'} \\
\int d\hat{r} \left( \hat{r} \cdot \hat{z} \times \hat{E}_0 \right) Y^{J'}_M (\hat{r}) Y^J_0 (\hat{r}) \\
&= (-)^{M+1} i3 \sqrt{\frac{2}{3}} \sum_{J', L', m_1, m_2} \sqrt{\frac{L L'}{4\pi}} C_{-M-0-M}^{L} C_{0-M}^{L'} C_{0-0-M}^{L} C_{0-M-M}^{1} C_{m_1, m_2}^{1} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
&= 3i (-)^{M+1} \sqrt{\frac{2}{3}} \sum_{J', L', m_1} \sqrt{\frac{L L'}{4\pi}} C_{-M-0-M}^{L} C_{0-M}^{L'} C_{0-0-M}^{L} C_{m_1}^{1} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
&= 3i (-)^{M+1} \frac{2}{3} \sum_{J', L', m_1} \sqrt{\frac{L L'}{4\pi}} C_{-M-0-M}^{L} C_{0-M}^{L'} C_{0-0-M}^{L} C_{m_1}^{1} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
&= 3i (-)^{M+1} \frac{2}{3} \sum_{J', L', m_1} \sqrt{\frac{L L'}{4\pi}} C_{-M-0-M}^{L} C_{0-M}^{L'} C_{0-0-M}^{L} C_{m_1}^{1} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
&= \frac{1}{\sqrt{3}} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
\int d\hat{r} \left( \hat{r} \cdot \hat{z} \times \hat{E}_0 \right) Y^{J'}_M (\hat{r}) Y^J_0 (\hat{r}) \\
&= \frac{i}{3} (-)^{M} \sqrt{2} \frac{L L'}{4\pi} C_{-M-0-M}^{L} C_{0-M}^{L'} C_{m_1}^{1} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
&= \frac{i}{3} (-)^{M} \sqrt{2} \frac{L L'}{4\pi} C_{-M-0-M}^{L} C_{0-M}^{L'} C_{m_1}^{1} \left[ E^I \right]_{-M}^{J'} W (1,1,L',J;1,J') \int d\hat{r} Y^{J'}_{m_2} \\
\end{align*}
\]

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Now \( W(1,1,1,1,0) = W(1,1,1,1,0) = \frac{-1}{3} \Rightarrow \)

\[
\int d\hat{r} \left( \hat{r} \cdot \hat{z} \times \vec{E}_0 \right) Y_M^{l^*} (\hat{r}) Y_0^l (\hat{r}) = \frac{i}{9} (-)^{M+1} \sqrt{2L+1} \frac{C_{-M}^L C_{000}^{L+1} C_{-M}^1 C_{-M}^1 E_M^1}{C_{-M}^1 C_{+M}^1 C_{000}^{L+1} C_{-M}^1 C_{-M}^1 E_M^1} \]

So that finally

\[
\frac{1}{k\alpha} a_{LM}^{(M)} \sqrt{L(L+1)} j_L (k\alpha) = \frac{\sqrt{4\pi}}{9Z_2} \sum_l \rho_{2l+1} (-)^{M+1} (2l+1) \sqrt{2\ell + 1} j_{\ell} (k\alpha) C_{-M}^L C_{000}^{L+1} C_{-M}^1 C_{-M}^1 E_M^1 \]

\[
+ \frac{1}{k\alpha} b_{LM}^{(M)} \sqrt{L(L+1)} h_L^1 (k\alpha) \]

Equations (4.2) and (2.4) together determine the magnetic multipoles.

V. Dipole Moment

\[
\hat{P} = [\varepsilon (\vec{x}) - \varepsilon_0] \vec{E} (\vec{x}) \Rightarrow \rho_{pol} = -\nabla \cdot \hat{P} = -\nabla \left[ \varepsilon (\vec{x}) - \varepsilon_0 \right] \vec{E} (\vec{x})
\]

\[
= -\nabla \left[ \varepsilon (\vec{x}) - \varepsilon_0 \right] \cdot \vec{E} (\vec{x}) - \left[ \varepsilon (\vec{x}) - \varepsilon_0 \right] \nabla \cdot \vec{E} (\vec{x})
\]

The dielectric functions are spatially constant in each region and there is no free electric charge, so \( \nabla \cdot \vec{E} (\vec{x}) = 0 \).

\[
\rho_{pol} = -\nabla \cdot \hat{P} = -\nabla \cdot \left[ \varepsilon (\vec{x}) - \varepsilon_0 \right] \vec{E} (\vec{x}) = -\nabla \left[ \varepsilon (\vec{x}) - \varepsilon_0 \right] \cdot \vec{E} (\vec{x})
\]

\[
\varepsilon (\vec{x}) = \varepsilon_1 \Theta (a - r) + \varepsilon_2 \Theta (r - a) \Rightarrow \nabla \left[ \varepsilon (\vec{x}) - \varepsilon_0 \right] = (\varepsilon_2 - \varepsilon_1) \delta (r - a) \hat{r}
\]

\[
\rho_{pol} = - (\varepsilon_2 - \varepsilon_1) \delta (r - a) \hat{r} \cdot \vec{E} (\vec{x})
\]

\[
\vec{d} = \int d^3 x \rho_{pol} \hat{x} = - (\varepsilon_2 - \varepsilon_1) \int d^3 x \hat{r} \cdot \vec{E} (\vec{x}) \delta (r - a) \hat{r}
\]

\[
= -(\varepsilon_2 - \varepsilon_1) a^3 \left( \hat{r} \cdot \vec{E} (\vec{r}) \right) \hat{r}
\]

\[
\hat{r} \cdot \vec{E}_{int} (\vec{r}) = \frac{-Z_1}{k\alpha} \sum_{l,m} a_{LM}^{(E)} \sqrt{l(l+1)} j_l (kr) Y_m^l (\hat{r}) \quad \text{(see discussion in section III above).}
\]
\[
\mathbf{d} = (\varepsilon - \varepsilon_l) Z_i a^2 \sum \left| a_{iM}^{(E)} \right|^2 j_i(k a) \int d\hat{r} Y_m^l(\hat{r}) \hat{r}
\]

\[
\int d\hat{r} Y_m^l(\hat{r}) \hat{r} = \int d\hat{r} (-)^m Y_m^l(\hat{r}) \hat{r} = (-)^{m+1} \sqrt{4\pi} \int d\hat{r} Y_m^l(\hat{r}) \hat{X}_m^0
\]

\[
= (-)^{m+1} \sqrt{4\pi} \sum M C_{-M M 0}^{-1 0} \hat{X}_M^0 \int d\hat{r} Y_m^l(\hat{r}) Y_m^l(\hat{r}) = (-)^{m+1} \sqrt{4\pi} C_{-M M 0}^{-1 0} \hat{X}_M^0 \delta_l^l = \sqrt{\frac{4\pi}{3}} \hat{x}_M^0 \delta_l^l
\]

\[
\therefore \mathbf{d} = (\varepsilon - \varepsilon_l) \sqrt{\frac{8\pi}{3}} Z_i a^2 j_i(k a) \sum M a_{iM}^{(E)} \hat{x}_M^0
\]

(5.1)

\[
|\mathbf{d}| = \left| \sqrt{\frac{8\pi}{3}} Z_i a^2 j_i(k a) \sum M a_{iM}^{(E)} \hat{x}_M^0 \right| (\varepsilon - \varepsilon_l)
\]

Example:

We can solve (1.3) and (3.1) with \(L = 1\) to get:

\[
(-)^3 \sqrt{24\pi} E_M^{(E)} \left\{ i j_0(\kappa a) C_{0 M M 0}^{0 1 11} C_{0 0 0 0}^{0 1 11} W(0,1,1,1,1) + \right\}
\]

\[
+ \sqrt{\frac{4\pi}{3}} \varepsilon (E_0)_{-M}^{(E)} \left\{ (-)^M C_{0 M M 0}^{0 1 11} C_{0 0 0 0}^{0 1 11} j_0(\kappa a) + \right\}
\]

\[
\therefore \frac{Z_2 \varepsilon}{\kappa a} \sqrt{2} \hat{h}_l^1(\kappa a) = \frac{-Z_2 \varepsilon}{\kappa a} a_{lM}^{(E)} \sqrt{2} j_i(k a)
\]

Now,

\[
C_{0 M M 0}^{0 1 11} = C_{0 0 0 0}^{0 1 11} = 1, W(0,1,1,1,1,1) = 1/3
\]

\[
C_{0 M M 0}^{2 1 11} = \frac{1}{\sqrt{10}}, C_{0 0 0 0}^{2 1 11} = -\frac{\sqrt{2}}{5}, W(2,1,1,1,1,1) = 1/3
\]

So that

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\(-\sqrt{24\pi} \left\{ j_0 (\kappa a) + j_2 (\kappa a) \right\} E_{-M}^i + Z_2 \left[ h_2^i (\kappa a) - 2 h_0^i (\kappa a) \right] b_{iM}^{(E)} = Z_1 \left[ j_2 (\kappa a) - 2 j_0 (\kappa a) \right] a_{iM}^{(E)} \) 

and

\[
\sqrt{\frac{2\pi}{3}} \varepsilon_2 (E_0)_{-M} \left\{ j_0 (\kappa a) + j_2 (\kappa a) \right\} + \frac{Z_2 \varepsilon_2}{\kappa a} b_{iM}^{(E)} h_1^i (\kappa a) = \frac{Z_1 \varepsilon_1}{\kappa a} a_{iM}^{(E)} j_1 (\kappa a)
\]

The solutions of these equations in the long wavelength limit are:

\[
a_{i,m} = \frac{3 \varepsilon_2 \sqrt{6\pi E}}{Z_1 (\varepsilon_1 + 2 \varepsilon_2)} + \frac{1}{10} \frac{\varepsilon_2 \sqrt{6\pi E} \left( 17 \varepsilon_1 \kappa^2 - 26 \varepsilon_2 \kappa^2 + 3 \varepsilon_1 k^2 + 12 \varepsilon_2 k^2 \right)}{Z_1 (\varepsilon_1 + 2 \varepsilon_2)^2} + O(a^3)
\]

And

\[
b_{i,m} = \frac{2 i \kappa^2 \sqrt{6\pi E} (\varepsilon_1 - \varepsilon_2)}{Z_2 (\varepsilon_1 + 2 \varepsilon_2)} a^3 + O(a^3)
\]

Where \( E \) is \( (E_0)_{-M} \) in these equations. Therefore:

\[
\sqrt{\sum_M^M \left| a_{iM}^{(E)} \right|^2} = \frac{3 \varepsilon_2 \sqrt{6\pi}}{Z_1 (\varepsilon_1 + 2 \varepsilon_2)} \sqrt{\sum_M^M \left| (E_0)_{-M} \right|^2} = \frac{3 \varepsilon_2 \sqrt{6\pi E_0}}{Z_1 (\varepsilon_1 + 2 \varepsilon_2)}
\]

So that

\[
|d| = 4 \pi \varepsilon_2 \frac{|\varepsilon_2 - \varepsilon_1| a^3}{(\varepsilon_1 + 2 \varepsilon_2)} E_0.
\]
VI. Cross-Sections

\[ |\hat{\mathbf{\varepsilon}}_p \cdot \hat{\mathbf{E}}_{sc}|^2 = \left| \sum_{l,m} \hat{\mathbf{\varepsilon}}_p \cdot Z_2 \left( i \frac{h_{lm}^{(E)}}{\kappa} \nabla \times [ h_l^{(i)}(\kappa r) \hat{\mathbf{X}}_m + b_{lm}^{(M)} h_l^{(i)}(\kappa r) \hat{\mathbf{X}}_m ] \right) \right|^2 = |Z_2|^2 \left| \sum_{l,m} \hat{\mathbf{\varepsilon}}_p \cdot i h_{lm}^{(E)} \left( -2L \frac{l+1}{2l+1} h_{l+1}^{(i)}(\kappa r) \hat{\mathbf{X}}_{m+1}^{l+1} + \frac{l+1}{2l+1} h_{l-1}^{(i)}(\kappa r) \hat{\mathbf{X}}_{m-1}^{l+1} \right) + b_{lm}^{(M)} h_l^{(i)}(\kappa r) \hat{\mathbf{X}}_m \right|^2 \]

\begin{align*}
\lim_{r \to \infty} |\hat{\mathbf{\varepsilon}}_p \cdot \hat{\mathbf{E}}_{sc}|^2 = & \frac{|Z_2|^2}{\kappa^2} \left| \sum_{l,m} \hat{\mathbf{\varepsilon}}_p \cdot \left( b_{lm}^{(E)} \left( \frac{l+1}{2l+1} \hat{\mathbf{X}}_m^{l+1} - \frac{l}{2l+1} \hat{\mathbf{X}}_m^{l+1} \right) - b_{lm}^{(M)} \hat{\mathbf{X}}_m \right) \right|^2 \end{align*}

So the differential scattering cross section is:

\[ \frac{d\sigma}{d\Omega}(\theta, \phi, \hat{\mathbf{\varepsilon}}_p) = \frac{|Z_2|^2}{\kappa^2 |E_0|^2} \left| \sum_{l,m} \hat{\mathbf{\varepsilon}}_p \cdot \left( b_{lm}^{(E)} \left( \frac{l+1}{2l+1} \hat{\mathbf{X}}_m^{l+1} - \frac{l}{2l+1} \hat{\mathbf{X}}_m^{l+1} \right) - b_{lm}^{(M)} \hat{\mathbf{X}}_m \right) \right|^2 \]

For example, in the long wavelength limit,

\[ \frac{d\sigma}{d\Omega}(\theta, \phi, \hat{\mathbf{\varepsilon}}_p) = \frac{|Z_2|^2}{\kappa^2 |E_0|^2} \left| \sum_{m} \hat{\mathbf{\varepsilon}}_p \cdot b_{lm}^{(E)} \left( \frac{2}{3} \hat{\mathbf{X}}_m^{0,1} - \frac{1}{3} \hat{\mathbf{X}}_m^{2,1} \right) \right|^2 \]

Now:

\[ \hat{\mathbf{\varepsilon}}_p \cdot \hat{\mathbf{X}}_m^{l+1} = \left[ Y^l \left( \hat{\mathbf{\varepsilon}}_p \right) \right]_M^l \] so that the cross-section for incident polarized radiation is:

\[ \frac{d\sigma}{d\Omega}(\theta, \phi, \hat{\mathbf{\varepsilon}}_p) = \frac{|Z_2|^2}{\kappa^2 |E_0|^2} \left| \sum_{m} b_{lm}^{(E)} \left[ \frac{2}{3} \left[ Y^0 \left( \hat{\mathbf{\varepsilon}}_p \right) \right]_M^0 - \frac{1}{3} \left[ Y^2 \left( \hat{\mathbf{\varepsilon}}_p \right) \right]_M^2 \right] \right|^2 \]

(6.1)

For unpolarized incident radiation, we must average (6.1) over \( \alpha \) and \( \beta \).

For example, in the long wavelength limit,
\[ b_{l,1}^{(E)}(E_0) = B(E_0) = \frac{BE_0}{\sqrt{2}}(\cos \alpha - ie^j \sin \alpha), \quad b_{l,1}^{(E)} = B'(E_0) = -\frac{BE_0}{\sqrt{2}}(\cos \alpha + ie^j \sin \alpha) \]

where \( B = \frac{2i(\kappa a)^3 \sqrt{6\pi}}{3Z_2} \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} \)

\[
\langle |b_{l,m}^{(E)}|^2 \rangle = \frac{|BE_0|^2}{2} \left( 1 - i(e^{i\beta} - e^{-i\beta}) \cos \alpha \sin \alpha \right) = \frac{|BE_0|^2}{2}
\]

\[
\langle b_{l,1}^{(E)} b_{l,1}^{(E)} \rangle = \frac{|BE_0|^2}{2} \left( \cos^2 \alpha - \sin^2 \alpha + i(e^{i\beta} + e^{-i\beta}) \cos \alpha \sin \alpha \right) = 0
\]

So that the cross-section for incident unpolarized radiation is;

\[
\frac{d\sigma}{d\Omega} = \frac{Z_2 B^2}{6\kappa^2} \sum_m \sqrt{2} \left[ Y^0 \left( \hat{\varepsilon}_m \right)^* \right] \left[ \left( \hat{\varepsilon}_m \right)^* \right] \left[ Y^2 \left( \hat{\varepsilon}_m \right)^* \right] \left[ \left( \hat{\varepsilon}_m \right)^* \right]
\]

The polarization vectors are (see Jackson section 10.1)

\[
\hat{\varepsilon}_\parallel = \cos \theta \cos \phi \hat{x} + \cos \theta \sin \phi \hat{y} - \sin \theta \hat{z} = \hat{\Theta}, \quad \hat{\varepsilon}_\perp = -\sin \phi \hat{x} + \cos \phi \hat{y} = \hat{\Phi}
\]


M. Bayer and A. Forchel, (2002) Temperature dependence of the exciton homogeneous linewidth in In_{0.60}Ga_{0.40}As/GaAs self-assembled quantum dots. Physical Review B 65, 041308(R).


