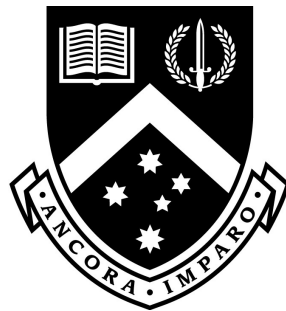


Nanoparticle Based Photoacoustic Treatment of Cancer

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BSc Eng (Hons)

Submitted in total fulfilment of the requirements for the degree
of
Doctor of Philosophy



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Summary

Cancer afflicts a compelling portion of the worldwide population with exceedingly high figures of morbidity and mortality. In fact, cancer causes more than 8 million deaths and seizes 14 million new victims each year. For decades, scientists have tirelessly attempted to devise methods at battling this deadly malady with little success. Even the state-of-the-art treatments are tainted with myriads of pernicious side-effects that debilitate the quality of life of patients. A conspicuous requirement exists for researchers to get involved in the formulation of novel therapeutic measures to battle this calamitous ailment. Modalities such as *cellular hyperthermia*, which are conceived based upon the recent coalescence of nanotechnology and medicine, shed hope on a cancer averted future. Nanoparticle based cellular hyperthermia utilizes nanoparticles to deliver controlled thermal energy to selectively destroy cancer cells while sparing adjacent healthy tissue. Achieving this requires the development of a complete nanophotonic device with controlled emission characteristics.

In fact, there has been a keen interest in utilizing nanoscopic structures in cancer therapy for several years. The minute nature of these devices theoretically enables minimally-invasive treatment of cancers. The design of a complete nanoscopic photothermal cancer therapy device requires the coalescence of multiple domains of knowledge. Furthermore, the end goal of such a device should be the generation of a controlled thermal pulse of energy. This is due to the fact that rather than using conventional energy sources, a pulse of thermal energy

has a significant potential towards improving cancer hyperthermia by not only increasing the internal tumour temperature to sufficient levels, but also by sparing the thermal damage to the surrounding healthy tissues significantly.

The proposed technique in this thesis to achieve this task of generating a pulse of thermal energy is via the utilization of the quantum mechanical principle of superradiance. In order to generate a superradiant thermal pulse, a highly symmetric and coupled group of nanoparticles is designed to depict superradiant characteristics. The conceptualization is based on a quantum mechanical and quantum electrodynamical framework and the emission dynamics are obtained based on coupled mode theory. Photon statistics are performed to obtain the amount of energy generated from a pulse. These results are validated using tissue model simulations on both breast cancer and liver cancer tumours. The obtained results indicate that through the utilisation of a thermal superradiant pulse, delivering a sufficient amount of thermal energy to the tumour volume while maintaining temperatures of adjacent healthy tissues at safe levels is indeed possible.

This thesis is organized as follows: an introduction to superradiance, cancer hyperthermia and motivation are provided in Chapter 1, which is followed by a fully quantum mechanical discussion of superradiant emission, both in general and in the context of thermal emission, in Chapter 2. A discussion on the study of multi-nanoparticle systems is presented in Chapter 3, followed by the details of generating a superradiant thermal pulse and its emission characteristics, which are discussed in Chapters 4 and 5. The potential enhancement to cancer hyperthermia through the utilization of a superradiant thermal pulse is presented in Chapter 6. Finally, a summary of contributions and potential future work is detailed in Chapter 7.

General Declaration

This thesis contains no material which has been accepted for the award of any other degree or diploma at any university or equivalent institution and that, to the best of my knowledge and belief, this thesis contains no material previously published or written by another person, except where due reference is made in the text of the thesis.

Signature : _____

Name : Sudaraka Mallawaarachchi

Date : 26th September 2019

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*This thesis is dedicated to my forever supportive parents, eternally
mischievous sister and breathtakingly remarkable wife.*

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Contents

1	Introduction	1
1.1	Superradiance	1
1.1.1	Current developments involving superradiance	2
1.1.2	Superradiance and energy conservation	3
1.2	Nanoparticles in Recent Research	4
1.2.1	Some properties of nanoparticles	4
1.2.2	Nanoparticles in biomedical applications	5
1.3	Motivation	7
1.4	Research Aims and Thesis Scope	8
1.4.1	Analysis of superradiance in nanoparticles	8
1.4.2	Design a high particle density superradiant structure	8
1.4.3	Determine the optimal design criteria for the assembly	9
1.4.4	Thermal requirement analysis for cancer hyperthermia	9
1.4.5	Simulation and verification	10
2	Superradiance and Emission of Thermal Energy	11
2.1	Overview	11
2.2	Case Study: Emission from N interacting QDs	12
2.3	The Superradiant Master Equation	14
2.3.1	Analysis of the real component	17
2.3.2	Analysis of the imaginary component	18
2.4	Emission Rate Characterization	18
2.5	Superradiant Criteria	19
2.5.1	Size of the assembly	20
2.5.2	Symmetry of the emitters	20
2.6	Steady State Superradiance	22
2.7	Extendibility to the Thermal Domain	24
3	Analysis of Multi-Nanoparticle Systems	27
3.1	Overview	27
3.2	Fundamental Approaches	28
3.2.1	Born-Oppenheimer approximation	29

3.2.2	Density functional theory	30
3.2.3	Mie's theory of scattering	31
3.3	Numerical Simulation Techniques	32
3.3.1	Computational electromagnetics techniques	33
3.4	Coupled Mode Theory	35
3.5	Inter Particle Interactions	37
3.5.1	Polarizability	38
3.5.2	Self-energy of a dipole	38
3.5.3	Static inter-dipole interactions	39
3.5.4	Keesom interactions	39
3.5.5	Van der Waals interactions	40
4	Controlled Generation of a Thermal Superradiant Pulse	43
4.1	Mathematical Characterization	43
4.1.1	Free space model	44
4.1.2	Inter-emitter Couplings	49
4.2	Derivation of Emission Cross Sections (ECS)	50
4.2.1	ECS - semi-generalized	51
4.2.2	ECS - conventional	57
4.2.3	ECS - fully generalized	58
4.2.4	Analytical validation of derivations	67
4.3	Tuning the Resonance Frequency	68
5	Characterisation of the Generated Thermal Superradiance Pulse	73
5.1	PEC Simulations for Thermal Superradiance	73
5.1.1	Simulation details	73
5.1.2	Semi generalized model	74
5.1.3	Fully generalized model	77
5.2	Ring Arrangement of Quantum Dots	83
5.2.1	Simulation parameters	84
5.2.2	Emission characteristics	84
5.3	Buckyball Arrangement of Quantum Dots	84
5.3.1	Superradiant controllability	90
5.3.2	Simulation parameters	92
5.3.3	Emission characteristics	93
6	Superradiance as a Viable Method for Biomedical Applications	97
6.1	Overview of Nanomedicine	97
6.1.1	Enhanced permeability and retention effect	98
6.1.2	Drug delivery	99
6.1.3	Imaging of tumours	99
6.1.4	Nanomedicine and cancer therapy	100
6.2	Optimally Tuning the Superradiant Pulse	103

6.3	Tissue Model Simulations	105
6.3.1	QD ring assembly	105
6.3.2	QD buckyball assembly	107
7	Summary of Contributions and Suggestions for Future Work	111
7.1	Summary of Contributions	111
7.2	Recommendations for Future Research	114
	Bibliography	117

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List of Figures

2.1	The Dicke Ladder	12
2.2	Ideal Characteristics of a Superradiance Pulse	17
2.3	Nearest Neighbour Interactions	21
2.4	Non-resonant Emitters	24
2.5	Conventional vs. Thermal Superradiance	25
3.1	Operation Parameters of an Emitter	36
4.1	Free Space Model	45
4.2	Cylindrical PEC Assembly Design	51
4.3	2 Emitter PEC Assembly Design	54
4.4	Spherical PEC Assembly Design	59
4.5	Resonance Frequency	69
4.6	Cavity Based Resonance Tuning	71
5.1	Spectral Characterisation for the Semi Generalized Model	75
5.2	Spectral Characterisation for the Fully Generalized Model	78
5.3	Spectral Characterisation with Startling Phenomena	80
5.4	Quantum Dot Ring Arrangement	83
5.5	Pulse Characterisation from the QD Ring Assembly	85
5.6	Spectral Characteristics of the QD Ring Assembly	86
5.7	Buckyball QD Arrangement	87
5.8	Nearest Neighbour Map	89
5.9	Permanent, Induced and Resultant Dipoles	90
5.10	Spectral Characterisation of the Buckyball Model	92
5.11	Pulse Characterisation of the Buckyball Model	95
6.1	Potential Resonant Frequencies in the Cavity Model	104
6.2	Liver Tissue Model	106
6.3	Breast Tissue Model	108

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Chapter 1

Introduction

This chapter provides a brief introduction to the concept of superradiance, its characteristics and recent developments, along with an overview of nanoparticles and their utilization in biomedical applications. Research motivation, aim and the thesis scope are also presented.

1.1 Superradiance

Superradiance is a signature effect in quantum photonics that explains the collective enhancement of emission power, approximately by a factor of N^2 , when N quantum emitters are placed in subwavelength proximity with adherence to certain symmetry criteria. The nature of this effect is apparently counter-intuitive, given the fact that N quantum emitters should emit at a collective emission power, approximately scaled by a factor of N , instead of N^2 . However, in nanoscopic dimensions, light fails to distinguish between quantum emitters and thus, interacts with the entire assembly collectively. This is one of the main reasons bolstering this unexpected behaviour. The effect itself is inherently transient and ephemeral, although successful attempts have been made recently to sustain it in the steady-state regime.

Superradiance was first conceptualized by Robert H. Dicke in 1954 [1]. It was originally proposed to explain the ephemeral, enhanced and cooperative sponta-

neous emission characteristics depicted by excited gases, where a flash of bright light is observed when the ionized gas particles are coherently coupled. Since then, the theory has undergone rigorous mathematical and experimental studies. Recently, based on time reversal symmetry, the concept of superradiance has been proposed to be extended to its absorption counterpart: *superabsorption* [2].

1.1.1 Current developments involving superradiance

Over the recent years, a considerable effort has been rendered towards mathematically understanding, observing and utilizing the superradiant effect. The mathematical aspects of the effect are presented in detail in chapter 2.

Observation of superradiance has been a topic of interest for many years. The effect has been observed in optically pumped HF gas (1973) [3], in a pair of trapped ions (1996) [4] and in a ring cavity (2007) [5], to name a few of many instances. More recent observations include Bose-Einstein Condensates [6], Mott insulators [7] and colour centres in diamond [8].

One major achievement in terms of utilizing superradiance is the invention of the superradiant laser. The concept was first proposed in 1993 by Haake *et al.* [9] and it was several years in development, until the first demonstration, which took place in 2012 [10]. The speciality of the concept is that, unlike a conventional laser, a superradiant laser does not rely on a large number of photons to be present within the lasing cavity to maintain coherence. In fact, the 2012 design has less than one intracavity photon.

The concept was further extended into a cold-strontium laser that operates in the superradiant crossover regime by the same research group [11]. This laser is demonstrated to circumvent the linewidth limitations of conventional lasers by utilizing the dipole-forbidden transitions. Their design relies upon the coherence of the atoms in the superradiant regime rather than the coherence of the light

field, which is the typical operating mechanism of a conventional laser [12].

Utilization of the superradiant effect for the development of semiconductor devices is another area of significant research interest [13, 14]. While the concept was originally proposed to enhance semiconductor lasers, proposals for designing components such as diodes and transistors [15, 16] have been around for a long time. The idea has also been extended to the development of quantum biological switches [17].

1.1.2 Superradiance and energy conservation

Superradiance enhances the power emission by a factor that approximately scales quadratically with the number of emitters involved in the emission process. This abnormal power scaling should technically violate the principle of energy conservation.

However, it is noted that the enhancing effect is only transient. For the case of a singular pulse, a ‘bright flash’ of energy is noticed for a very short duration and the emitter assembly ceases from emitting any further energy until $t \rightarrow \infty$. In fact, if a comparison is made between an ideal superradiant and non-superradiant emitter assembly, both assemblies identical and starting the emission process at $t = 0$, at the observation point ($t \rightarrow \infty$), the non-superradiant assembly would be releasing more energy than its superradiant counterpart. However, due to the pulsation effect, if an observation is made at a time point soon after $t = 0$, the total energy emitted from the superradiant assembly will be much higher than its non-superradiant counterpart.

The comparative reduction in the emitted energy observed at $t \rightarrow \infty$ by the superradiant assembly is due to the fact that a certain amount of energy in the assembly is consumed by the interactions that occur between the emitters. Therefore, the effect of superradiance does not violate the laws of energy conservation.

1.2 Nanoparticles in Recent Research

With the commencement of the 'nano-revolution', a considerable research interest has been dedicated towards understanding the behaviour and the utilization of nanoparticles in various applications.

Due to the extremely minute nature of these particles, the laws of physics that govern macroscopic bodies fail to describe the behaviour of these particles. Since the size of these particles is comparable with the wavelengths of light that interact with them, it has become possible to manipulate light in unforeseen mechanisms. Furthermore, nanoparticles can be grown in different sizes, shapes and materials. This diversification results in a plethora of interesting behaviours and characteristics [18].

1.2.1 Some properties of nanoparticles

Nanoparticles are typically considered to range in dimension from 1nm to 100nm. In these nanoscopic dimensions, materials tend to depict unprecedented characteristics. For instance, copper, which is an opaque material in bulk converts to a transparent material in nanoscopic dimensions. These properties are mostly attributed to the confinement of electrons within nanoparticles.

Electrons within bulk material can travel relatively long distances between successive impacts, however, in the case of nanoparticles, due to the nanoscopic dimensions, the electron mean free path is significantly reduced. This leads to an effect known as electron surface scattering, which is not prominent in bulk materials. This confinement completely changes the optical properties of nanoparticles and characteristics such as permittivity become dependent on the size and shape of the nanoparticle. These characteristics obviously make it difficult to analytically model and predict the behaviour of nanoparticles, however, it also becomes possible to tune these nanoparticles by varying shape and size criteria

to make them suitable for myriads of applications.

In metallic nanoparticles, collective oscillations of the conduction band electrons are observed when coupled with an external electromagnetic field. These well known oscillations are referred to as localized surface plasmons and are at the heart of the area of research known as 'plasmonics'. This field of research is currently at the verge of developing the nanoscopic counterpart of the convention LASER, referred to as the SPASER [19]. These localised surface plasmons are also used to generate strong electric fields within nanoparticles, which are then known to generate 'hot electrons', a conceptual phenomenon with many applications in optics and physics [20].

Furthermore, due to the strong inter particle coupling forces present in these nanoscopic dimensions, the inter particle energy transfer processes are significantly different from those observed in bulk materials. In fact, the well-known resonance energy transfer principle governs the energy transfer at these scales and is used to explain the fundamentals of processes such as photosynthesis [21].

1.2.2 Nanoparticles in biomedical applications

Due to the minute nature of nanoparticles, they possess the ability to penetrate blood vessels and directly reach into tissues and cells of in-vivo systems. This effect is of particular interest to biomedical applications. For instance, with the aid of chemical aptamers, it is possible to use nanoparticles to precisely target specific organs or tumours. This ability is used to greatly enhance targeted drug delivery and more recently, cancer therapy [22,23].

Spherical gold nanoparticles are the most utilized type of particle for biomedical applications. However, recent works have proposed the use of gold nanorods, nanoshells and other shapes due to their inherent and favourable optical controllability characteristics. Since nanoparticles can be grown using different types

of organic and inorganic materials, designing biocompatible particles should be possible [24]. More recently, the utilization of quantum dots has also been suggested for biomedical applications such as cancer therapy [25,26].

Besides gold nanoparticles, materials such as Silver, Copper, Platinum and Graphene have also been suggested for biomedical applications. These biomedical applications utilizing nanoparticles include a wide range of techniques such as photothermal cancer therapy, radio frequency induced hyperthermia, magnetic hyperthermia, ultrasound induced hyperthermia, sonodynamic hyperthermia and radiotherapy. Although each treatment modality is unique, the concept remains the same. Nanoparticles are directed to the tumour site and once gathered, are excited externally using an energy source. The variability of the type of energy source is what gives rise to a multitude of treatment modalities. Currently, besides significant analytical and numerical computer simulations, some of these techniques have undergone both in-vitro and in-vivo studies successfully. However, human trials are still several years away from taking place [27].

Biocompatibility

Biocompatibility of nanoparticles remains an open topic with myriads of publications appearing each year. Currently, there are nanoparticles that are being utilized for drug delivering applications that are classified as biocompatible. Biocompatibility itself is a complicated topic that requires stringent study where one has to consider factors such as immunocompatibility, blood interactions and biodegradability. Typically, carbon based nanoparticles, particularly fullerenes, are continuously being studied for biocompatibility. In certain instances, carbon based nanoparticles have shown to cause inflammation and granuloma formation whereas other research claims that these particles are completely biocompatible [28].

Quantum dots are a type of nanoparticles that depict various and highly at-

tractive optical and thermal properties. In their raw form, these nanoparticles induce cytotoxic effects, specifically to the mitochondria, cellular membranes and plasma. However, properly coated quantum dots have demonstrated biocompatibility in experimental scenarios [28].

1.3 Motivation

Cancer is a prominent cause of distress due to its attribution with extremely high figures of morbidity and mortality. Cancer is responsible for more than 8 million tragedies each year while capturing more than 14 million new victims along the way. For many years, scientists have tried battling cancer with low rates of success. In fact, even the most advanced treatments are accompanied with many side effects that degrade the quality of life.

Recently, with the advent of nanomedicine, there has been a keen interest in utilizing nanoscopic structures in cancer therapy. The minute nature of these devices theoretically enables minimally-invasive treatment of cancer. The modality known as ‘cellular hyperthermia’, which utilizes nanoparticles to deliver thermal energy to destroy cancer cells while sparing adjacent healthy tissues, is of particular significance. However, designing a proper nanoscopic assembly of thermal emitters to deliver a controlled dosage of thermal radiation remains an open challenge.

There are many aspects to consider when designing such assemblies for cellular hyperthermia. Increasing the amount of thermal energy delivered to the tumour itself while maintaining the temperatures of adjacent healthy tissues within safe limits is a major concern. In a general sense, controlling the temperature variation across the tumour boundary using conventional nanoparticle assemblies is not a possibility. Once the nanoparticles reach a particular temperature, over time, the in-vivo temperature gradient is purely controlled by the principles of

thermodynamics. This leads to intractable temperatures within adjacent healthy tissues, ultimately resulting in irreparable damage.

However, utilizing a nanoparticle assembly that obeys the quantum mechanical principle of superradiance allows the generation of a controlled pulse of thermal energy that can be used to significantly improve upon the controllability of the thermal energy delivered to the tumour. The only nanoparticle structure that is popularly known to be superradiant is the ring structure, which has a serious limitation on the number of particles that can be included within a single assembly. Without including enough particles in a single assembly, it is not possible to store the thermal energy required to deliver cancer hyperthermia successfully. Designing another nanoparticle structure that depicts superradiance while supporting a higher particle density remains an open challenge.

1.4 Research Aims and Thesis Scope

1.4.1 Analysis of superradiance in nanoparticles

Recent observations involving superradiance in nanoparticles have led to a surge of literature in this area. While a generally complete quantum mechanical study of superradiance is well established (summarised in chapter 2), certain analytical properties that are unique to perfect electrical conductor materials and quantum dots are still not available in the literature. Our analysis pertaining to these special cases is presented in chapters 4 and 5.

1.4.2 Design a high particle density superradiant structure

Analysis of multi nanoparticle systems is a key requirement in any nanoparticle based design. There are myriads of ways to analyse multi nanoparticle systems.

These techniques vary from purely analytical quantum mechanical approaches to first principle approaches such as density functional theory and purely numerical finite element approaches. It is vital to use the correct simulation technique in order to obtain valid results for the nanoparticle system under study. A detailed summary of such techniques is provided in chapter 3. The superradiant pulses generated from both the commonly known ring structure and the newly proposed buckyball structure are analysed in detail in chapter 5. Several of the analysis techniques are used to completely characterise the behaviour accurately.

1.4.3 Determine the optimal design criteria for the assembly

This involves the analytical formulation of an emitter configuration where enhanced power delivery at the resonant frequency is achieved using principles of superradiance. The particular dipole moment, size, material, optical and other parameters of each quantum dot emitter needs to be carefully determined and validated in the buckyball structure in order to deliver the required superradiant pulse. Since these values need to be experimentally measured, most of the parameters are obtained from the literature and tuned according to our requirements. The analysis pertaining to the tuning and the influence of these criteria are discussed in chapter 4.

1.4.4 Thermal requirement analysis for cancer hyperthermia

This requires the utilisation of practically observed tumour and tissue parameters to develop simple thermal models to identify the energy requirements to successfully deliver cancer hyperthermia. Practically, each tumour type should have a threshold temperature to achieve hyperthermia. Similarly, each tissue type should also have an upper cut off temperature and exposure time to safely avoid undergoing apoptosis. However, experimental observations for most tu-

mour and tissue types pertaining to hyperthermia are currently not available in the literature. Since measuring such practical values is beyond the scope of this thesis, some model criteria will need to be based on assumptions. Complete details of this analysis is presented in Chapter 6.

1.4.5 Simulation and verification

The generated superradiant pulse requires to be simulated and validated using tissue model simulations. This provides the vital understanding of the resulting temperature profile within the tumour as well as the adjacent healthy tissues. The temperature within the tumour should be high enough to sufficiently induce apoptosis while maintaining close to body temperature values at the tumour boundary. Since the tumour and the healthy tissue have different thermal characteristics, a numerical simulation is required in this instance. These details are discussed in chapter 6.

Chapter 2

Superradiance and Emission of Thermal Energy

THis chapter presents a literature based discussion of the concept of superradiance and its extension to the thermal regime. A quantum mechanical approach is used to provide a complete mathematical characterisation of the effect and the criteria that an assembly of emitters must fulfil for superradiant emission are established using first principles.

2.1 Overview

Superradiance is a phenomenon conceptualized by Dicke in 1954 [1]. It is a fundamental theory in quantum photonics that has undergone rigorous studies for several decades [29–31]. It was originally proposed to explain the ephemeral, enhanced, cooperative spontaneous emission characteristics of excited quantum emitters that are coherently coupled and are placed within a sub-wavelength proximity. Recently, based on quantum mechanical time reversal symmetry, the same concept has been extended to describe its absorption counterpart known as *superabsorption* [2].

In order to establish the mathematical background for superradiance, purely in the context of a case study, next we consider the superradiant emission of N interacting Quantum Dot (QD) emitters.

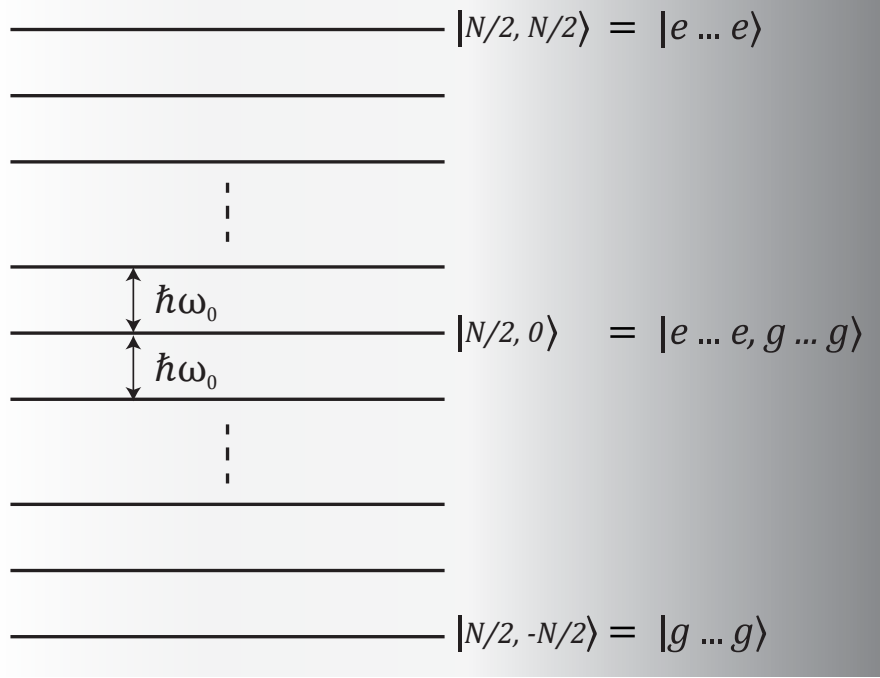


Figure 2.1: The Dicke states form an equidistant ladder of $N + 1$ levels. The emission rate scales quadratically near the ladder centre, implying the existence of strong superradiance. At the top state: $|N/2, N/2\rangle$, all QDs are excited, whereas at the bottom state: $|N/2, -N/2\rangle$, all QDs are in the ground state. At the mid point of the ladder, exactly $N/2$ QDs are excited, whereas the remaining $N/2$ QDs are in the ground state. This is the classical description of the superradiant state.

2.2 Case Study: Emission from N interacting QDs

We begin our analysis by considering an assembly of N interacting QDs confined within sub-wavelength dimensions without adhering to any geometric constraints. Given that the atomic dimensions are much smaller than the emitted wavelength, we approximate each QD by a two-level dipole (excited: $|e\rangle$, ground: $|g\rangle$), and assume that the system is initially excited: $|\psi(0)\rangle = |e\dots e\rangle$. We assume that each QD is identical and the system possesses perfect permutation symmetry; *i.e.* where any two QDs can be interchanged without altering $|\psi(0)\rangle$.

Let:

$$\hat{R}_p^+ = |e\rangle \langle g|, \quad (2.1)$$

$$\hat{R}_p^- = |g\rangle \langle e|, \quad (2.2)$$

$$\hat{R}_p^3 = \frac{1}{2}[|e\rangle \langle e| - |g\rangle \langle g|], \quad (2.3)$$

be the raising, lowering and diagonal Pauli spin operators of the p th QD, respectively. Considering the system evolution and iteratively applying the lowering operator yields the following:

$$|\frac{N}{2}, M\rangle = \sqrt{\frac{(\frac{N}{2} + M)!}{N! (\frac{N}{2} - M)!}} \sum_{p=1}^{\frac{N}{2}-M} \hat{R}_p^- |\psi(0)\rangle, \quad (2.4)$$

where $-\frac{N}{2} \leq M \leq \frac{N}{2}$.

The resulting $|\frac{N}{2}, M\rangle$ represents a fully symmetrical state where $\frac{N}{2} + M$ QDs are excited in the state $|e\rangle$ and $\frac{N}{2} - M$ are in the ground state represented by $|g\rangle$. This implies that the system evolves along a ladder of $N + 1$ equidistant levels known as Dicke states [29, 32], as shown in Fig. 2.1.

Furthermore, it is possible to define the collective operators $\hat{R}^\pm = \sum_{p=1}^N \hat{R}_p^\pm$ and $\hat{R}^3 = \sum_{p=1}^N \hat{R}_p^3$ to obtain the traversing relationship for Dicke states:

$$\hat{R}^\pm |\frac{N}{2}, M\rangle = \sqrt{(\frac{N}{2} \pm M + 1) (\frac{N}{2} \mp M)} |\frac{N}{2}, M \pm 1\rangle. \quad (2.5)$$

Based on these definitions, an expression for inter-Dicke state photon transition rate ($= \Lambda_{at}[\langle \hat{R}^+ \hat{R}^- \rangle]$) can be expressed as:

$$\Lambda_{M \rightarrow M \pm 1}^T = \frac{8\pi^2 d^2}{3\epsilon_0 \hbar \lambda^3} \left[\left(\frac{N}{2} \pm M + 1 \right) \left(\frac{N}{2} \mp M \right) \right], \quad (2.6)$$

where Λ_{at} is the single QD spontaneous emission rate, d is the dipole matrix

element, ϵ_0 is the bath permittivity and $\hbar$ is the reduced Planck constant.

A quadratic emission enhancement is observed when $M \rightarrow 0$ in equation (2.6), which implies that superradiant emission is prominent at the center of the ladder as shown in Fig. 2.1 [1].

Although this quantum mechanical analysis is simple and straightforward, it provides minimal insights about the emitter assembly. In order to investigate the finer details, we must continue the analysis using a master equation approach.

2.3 The Superradiant Master Equation

In order to establish the symmetry requirement of superradiance, it is necessary to study the superradiant master equation (SME) in detail. For the sake of completeness, we outline the derivation steps of this well-known result as follows [29, 32].

For convenience, we begin by separating the emitted electromagnetic field, as a function of the position vector: $\mathbf{s}$, into its positive: $E^+(\mathbf{s})$ and negative: $E^-(\mathbf{s}) = (E^+(\mathbf{s}))^*$ frequency components. The positive component has the form:

$$E^+(\mathbf{s}) = -i \sum_{\mathbf{k}, \mathbf{q}} \frac{\hbar c \mathbf{k}}{2\epsilon_0 \mathcal{V}} \hat{a}_{\mathbf{k}, \mathbf{q}} \exp(i\mathbf{k} \cdot \mathbf{s}), \quad (2.7)$$

where $\mathbf{k}$ is the wave-vector, $\mathbf{q}$ is the polarization vector and $(\mathcal{V} \gg \mathcal{S})$ is the arbitrary quantization volume. Field creation and annihilation operators are denoted by $\hat{a}_{\mathbf{k}, \mathbf{q}}^\dagger$ and $\hat{a}_{\mathbf{k}, \mathbf{q}}$, respectively.

The system Hamiltonian:

$$\mathcal{H}_{sys} = \mathcal{H}_{atm} + \mathcal{H}_{rad} + \mathcal{H}_{int}, \quad (2.8)$$

is the summation of the free atom, radiation field and interaction Hamiltonians,

given by:

$$\mathcal{H}_{atm} = \hbar\omega_0 \hat{R}^3, \quad (2.9a)$$

$$\mathcal{H}_{rad} = \sum_{\mathbf{k}, \mathbf{q}} \hbar\omega_{\mathbf{k}} \left(\hat{a}_{\mathbf{k}, \mathbf{q}}^\dagger \hat{a}_{\mathbf{k}, \mathbf{q}} + \frac{1}{2} \right), \quad (2.9b)$$

$$\mathcal{H}_{int} = - \sum_{p=1}^N (E^+(\mathbf{s}_p) + E^-(\mathbf{s}_p)) \cdot (\hat{R}_p^+ + \hat{R}_p^-) d\tilde{\mathbf{q}}_p, \quad (2.9c)$$

where $\tilde{\mathbf{q}}_p$ is the unit polarization vector of the p th QD.

In the Schrödinger picture, it is possible to define the evolution as: $i\hbar\dot{\Phi} = [\mathcal{H}, \Phi]$, where:

$$\mathcal{H} = \mathcal{H}_{atom} + \mathcal{H}_{rad}. \quad (2.10)$$

This evolution can be expressed in the interaction representation as:

$$i\hbar\dot{\tilde{\Phi}} = [\tilde{\mathcal{H}}_{int}, \tilde{\Phi}]. \quad (2.11)$$

Here the unitary transformations:

$$\tilde{\Phi} = \exp(i\mathcal{H}t/\hbar)\Phi\exp(-i\mathcal{H}t/\hbar), \quad (2.12)$$

$$\tilde{\mathcal{H}}_{int} = \exp(i\mathcal{H}t/\hbar)\mathcal{H}_{int}\exp(-i\mathcal{H}t/\hbar), \quad (2.13)$$

have been used as required.

Quantum electrodynamic theory allows us to express the evolution of the atomic density operator as follows [33]:

$$\dot{\rho} = -\frac{1}{\hbar^2} \text{tr} \int_0^t d\tau [\tilde{\mathcal{H}}_{int}(t), [\tilde{\mathcal{H}}_{int}(t-\tau), \tilde{\Phi}(t-\tau)]]. \quad (2.14)$$

Next we enforce the small volume and Born-Markoff approximations [33], along

with the unitary transformation:

$$\rho(t) = \exp(i\mathcal{H}_{atom}t/\hbar)\tilde{\rho}\exp(-i\mathcal{H}_{atom}t/\hbar), \quad (2.15)$$

on equation (2.14) to derive the model-specific SME.

For analytical simplicity, we will consider the real and imaginary parts of the SME separately as follows:

$$\Re\dot{\rho} = -\frac{\Gamma_{at}}{2}[\hat{R}^+\hat{R}^-, \rho]_+ + \Gamma_{at}\hat{R}^-\rho\hat{R}^+, \quad (2.16a)$$

$$\Im\dot{\rho} = \frac{1}{i\hbar} \left[\sum_{\substack{p,q=1 \\ p>q}}^N \Lambda_{int}(p,q)\hat{R}_p\hat{R}_q, \rho \right], \quad (2.16b)$$

where $\Lambda_{int}(p,q)$ corresponds to the electrostatic Van der Waal's interaction between the QDs. This term is commonly referred to as the hopping-interaction strength [2] because it defines the virtual photon exchange rate among emitters. This concept is discussed in detail in section 3.5.

The pulsating behaviour shown in Fig. 2.2 depicts an ideal scenario where all environmental losses are ignored. In a realistic scenario, there will be several environmental factors determining the actual shape, duration, frequency, amplitude and the controllability of the pulsating behaviour. Here we have assumed that the behaviour is completely controllable by just turning the control field ON and OFF. Furthermore, we have assumed that the zero inter-emitter interaction assembly delivers thermal photons at a constant rate. Both these assumptions are only valid under ideal conditions. However, when a model presents with a controllable superradiant phenomenon, such as our proposed emitter assembly, the pulsating ability is inherent and will definitely be present within practical limitations.

Without a complete QED analysis of the pulsating emission, it is impossible

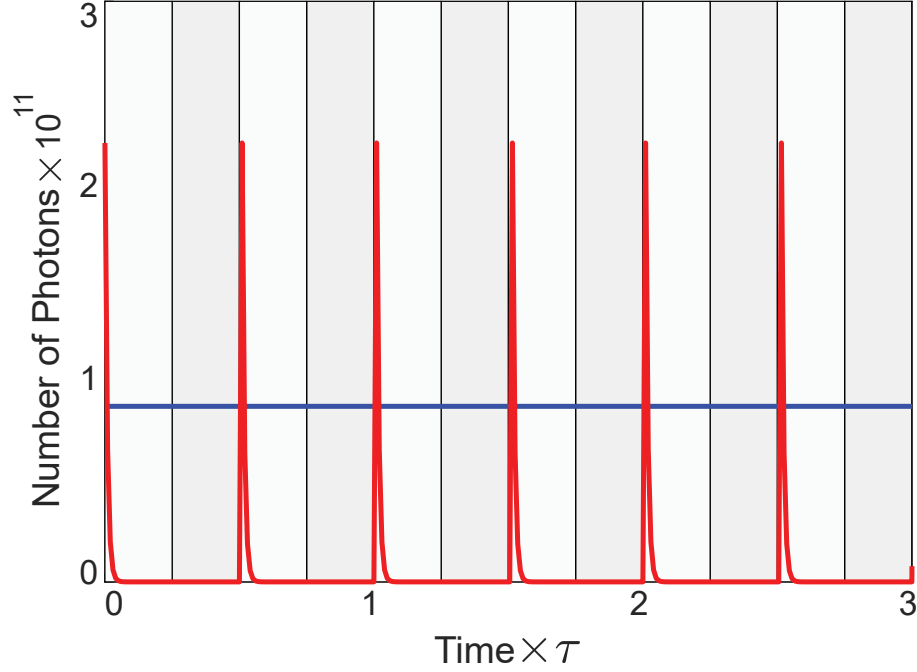


Figure 2.2: This figure shows the ideal emission pattern of repeated superradiant pulses (red). Note the ephemeral nature of the effect and the amount of energy within each pulse. Generating repeated pulses as shown here requires the steady state sustenance of the effect. The blue line is shown as a comparison for a non-superradiant emission case from the same number of emitters. In the presence of environmental and other losses, these emission patterns will depict decay.

to predict the exact details of the pulsation. However, owing to the clearly controllable superradiant phenomenon, we can assume that the pulsating emission will be characteristic to our proposed model.

2.3.1 Analysis of the real component

The real part of the SME in equation (2.16a) confirms our previous derivation and merely reaffirms the symmetrical damping process of the superradiant emitter assembly with intermediary states $|\frac{N}{2}, M\rangle$, as shown in equation (2.4). This is the expected result for the system. Furthermore, the projection of equation (2.16a) along the system states, derives a result similar to equation (2.6) [29].

2.3.2 Analysis of the imaginary component

Careful observation of the imaginary part of the SME presented in eq. (2.16b) reveals a surprising connection regarding the permutation symmetry criteria. It is obvious that any information derived through equation (2.16b) is already implicit in $\mathcal{H}_{sys}$, however, without the separation of real and imaginary parts, an observer will remain oblivious.

The electrostatic Van der Waals interaction is of the form:

$$\Lambda_{int}(p, b) = \frac{d^2}{4\pi\epsilon_0\hbar} \frac{1}{r_{pb}^3} \left[1 - 3 \frac{(\tilde{\mathbf{q}}_p \cdot \mathbf{r}_{pb})^2}{r_{pb}^2} \right], \quad (2.17)$$

and the presence of the dot product $\tilde{\mathbf{q}}_p \cdot \mathbf{r}_{pb}$ clearly challenges the permutation symmetry of an arbitrary geometry of QDs. This fact is discussed in detail in section 2.5.2.

2.4 Emission Rate Characterization

Assuming that the QDs are placed sufficiently far away from each other, there will be no inter-emitter interactions. This is the conventional arrangement and corresponds to the typical scenario of spontaneous emission. The emission of each photon is governed by the single QD spontaneous emission rate, denoted by [29]:

$$\Lambda_{at} = \frac{8\pi^2 |\mathbf{q}|^2}{3\epsilon_b \hbar \lambda^3}, \quad (2.18)$$

where $|\mathbf{q}|$ is the norm of the dipole vector (dipole matrix element), ϵ_b is the bath permittivity, $\hbar$ is the reduced Planck constant and λ is the emitted wavelength.

Let $\mathcal{P}_\omega$ denote the power emitted by the entire assembly at frequency ω . Now, without loss of generality, it is possible to approximate the emission characteris-

tics for the assembly using [34]:

$$\tilde{\mathcal{P}}_{\omega,t} \approx N \exp(-t/\Lambda_{at}) \mathcal{P}_{\omega}. \quad (2.19)$$

Equation (2.19) describes an exponentially decaying pulse of energy, the angular frequency characteristics of which are completely governed by the power spectrum of the emitter assembly.

An emitter assembly needs to adhere with two SRC in order to exhibit superradiant characteristics. These SRC are discussed in detail in section 2.5. Without loss of generality, we now assume that our QD assembly fulfils these criteria. Due to superradiance, the emitted pulse will scale quadratically and its duration will scale inversely with the number of emitters [34]:

$$\tilde{\mathcal{P}}_{\omega,t} \approx N^2 \exp(-t/(N\Lambda_{at})) \mathcal{P}_{\omega}. \quad (2.20)$$

Once a complete spectral characterization is performed on any proposed emitter assembly model, equations (2.19) and (2.20) will allow us to simulate its emission dynamics.

2.5 Superradiant Criteria

Superradiance is a well-studied phenomenon that has attracted attention due to its unique temporal and spectral characteristics. There are strict requirements to which an emitter assembly must adhere, in order to depict superradiance.

Furthermore, it is hypothesized that an assembly maintaining superradiant criteria should also exhibit superabsorption characteristics due to quantum time reversal symmetry [2]. This implies that the proposed assembly should theoretically depict an analogous enhancement in the initial heating process involving the absorption of electromagnetic radiation.

2.5.1 Size of the assembly

This SRC is intuitive; when the linear size of the assembly ($\mathcal{S}$) is much smaller than the wavelength with which it interacts ($\mathcal{S} \ll \lambda$), the emitters become indiscernible to the emitted energy. Therefore, rather than functioning as individual emitters, the entire assembly functions as a global radiating dipole. This phenomenon is fundamental to superradiance. Therefore, any emitter assembly confined within sub-wavelength volumes fulfils the size criterion and is a potential candidate for superradiance. For the case of thermal emission, λ could be as large as several micrometers.

2.5.2 Symmetry of the emitters

This SRC depends on the form of inter-emitter interactions. For QDs that are sufficiently proximal to each other, but not close enough for Förster type coupling or tunnelling, it is possible to assume that Van der Waal's coupling is the main form of inter-emitter photon transfer. In order to understand the behaviour of this coupling, it is necessary to begin with fundamental QED theory to compose and solve the master equation for the superradiant system. This lengthy derivation is detailed elsewhere in the literature [29].

As discussed previously, the imaginary part of the master equation presents the inter-emitter Van der Waal's interaction term defined in equation 2.17. The dot product term: $\hat{\mathbf{q}}_p \cdot \mathbf{r}_{pb}$, as shown in Fig. 2.3b, is pivotal to the symmetry SRC. An assembly of QDs must maintain this coupling rate at a uniform value across all interacting QDs.

If this criterion does not hold, then the emitters will couple with each other at different rates and superradiance will cease to exist. An arbitrary geometric distribution of QDs will obviously breach the symmetry SRC because $\Lambda_{int}(p, b)$ will take arbitrary values for different interacting QDs, depending on the inter-

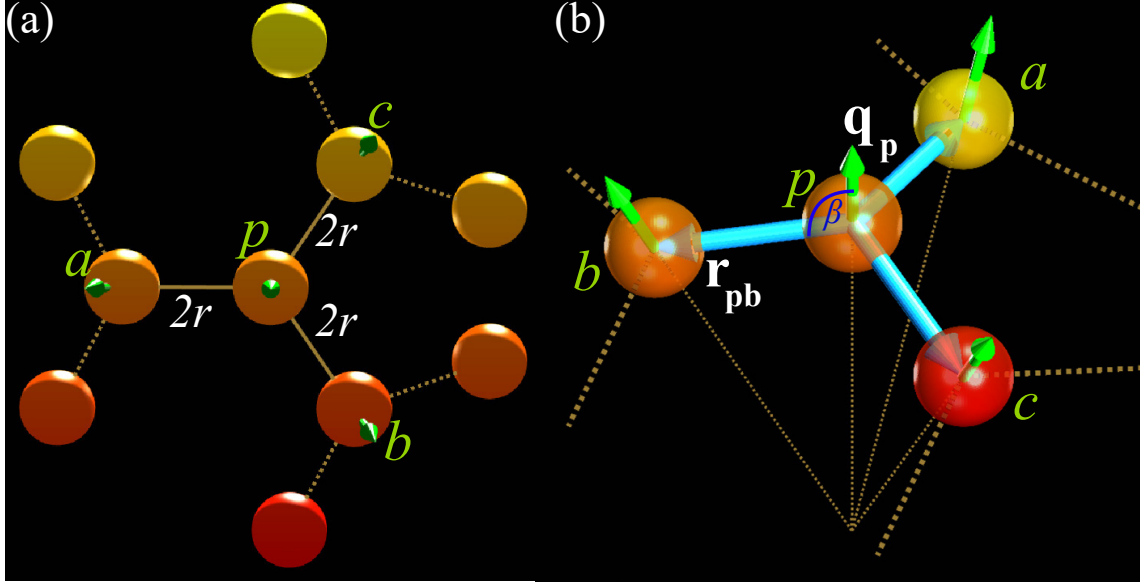


Figure 2.3: (a) Each QD in the assembly (say p) is assumed to interact with 3 nearest neighbours (a, b, c), each at a distance of $2r$. Interactions beyond this limit can be safely ignored for sufficiently large r . (b) The symmetry SRC of the assembly depends on the dot product quantity: $\hat{\mathbf{q}}_p \cdot \mathbf{r}_{pb}$, the value of which relies on the shown inter-vector angle β . Due to the symmetry of a truncated icosahedron, this angle is constant for all QDs across the assembly. This implies that the model is capable of fulfilling the symmetry SRC.

emitter distances and their unique resultant dipoles. This clearly implies that the symmetry SRC cannot be guaranteed for an arbitrary geometric distribution of interacting QDs.

However, a symmetric ring or an equidistant linear QD configuration clearly satisfies the SRC, since it is possible to maintain:

$$\hat{\mathbf{q}}_p \perp \mathbf{r}_{pq} \implies \hat{\mathbf{q}}_p \cdot \mathbf{r}_{pq} = 0, (\forall p, q), \quad (2.21)$$

which explains the popularity of these geometries in the literature [35,36]. However, it is well-known that such structures support superradiant emission only up to a few emitters. This is due to the fact that the number of emitters in the assembly can only be increased until limitation set by the size SRC is met. This is clearly

a limitation and will hinder the practical applicability of superradiance [26].

2.6 Sustaining Superradiance in the Steady State

Recently, many efforts have been made to sustain the effects of superradiance in the steady-state regime for better utilization in applications [9, 37–39]. One major breakthrough in these efforts is the designing and development of the superradiant laser [10]. Several mechanisms are being used to achieve steady-state superradiance. One such method is through the application of quantum feedback control, which involves regulating the number of excitons within different quantum states of the Dicke ladder [40].

This is realized through the careful designing of a photonic cavity or a band-gap crystal that can eliminate undesirable transitions, thus, sustaining the effects of superradiance in the steady-state regime [2].

As proven earlier, superradiance describes a process where the initially excited N QDs traverse down the Dicke ladder (Fig. 2.1a) to its ground state. For practical utilization of superradiance, it is necessary to periodically reinitialize the system to an upper ladder state. However, the enhanced emission is only observed at ladder midpoint state $M = 0$, which is the ideal emission setting for the assembly.

In order to lock the emission to this state, we begin by further analysing the state $|\frac{N}{2}, M\rangle$ in terms of its nearest neighbour interactions. Due to the couplings, it is noted that each state is now associated with a uniquely different dipole state, and the overall emission undergoes chirping [2, 29, 32]. To obtain an expression for the frequency shifts, we write:

$$\hbar\Omega = d^2/4\pi\epsilon_0 r^3 \sum_{p>q} \hat{R}_p^+ \hat{R}_q^- + \hat{R}_p^- \hat{R}_q^+, \quad (2.22)$$

where Ω is the symmetrical perturbation due to the nearest neighbour Van der Waals interactions. Now the value of $\langle \hbar \Omega \rangle$ corresponds to the energy associated with the state and takes the form:

$$\hbar \langle \frac{N}{2}, M | \Omega | \frac{N}{2}, M \rangle = \frac{d^2}{4\pi\epsilon_0 r^3} \frac{(\frac{N}{2})^2 - M^2}{\frac{N}{2} - \frac{1}{2}}. \quad (2.23)$$

The shift in frequencies can be directly calculated using equation; (2.23) and represented using the operator $\hat{\delta}_\omega$:

$$\hat{\delta}_\omega \left| \frac{N}{2}, M \right\rangle = -\frac{d^2}{4\pi\epsilon_0 r^3} \left[1 - \frac{\frac{N}{2} - M}{\frac{N}{2}} \right] \left| \frac{N}{2}, M - 1 \right\rangle. \quad (2.24)$$

The result in equation (2.24) implies that the Dicke ladder in Fig. 2.1 should be modified in the presence of Van der Waals interactions. Each stage has a unique photon transition rate, given by equation (2.4), and a unique transmission frequency, denoted by equation (2.24). It is noted that for $N/2 < M < 0$, the frequencies appear to red-shift and for $0 < M < -N/2$, the frequencies appear to blue-shift during the emission. This frequency separation makes it possible to engineer an effective two-level system by confining the dynamics of the emitter assembly around the favourable $M = 0$ transition. This is achieved with well established quantum control techniques, such as through using photonic band-gap crystals to curtail unfavourable frequencies from the emitter assembly [41, 42]. The use of this technique in detail is analysed in [2] for a superabsorbing system. This modification allows the sustenance of superradiance in the steady-state regime. Once this is achieved, it is possible to utilize the effect meaningfully.

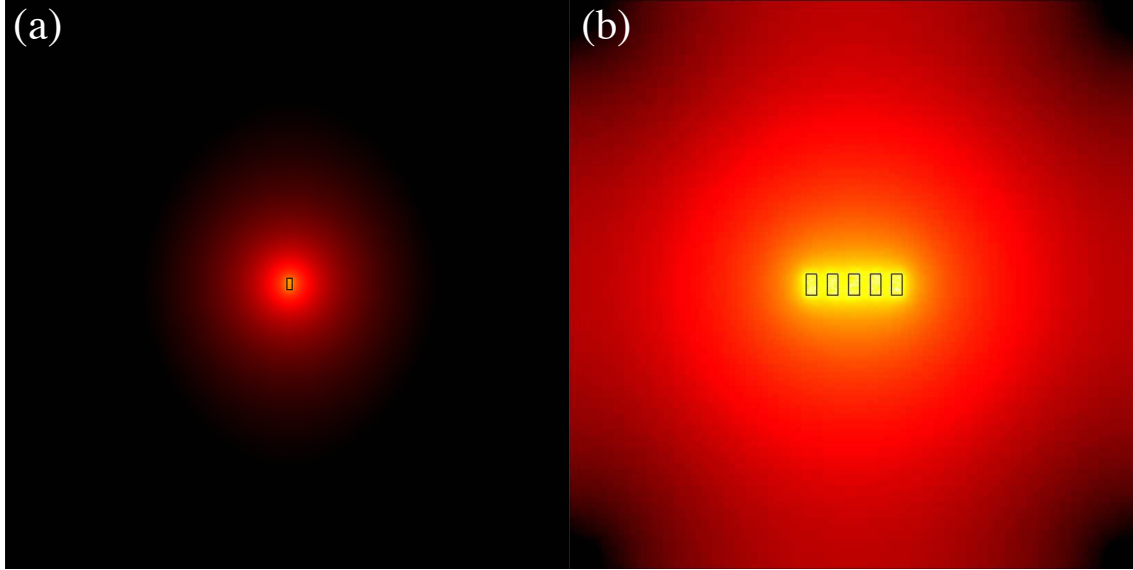


Figure 2.4: COMSOL simulation of (a) 1 and (b) 5 non-resonant emitters with 100 dipoles within each emitter. For the case of multiple emitters, note that the emitters are placed in greater than subwavelength proximity. Notice the linear power enhancement observed with the increase of the number of emitters.

2.7 Extendibility to the Thermal Domain

Until recently, the analogue effect of thermal superradiance was not a topic of interest due to the incoherent nature of bulk thermal emitters [43,44]. Therefore, most of the literature in this area discussed the application of non-resonant bulk emitters as depicted in Fig. (2.4).

However, recent studies show that nanophotonic emitters show interesting coherent and enhanced thermal properties that compel them to acquiesce to the principles of superradiance [45–51]. Beyond displaying coherent characteristics, nanophotonic thermal emitters also possess the ability to radiate beyond the traditional blackbody limit [52,53].

Recently, Zhou *et al.* [34] have studied the superradiant characteristics of a conventional thermal emitter assembly where all emitters are identical and possess similar absorption and far field coupling characteristics. For such an assembly, when the emitters obey the concept of optical resonance and are placed

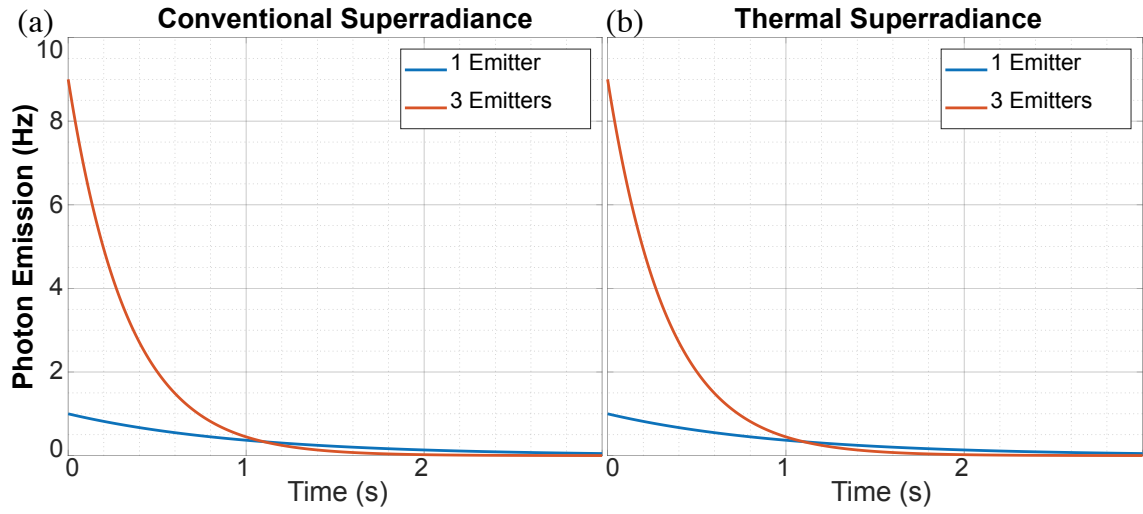


Figure 2.5: Free-decaying behaviour of the (a) conventional and (b) thermal superradiant emitter systems. It is noted that the temporal behaviour of both pulses are identical.

within sub-wavelength dimensions, superradiant phenomena can be observed at a controllable resonance frequency as long as the over-coupling relationship is maintained. However, the sub wavelength emitter placement requirement is difficult to be achieved using conventional dielectric resonators because their dimensions exceed required criteria. Therefore, the conventional emitter assembly in [34] assumes that all emitters possess ultra-compact electromagnetic resonance features. In practice, these resonance features can be simulated using perfect electric conductor (PEC) deep sub wavelength slits [54].

While the transient, ephemeral character of the effect is unaltered as observed in Fig. 2.5, thermal superradiance introduces significant spectral broadening. For conventional assemblies, they prove that the emission power at resonance is scaled inversely as a function of the number of emitters in the assembly. Due to this anomalous phenomenon, conventional superradiant thermal emitter assemblies always fail to outperform non-coupled emitter assemblies at resonance, and thus, it curtails the practical applicability of thermal superradiance. This fact is discussed in detail in Chapter 5.

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Chapter 3

Analysis of Multi-Nanoparticle Systems

This chapter provides a detailed assessment of various techniques involved in the analysis of complex multi-nanoparticle systems in the framework of a literature review. This well-known analysis in theoretical physics is a multi-faceted problem that often times requires the utilization of several techniques.

3.1 Overview

The principles of classical physics that govern macroscopic systems are both simple and elegant. They are comparatively easy to understand, model and simulate. Furthermore, in almost all macroscopic cases, the explanations provided by the fully classical analysis is quite sufficient to study and predict the behaviour of multi-particle systems. Many modern examples of multi-particle analysis using classical physics can be found in popular 3-D animation and video game development software packages [55].

However, the analysis of multi-nanoparticle systems is a comparatively complex process. The main difference influencing the observed variations between bulk and nano-particles is due to ‘electron confinement’. When atoms are considered in isolation, it is well known that the energy levels within the atoms are discrete. However, when the atoms are considered in bulk, the energy levels un-

dergo splitting and form bands. The observations made in nanoparticles are in the intermediary stage between these two observations.

These effects arise due to the fact that the size of these nanoparticles are now comparable with the de Broglie wavelength of the electron wave function. This discretization of the permitted energy levels is simple to solve and compute for a single particle system. In fact, an electron confined in a potential well is one of the most well-known problems in quantum mechanics [56]. However, the problem of solving multi-nanoparticle systems become extremely complex with the increase of the number of particles.

3.2 Fundamental Approaches

It is possible to mathematically approximate and express any multi-particle system using an eigenvalue equation, as long as the wave functions of each and every electron and nuclei component within the multi-particle system is known.

$$\hat{\mathcal{H}}\psi(r_i^-, R_i^+) = E\psi(r_i^-, R_i^+), \quad (3.1)$$

where r_i^- and R_i^+ correspond to the coordinates of the i^{th} electron and nuclei of the particles and E is the total energy. The simplified Hamiltonian can be expressed as,

$$\hat{\mathcal{H}} = \hat{\mathcal{T}} + \hat{\mathcal{V}}, \quad (3.2)$$

where $\hat{\mathcal{T}}$ and $\hat{\mathcal{V}}$ correspond to the Kinetic and Potential energy operators, respectively. Without any simplifications, this equation is cumbersome and difficult to fathom. The classical approach follows with a set of approximations.

3.2.1 Born-Oppenheimer approximation

This famous approximation, also known as the clamped nuclei approximation, begins by assuming that the mass of the nuclei are comparatively much larger than the mass of electrons and the speed of motion of nuclei are comparatively much smaller than the speed of motion of electrons. This assumption means that the kinetic energy of the nuclei can be safely neglected in the system. While complex mathematics is required to fully understand the validity and applicability of this approximation, the pivotal end result is the decoupling of the wavefunction,

$$\psi(r_i^-, R_i^+) \rightarrow \psi_N(R_i^+) \otimes \psi_e(r_i^-), \quad (3.3)$$

to the nuclei: $\psi_N(R_i^+)$ and electron: $\psi_e(r_i^-)$ components.

This permits the restructuring of the total Hamiltonian equation. For simplification purposes, based on the approximation, now we study only the kinetic energy of electrons and the potential energies due to the attractions between nuclei-electrons and the repellents between electrons-electrons.

$$\hat{\mathcal{H}}\psi(r_i^-) = E\psi(r_i^-), \quad (3.4)$$

where the Hamiltonian operator now has the form:

$$\hat{\mathcal{H}} = -\frac{\hbar^2}{2me} \sum_i^N \nabla_i^2 + \sum_i^N V^{--}(r_i) + \sum_i^N \sum_{j>1}^N V^{+-}(r_i, r_j). \quad (3.5)$$

Note that short have notations have been used for the repellent (V^{--}) and attraction (V^{+-}) potential operators, respectively. The mathematical nature of these operators are discussed further in section 3.5.

It is noted that this simplified approximation, when applied to a case study of a 3-dimensional cluster of 100 Pb atoms, results in a 24600 dimension problem

that is still quite cumbersome and difficult to solve. Therefore, further simplifications are clearly necessary to solve multi-particle systems.

3.2.2 Density functional theory

Density functional theory (DFT) is referred to as an *ab initio* method with a profound reputation for modelling the electronic structure of matter [57]. It begins with the understanding that the complexity of solving multi-particle systems is due to modelling each electron as a unique wave function. Instead, DFT approaches the problem based on electron density: $n(r)$.

Now, an arbitrary i_{th} electron in the multi-particle system is treated as a single point charge in the field of all electrons. This simplifies the ‘many electron problem’ to a ‘many-one electron problem’. This mean-field assumption allows the following formulation, known as the Hartree product, to be made:

$$\psi(r_1, r_2, \dots, r_N) = \psi(r_1) \times \psi(r_2) \times \dots \times \psi(r_N), \quad (3.6)$$

It is noted that in the most abstract sense, the direct Hartree product does not satisfy fermionic dynamics.

The more complex Slater determinant is required for a complete description. In fact, more advanced and accurate multi-particle solving techniques such as ‘configuration interaction’ and ‘multi configurational self consistent field’ rely upon the Slater determinant instead of the Hartree product to define the ‘many-one electron problem’.

Now it is possible to express the electron density as follows:

$$n(r) = 2 \sum_i \psi^*(r_i) \psi(r_i), \quad (3.7)$$

The Hohenberg-Kohn theorems, which state that:

1. the ground state energy E is a unique functional of the electron density:
 $E = E[n(r)]$ and,
2. the electron density that minimizes the energy of the overall functional is the ground state energy density: $(E[n(r)] > E[n(r_0)], \forall r)$,

are the fundamental principles governing DFT.

Now, the energy functional ($E[n(r)]$) described in this theorem can be divided into ‘known’ and ‘unknown’ components. The known components arise due to the kinetic and potential components of the Hamiltonian that were described in equation 3.5. The unknown components, also known as ‘exchange-correlation’ functionals, arise due to the presence of quantum mechanical effects. DFT does not present a complete method to calculate the exchange-correlation functional, instead, it has to be approximated using various techniques.

Based on these ideas, now it is possible to present the so-called ‘Kohn-Sham scheme’, which is a straight forward minimization algorithm that attempts to find the ground state electron density of the multi-particle system. In turn, the ground state electron density functional can be used to solve for and yield the electron states and orbitals of the multi-particle system.

Over many decades, the DFT technique has undergone many modifications and in the process, has acquired the ability to solve numerous complex problems and structures. However, its complexity when solving multi particle systems with complex structures and the lack of simulation accuracy during certain types of computations are serious problems that hinder the applicability of the DFT technique.

3.2.3 Mie’s theory of scattering

Mie’s theory provides the analytical solution to the diffraction problem of the time-harmonic electromagnetic field in the presence of a homogeneous spheri-

cal particle [58]. This classical theory in physics has been extended for studying the scattering and absorption behaviour of spherical and cylindrical multi-nanoparticle systems [59]. The analytical solution relies on the use of Bessel functions and is of significant practical interest for modelling the spectroscopic behaviour of either spherical or rod-like mixtures of nanoparticles.

3.3 Numerical Simulation Techniques

Often times, multi-nanoparticle systems comprise particle shapes that are highly irregular. Purely analytical equations such as Mie's theory, although can be written to define the physics of such arbitrary shapes, often times cannot be solved using ordinary techniques. Even if a solution can be obtained, it will be extremely cumbersome and will be of very limited use. Furthermore, although fundamental analytical models can be presented for most of the problems involving multi-nanoparticle systems, solving them requires the utilisation of various numerical techniques.

Often times, these numerical techniques depend on the use of sophisticated and parallelized differential equation or integral solvers. An interface is provided to design the geometry of the multi-nanoparticle system, which is then 'meshed' to discretize the space parameter. Meshing itself involves self-deterministic, sophisticated algorithms that can ideally vary the particle size depending on the complexity of the geometry involved. The rest of this section briefly summarizes a few of the popular numerical techniques that are widely used in mathematical physics in order to provide solutions for multi-nanoparticle systems.

3.3.1 Computational electromagnetics techniques

Computation electromagnetics (CEM) is an umbrella term that is used to identify technique that model the interactions between electromagnetic waves and various object types and baths (environments). For the most part, all of these techniques provide solutions to the Maxwell's equations, either in raw form, modified forms or through the use of approximations. Since Maxwell's equations can be expressed in either integral or differential form, these techniques are also divided based on the type of solver used. Techniques such as finite difference time domain and finite element method are differential techniques, whereas discrete dipole approximation and method of moments element rely upon integral solvers.

Finite difference time domain technique

One of the most popular numerical techniques of solving electromagnetic problems in physics is known as the finite difference time domain (FDTD) technique. The concept is to solve an electromagnetic problem by discretizing time/space parameters and then use Maxwell's equations with approximations to arrive at a converging solution. The method itself is highly versatile and can solve problems involving various material types, shapes, geometries, optical properties and baths. It is also possible to sweep across parameters to observe characteristic changes in problems such as scattering, absorption and emission. This allows researchers to easily validate and configure parameters such as the resonance frequency and ideal bath permittivity.

In the most simple context, the main concept of FDTD is to convert Maxwell's derivative equations into central difference approximations. The algorithm iteratively solves for both the E-field and H-field components in both space and time domains. Due to its intrinsic simplicity, this approach can be used to solve for

1-D, 2-D and 3-D problems without difficulty.

However, it is noted that the approach used here is purely semi-classical. By using an FDTD simulation, it is not possible to obtain any knowledge on the quantum mechanical behaviour of the structure. Therefore, FDTD simulations are better suited for confirming and validating absorption, scattering and emission data of multi-nanoparticle systems that are already experimentally investigated.

Finite element method

Finite element method (FEM) is another popular technique used in computational physics to analyse systems with various complexity levels. Unlike the FDTD technique that provides solutions in the context of a time varying electromagnetic field, the FEM technique assumes the presence of a time-harmonic dependence in the field. Furthermore, the FEM technique is highly suited to study the temperature variation of systems but studying ‘heat-flow’ problems, specifically relevant to this thesis in the context of ‘tissue models’.

Discrete dipole approximation

The discrete dipole approximation (DDA) method is used to study the scattering and absorption of electromagnetic radiation from arbitrarily shaped particles. The concept is to replace the complex geometry of the nanoparticles with a set of point dipoles, that can then be modelled using a linear set of equations where the inter-dipole interactions as well as field-dipole interactions are exclusively taken into account. The mathematical formulation depends on the dyadic Green’s function, which allows the expression of the electric field inside the dielectric scatterer to be expressed in the form of an integral.

Over the years, several DDA approaches have been developed with improved

accuracy and computational time. In fact, modern approaches to DDA relies on iterative solvers to solve the scattering problem with a higher degree of numerical accuracy. One such extension is popularly known as the ‘fast multipole method’, which utilizes the fast Fourier transform to reduce the computational complexity of the system [60].

Method of moments

This method is also referred to as the boundary element method, due to the fact that it only computes values at the boundaries of the structure. However, this technique lacks versatility due to the fact that it requires a complete Green’s function formalism and is generally only applicable to linear homogeneous media [61].

3.4 Coupled Mode Theory

Coupled mode theory (CMT) has a history dating back to 1954, where it was first used to study the characteristics of microwave travelling-wave tubes [62]. It is best described as a perturbational technique that allows to capture both spatial and temporal variations of vibrational systems [63]. Recently, this technique was extended to predict the behaviour of nanophotonic systems with a high degree of accuracy [64].

It is note that this model is somewhat restrictive due to the fact that at the formulation level, it assumes linearity and weak mode coupling to be present in the multi-nanoparticle system. However, due to its ability to elegantly describe the electric field intensity variation within each element of the multi-nanoparticle system, it was chosen as the primary analysis technique for the work presented in this thesis.

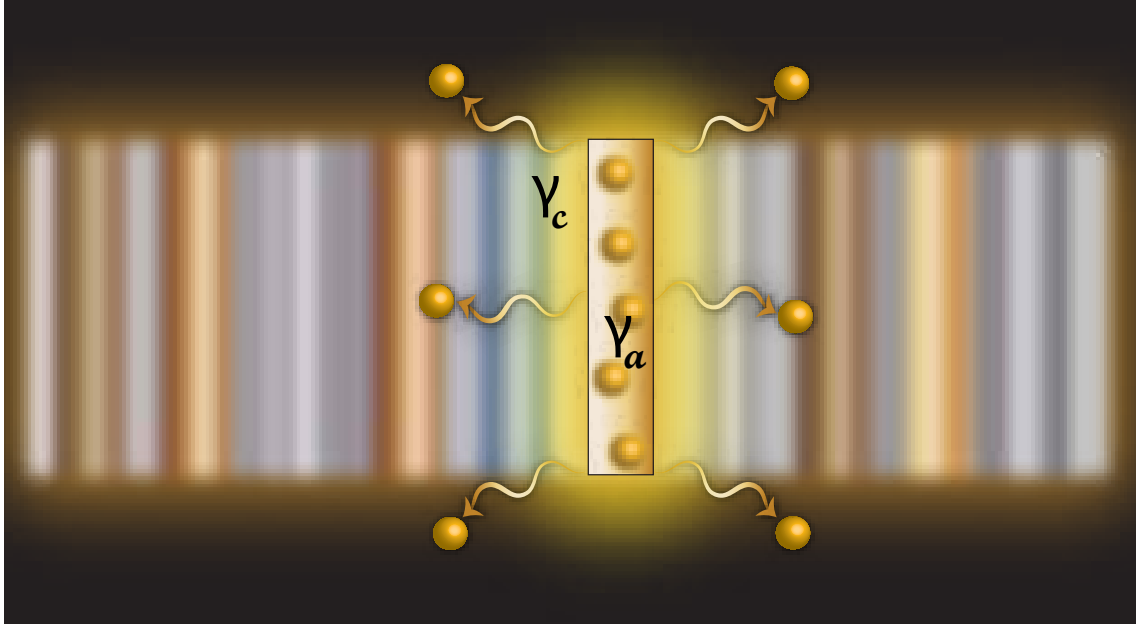


Figure 3.1: A singular emitter is highlighted to show the operation parameters of the assembly. The photons emanating from the emitter are coupled to the far-field at a rate of γ_c and the photons engulfed within the emitter are absorbed at a rate of γ_a . Note that each emitter has a unique configuration of γ_a and γ_c , which leads to myriads of interference configurations, resulting in the observed startling phenomena.

General CMT model for a nanophotonic assembly of emitters

Let the normalized, time and frequency varying, electric field amplitudes within the nanophotonic emitters be denoted by:

$$\mathbf{a}(\omega, t) = [a_1(\omega, t) \dots a_N(\omega, t)]^T. \quad (3.8)$$

Then the system dynamics are captured using the CMT equation:

$$\frac{\partial}{\partial t} \mathbf{a}(\omega, t) = [j\mathbf{\Omega}_0 - \mathbf{\Gamma}] \mathbf{a}(\omega, t), \quad (3.9)$$

where $\mathbf{\Omega}_0 = \text{diag}[\omega_0^1 \dots \omega_0^N]$ contains the resonant frequencies of each emitter. The $N \times N$ matrix:

$$\mathbf{\Gamma} = \mathbf{\Gamma}_A + \mathbf{\Gamma}_E + \mathbf{\Gamma}_{int}, \quad (3.10)$$

encodes the rates of photon absorption: $\mathbf{\Gamma}_A = \text{diag}[\gamma_A^1 \dots \gamma_A^N]$ and emission: $\mathbf{\Gamma}_E = \text{diag}[\gamma_E^1 \dots \gamma_E^N]$ of all emitters. Finally, the inter-emitter coupling rates are encoded within $\mathbf{\Gamma}_{int}$. It is noted that this set of linear differential equations completely characterise the first order energy transfers that occur within the entire multi-nanoparticle system, with adherence to the principle of energy conservation.

3.5 Inter Particle Interactions

Inter-atomic interactions and forces play a crucial role in the context of shaping the atomic structure. Furthermore, these interactions are the key to explaining the presence of unique characteristics in certain molecules such as water, where Hydrogen bonds govern its entire behavioural spectrum. Although the separations between nanoparticles are several orders of magnitude higher than the separations between atoms in molecules, when analysing multi-nanoparticle systems, a crucial understanding of the inter-particle interactions that apply to the situation is of significance.

Due to the nanoscopic dimensions, it is commonplace in the literature to treat nanoparticles as point objects and model them as dipoles. Furthermore, under the influence of external electric fields, nanoparticles are known to become polarized and therefore, depict characteristics of induced dipoles.

In the CMT framework discussed in section 3.4, a complete analysis of inter-particle interactions is required to model the properties of $\mathbf{\Gamma}_{int}$. In fact, being able to fully define $\mathbf{\Gamma}_{int}$, along with the application of the principle of energy conservation, is the sufficient condition to completely analyse a system using CMT. This fact is mathematically proven in chapter 4.

In order to understand how inter-particle interactions occur, it is important to begin by mathematically defining the concept of a dipole.

3.5.1 Polarizability

In order to comprehend the dipole moment of a nanoparticle, it is important to understanding how the concept of dipolar polarizability applies to the same particle. If the polarizability α of a particle is known, the induced dipole moment can be expressed using:

$$u_i = \alpha \times E, \quad (3.11)$$

where E is the electric field incident upon the particle. The description of E has to be varied according to the definition of the incident electric field [65]. Depending on the situation, it is possible to induce high dipolar moments within nanoparticles.

3.5.2 Self-energy of a dipole

A dipole is modelled as a pair of $\pm$ charges, separated by a certain distance, and is known to possess an electrostatic energy denoted by:

$$\mu = \frac{q^2}{4\pi\epsilon_0\epsilon r}, \quad (3.12)$$

where q is the charge amount and r is the separation. This formulation is similar to the Born self-energy of an ion [65].

3.5.3 Static inter-dipole interactions

Two polar nanoparticles that possess dissimilar dipole moments $u_{1,2}$ that are separated by a distance r results in an inter-dipole interaction energy denoted by:

$$w = \frac{u_1 u_2}{4\pi\epsilon_0\epsilon r^3} \times [2 \cos \theta_1 \cos \theta_2 - \sin \theta_1 \sin \theta_2 \cos \phi], \quad (3.13)$$

where $\theta_{1,2}$ are the intersecting angles between each dipole's charge-charge vector and the interacting dipole-dipole vector. When the two dipoles are lying on a line and the two vectors are normal, $\theta_{1,2} = 0$ results in the maximisation of interaction energy:

$$w = u_1 u_2 / (4\pi\epsilon_0\epsilon r^3). \quad (3.14)$$

When the two dipoles are aligned parallel to each other, $\theta_{1,2} = \pi/2, \phi = \pi$, the total interaction energy is half-maximum.

3.5.4 Keesom interactions

It is noted that unlike in complex molecules where static dipole interactions can occur and $\theta_{1,2}$ and ϕ angles become significant in evaluating the total dipole-interaction energy, in the case of nanoparticles, it makes sense to obtain the angle averaged interaction energies. This is due to the fact that at temperatures higher than absolute zero ($T > 0$), these nanoparticle dipoles are free to rotate.

In order to obtain the angle-averaged free, the potential distribution theorem is used [66]. Based on simple mathematical relationships, it is possible to obtain the following relationship for the angle averaged interaction energy between two dissimilar rotating dipoles.

$$w = \frac{u_1^2 u_2^2}{48\pi^2 \epsilon^2 \epsilon_0^2 k T r^6}, \quad (3.15)$$

which is valid when:

$$kT > u_1 u_2 / 4\pi\epsilon_0 \epsilon r^3. \quad (3.16)$$

This famous relationship of the Boltzmann-averaged interaction between two permanent dipoles is referred to as the *Keesom* interaction energy and is one of the three major contributors to the *van der Waals* interaction between particles. It is also referred to as the *orientation* energy.

3.5.5 Van der Waals interactions

Van der Waals interactions provide a somewhat complete model for the total interaction energy between two polarized nanoparticles. While the model is originally proposed for polar molecules, there are similarities that can be made between polar molecules and polarized nanoparticles. There are three components to Van der Waals interactions. The *induction* part is purely due to the polarizability impact of one molecule on the other. The *orientation* part was discussed in section 3.5.4 and the final part is the *dispersion*. These dispersion forces arise due to interactions between instantaneous multipoles.

A generalized theory for Van der Waals interaction energy has the following form:

$$w = -\frac{6kT}{(4\pi\epsilon_0)^2 r^6} \left(\sum_{n=1,2,\dots}^{\infty} \frac{\alpha_1(iv_n)\alpha_2(iv_n)}{\epsilon_3^2(iv_n)} + \frac{1}{2} \frac{\alpha_1(0)\alpha_2(0)}{\epsilon_3^2(0)} \right), \quad (3.17)$$

where terms corresponding to subscripts 1, 2 apply to the two nanoparticles and 3 applies to the medium. The complex frequency term $v_n = nkT/\hbar$.

Although these energy values seem straightforward and computable based on observables, it is worth noting that within a multi-nanoparticle system, taking the summation of all pairwise Van der Waals interactions between a particle of interest and all other particles to find the total interaction energy of a single particle is not accurate. The reason for this anomaly is simple to understand. In the pres-

ence of multiple particles, the field that emanates from a single particle not only interacts with one other particle directly, but gets reflected from each and every other particle, which in turn results in a field amplification around the particle in question. If all the pairwise energies are summed to find the total interaction energy of a single particle, the field amplification due to reflections are ignored.

Furthermore, in extremely proximal scenarios where the separation between nanoparticles is negligibly small (1-5nm), dispersion energies tend to govern the total Van der Waals interaction energy. This is the scenario that is applicable to molecular structure. However, for multi-nanoparticle mixtures, usually the separations are much larger and therefore, Keesom energy becomes the governing contributor [65].

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Chapter 4

Controlled Generation of a Thermal Superradiant Pulse

This chapter provides a comprehensive description of the mathematical efforts involved in the controlled generation of a superradiant pulse in the thermal regime. The analysis is primarily based on coupled mode theory and three types of emitter assembly configurations are presented and compared based on the analytical framework.

4.1 Mathematical Characterization

WE base the fundamental analysis of our model on coupled mode theory (CMT) [63, 67, 68]. CMT provides a solid platform for analyzing the behavior of intrinsic electromagnetic fields of an assembly of nanophotonic emitters (thermal or optical) placed within subwavelength proximity and has been used extensively in literature [64, 69]. Although CMT provides a straight forward and generally comprehensive analysis for the energy emission at the point of resonance, by definition, the framework tends to ignore other energy transfers especially akin to non-resonant modes. Therefore, within this model, we assume that all such non-resonant modes are sufficiently far away from the main mode of resonance and contribute less towards the power at resonance.

We begin by modifying the generalized CMT equation (eq. 3.9) as follows:

$$\frac{\partial}{\partial t} \mathbf{a}(\omega, t) = [j\mathbf{\Omega}_0 - \mathbf{\Gamma}] \mathbf{a}(\omega, t) + \diamond(\omega, t), \quad (4.1)$$

where the term $\diamond(\omega, t)$ is added as a direct consequence of thermal energy generation. Here we assume that each nanophotonic emitter is modelled based on a grey-body radiator. Since each source emits in accordance with the fluctuation dissipation theorem, and assuming a stationary characteristic for each source, $\diamond(\omega, t)$ converts to the time invariant form:

$$\diamond(\omega) = \sqrt{\mathbf{\Gamma}_A} \mathbf{n}(\omega). \quad (4.2)$$

Now, the $N \times 1$ matrix $\mathbf{n}(\omega)$ has the form [70]:

$$\langle n_p^*(\omega) n_b(\omega) \rangle = \Theta(\omega, T) \delta(\omega - \omega') \delta_{pb}, \quad (4.3)$$

where:

$$\Theta(\omega, T) = \frac{\varepsilon_\omega \hbar \omega}{(2\pi \exp(\hbar \omega / k_B T))}, \quad (4.4)$$

is the well-known Planck energy of thermal photons radiating at temperature T and angular frequency ω . Here, ε_ω and k_B denote the emissivity of the emitters and the Boltzman constant, respectively.

4.1.1 Simplified periodic boundary model for free space

Under resonant conditions, it is necessary to define a simplified mathematical model for free space in order to characterize the behaviour of $\mathbf{\Gamma}_{int}$.

In order to model free-space, we use the temporal coupled-mode theory based approach suggested by Verslegers *et al.* [71]. In this approach, free-space is assumed to comprise of an infinite number of modes and fragmented into an infi-

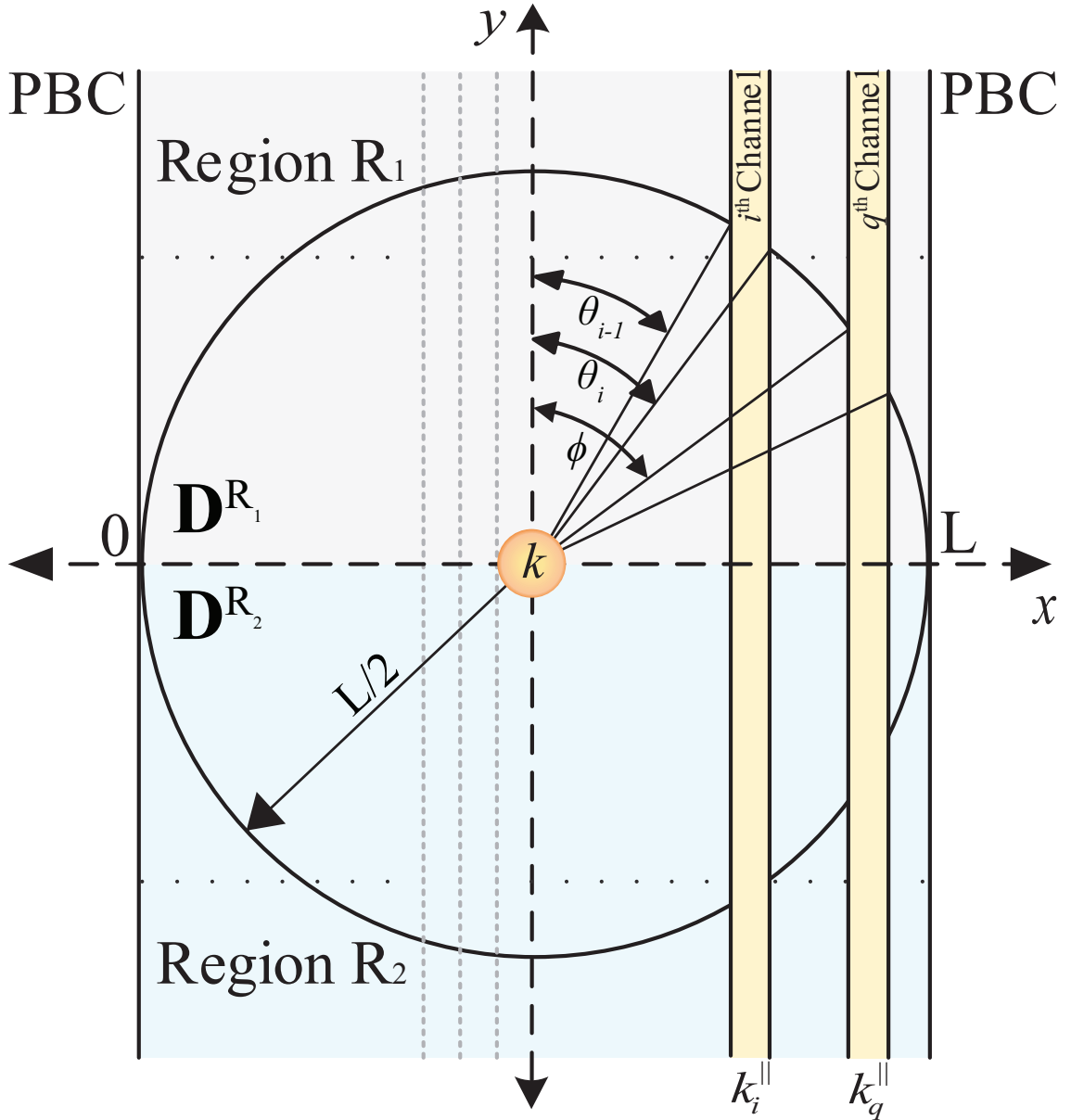


Figure 4.1: Diagram illustrating the vertical distribution of normalized and orthogonal radiation channels. The k^{th} emitter is located at the mid point betwixt the periodic boundaries located a distance of L apart. The infinite reality of free-space is achieved by considering $L \rightarrow \infty$.

nite number of resonating Fabry-Perot cavity based radiation channels. Depending on the type of metal used for emitter design, plasmonic-mode channels may also appear. However for this analysis, we assume that the emitter assembly is

constructed using non-plasmonic metals and neglect this phenomenon. This is a safe assumption for PECs due to their infinite permittivity characteristics that prevent them from creating surface plasmons [19] and can be safely extended to some metals when the emissions occur at higher optical frequencies [72–74].

Furthermore, modal analysis is used to prove that the plane wave propagating channels should be orthogonal [67]. Based on these assumptions and proofs, the 2-dimensional free-space is modelled using a set of orthogonal and normalized radiation channels denoted by $\mathbf{S}$, where:

$$P_i = |S_i|^2, \quad (4.5)$$

denotes the power of the i^{th} channel.

In order to deal with the infinite nature of free-space, a periodic boundary condition (PBC) of length L is introduced along the x direction such that, $L/\gamma_0 \in \mathbb{Z}^+$, where γ_0 is the resonant wavelength. Enforcing $L \rightarrow \infty$ enables the recovery of free-space conditions.

The i^{th} channel propagates a plane wave represented by the parallel component of the wave vector:

$$\mathcal{K}_i^{\parallel} = i(2\pi/m\gamma_0), \quad (4.6)$$

where $m = L/\gamma_0$.

In this model, there are $2m$ radiation channels enclosed within the periodic boundary and each channel is uniquely represented by its index:

$$i = (-m, -m+1, \dots, -1, 1, \dots, m-1, m). \quad (4.7)$$

Each channel has a spatial width of $\gamma_0/2$, which implies that the orthogonal set of channels uniformly sample the parallel wave vector space as shown in Fig. 4.1.

Overall emission to free-space from all emitters can be defined using the fol-

lowing relationship:

$$\mathbf{S} = \begin{bmatrix} \mathbf{D}^{R_1} \\ \mathbf{D}^{R_2} \end{bmatrix} \mathbf{a} = \mathbf{D}\mathbf{a}, \quad (4.8)$$

where $\mathbf{D}^{R_1}$ and $\mathbf{D}^{R_2}$ are of dimensions $2m \times N$. Due to the symmetric nature of the two regions, it is observed that $\mathbf{D}^{R_1} = \mathbf{D}^{R_2}$, and their elements $D_{i,k}^{R_1}$ and $D_{i,k}^{R_2}$ represent the coupling of the k^{th} emitter to the i^{th} channel in regions R_1 and R_2 , respectively.

Next we derive an analytical solution for $\mathbf{D}$ by considering the coupling from the k^{th} emitter to the free-space. We begin by analysing the geometry in Fig. 4.1. We assume that the k^{th} emitter of the assembly placed at the center has an isotropic radiation characteristic. Using the definition for:

$$\text{arc}(\Delta\theta_i) = \text{arc}(\theta_i - \theta_{i-1}) = (L/2)\Delta\theta_i. \quad (4.9)$$

The following result can easily be proven using simple trigonometric relations:

$$\text{As } L \rightarrow \infty, \cos \theta_i = \frac{\gamma_0/2}{\text{arc}(\Delta\theta_i)}, \quad (4.10a)$$

$$\cos \theta_i = \frac{L/2}{m} \frac{1}{(L/2)\Delta\theta_i}, \quad (4.10b)$$

$$\gamma_0/2 = (L/2)\Delta \sin \theta_i, \quad (4.10c)$$

$$\Delta \sin \theta_i = \left(\frac{i\gamma_0/2}{L/2} - \frac{(i-1)\gamma_0/2}{L/2} \right) \quad (4.10d)$$

$$\Delta \sin \theta_i = \gamma_0/L \quad (4.10e)$$

$$m\Delta\theta_i = \frac{1}{\cos \theta_i}. \quad (4.10f)$$

The normal projection of the coupling components of the k^{th} emitter to the i^{th}

channel can be expressed as follows:

$$\gamma_{c_{\perp \leftarrow k}} = \gamma_{c_{\theta_i \leftarrow k}} \cos \theta_i, \quad (4.11)$$

where $\gamma_{c_{\theta_i \leftarrow k}}$ denotes the coupling from the k^{th} emitter along the θ_i direction and $\gamma_{c_{\perp \leftarrow k}}$ denotes the coupling component of the k^{th} emitter along the normal direction (parallel to the y axis).

Based on the relationship in (4.11), we obtain the following equation for couplings from the k^{th} emitter to all channels in both regions R_1 and R_2 ,

$$\gamma_{c_k} = 2 \sum_{\substack{i=-m \\ m \neq 0}}^m \gamma_{c_{\theta_i \leftarrow k}} = 2\gamma_{c_{\perp \leftarrow k}} \sum_{\substack{i=-m \\ m \neq 0}}^m \frac{1}{\cos \theta_i} \quad (4.12)$$

By applying the summation condition to (4.10f) and considering either of the two symmetric regions (R_1 or R_2), the following can be derived to simplify (4.12):

$$\sum_{\substack{i=-m \\ m \neq 0}}^m \frac{1}{\cos \theta_i} = m \sum_{\substack{i=-m \\ m \neq 0}}^m \Delta \theta_i = m\pi, \quad (4.13)$$

where $\theta_i \in (0, \pi/2)$. Substituting the result from (4.13) in (4.12), the following answer is derived,

$$\gamma_{c_k} = 2\gamma_{c_{\perp \leftarrow k}} m\pi. \quad (4.14)$$

The coupling element can now be written using the results from (4.11) and (4.14) as follows:

$$D_{i,k}^{R_1} = D_{i,k}^{R_2} = \sqrt{\gamma_{c_{\theta_i \leftarrow k}}} = \sqrt{\frac{\gamma_{c_k}}{2m\pi \cos \theta_i}}, \quad (4.15)$$

which allows us to explicitly generate the following:

$$\mathbf{D}^{R_1} = \mathbf{D}^{R_2} = \begin{bmatrix} \sqrt{\frac{\gamma_{c_1}}{2m\pi \cos \theta_{-m}}} & \cdots & \sqrt{\frac{\gamma_{c_N}}{2m\pi \cos \theta_{-m}}} \\ \vdots & \ddots & \vdots \\ \sqrt{\frac{\gamma_{c_1}}{2m\pi \cos \theta_{-1}}} & \cdots & \sqrt{\frac{\gamma_{c_N}}{2m\pi \cos \theta_{-1}}} \\ \sqrt{\frac{\gamma_{c_1}}{2m\pi \cos \theta_1}} & \cdots & \sqrt{\frac{\gamma_{c_N}}{2m\pi \cos \theta_1}} \\ \vdots & \ddots & \vdots \\ \sqrt{\frac{\gamma_{c_1}}{2m\pi \cos \theta_m}} & \cdots & \sqrt{\frac{\gamma_{c_N}}{2m\pi \cos \theta_m}} \end{bmatrix}_{2m \times N} \quad (4.16)$$

which can be used to obtain the final form of $\mathbf{D}$ using the relationship in (4.8).

4.1.2 Generalisation of the solution for the inter resonant emitter coupling matrix

In order to obtain an analytical expression for Γ_{int} , we consider the power relationship with regard to the far field and inter-emitter coupling. In order to obey conservation laws, it is required to cease the generation of new thermal photons ($\mathbf{n} = 0$) and to cease the absorption of generated photons ($\gamma_a = 0$). These modifications are applied to (4.1) and used in the following derivations.

$$\frac{\partial}{\partial t} \mathbf{a}^* \mathbf{a} = \left[\frac{\partial}{\partial t} \mathbf{a}^* \right] \mathbf{a} + \mathbf{a}^* \left[\frac{\partial}{\partial t} \mathbf{a} \right], \quad (4.17a)$$

$$= \mathbf{a}^* (-j\omega_0 \mathbf{I} - (\Gamma_c + \Gamma_{int})) \mathbf{a} + \mathbf{a}^* (j\omega_0 \mathbf{I} - (\Gamma_c + \Gamma_{int})) \mathbf{a}, \quad (4.17b)$$

$$= \mathbf{a}^* (-2(\Gamma_c + \Gamma_{int})) \mathbf{a}. \quad (4.17c)$$

We now apply the same principle to the free-space model in Fig. 4.1 and obtain the following conservation relation:

$$\frac{\partial}{\partial t} \mathbf{a}^* \mathbf{a} = -\mathbf{S}^* \mathbf{S} = -\mathbf{a}^* \mathbf{D}^* \mathbf{D} \mathbf{a} \quad (4.18)$$

Comparing this result with (4.17c), the following relationship is derived:

$$\mathbf{D}^* \mathbf{D} = 2 (\mathbf{\Gamma}_c + \mathbf{\Gamma}_{int}). \quad (4.19)$$

Using values from (4.1) and (4.16), it is now possible to obtain an analytical solution for $[\Gamma_{int}]_{i,k}$ as follows:

$$[\Gamma_{int}]_{i,k} = \frac{1}{2} \sqrt{\gamma_{c_i} \gamma_{c_k}} (1 - \delta_{i,k}) = \frac{1}{2} \begin{bmatrix} 0 & \sqrt{\gamma_{c_1} \gamma_{c_2}} & \cdots & \sqrt{\gamma_{c_1} \gamma_{c_N}} \\ \sqrt{\gamma_{c_2} \gamma_{c_1}} & 0 & \cdots & \sqrt{\gamma_{c_2} \gamma_{c_N}} \\ \vdots & \vdots & \ddots & \vdots \\ \sqrt{\gamma_{c_N} \gamma_{c_1}} & \sqrt{\gamma_{c_N} \gamma_{c_1}} & \cdots & 0 \end{bmatrix} \quad (4.20)$$

4.2 Derivation of Emission Cross Sections (ECS)

In order to benchmark the system characteristics of our emitter assembly, we rigorously obtain an analytical expression for the total thermal energy of the system as a function of angular frequency (ω) using first principles [70,75,76]. An emitter system of this nature comprising N nanophotonic emitters is usually characterized by the emission cross section (ECS) denoted by σ_ω . When coupled with the blackbody spectral density, the total emission from the assembly can be expressed as:

$$P(\omega) = \left(\frac{\hbar \omega^2}{4\pi^2 c (\exp(\frac{\hbar \omega}{k_B T}) - 1)} \right) \sigma(\omega), \quad (4.21)$$

where c is the velocity of light in free space and T is the temperature. The constants $\hbar$ and k_B denote the reduced Plank constant and Boltzmann constant, respectively. It is noted that the equation represents the observed energy value in Joules, which is equivalent to the time integral of the received power [34].

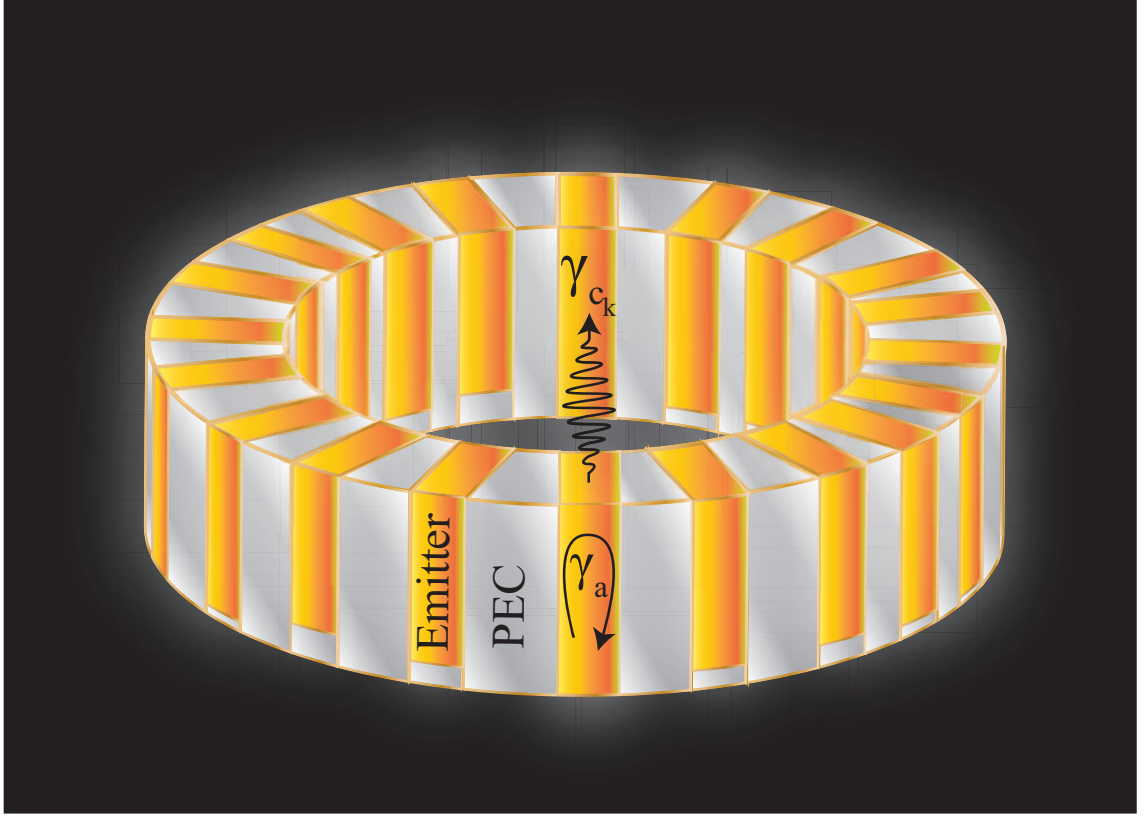


Figure 4.2: Conceptual design for a semi-generalized, ring-shaped superradiant emitter assembly with twenty nanophotonic emitters placed within subwavelength proximity. This structure of the emitter assembly can be engineered using PEC deep subwavelength slits. The absorption rate of the emissive material within each nanophotonic emitter is γ_a and the far field coupling constant of the k^{th} emitter is γ_{c_k} . Note that because the emitters in the assembly have different dimensions, some emitters do not traverse the entire thickness of the PEC material, thus resulting in a gap between the bottom surface of the ring and the emitter.

4.2.1 Semi-generalized emission cross section

We derive an analytical equation for the emission cross section of the novel emitter by considering spectral relations. Considering the physical nature of coupling constants and emitter amplitudes, it is possible to simplify the relations using:

$$\forall_{i,k}(a_k, D_{i,k}) \in \mathbb{R} \Rightarrow \mathbf{D}^* = \mathbf{D}^T. \quad (4.22)$$

Total power emission by all channels across the spectrum is then given by:

$$\langle P_t \rangle = \langle \mathbf{S}^* \mathbf{S} \rangle = \langle \mathbf{a}^T \mathbf{D}^T \mathbf{D} \mathbf{a} \rangle. \quad (4.23)$$

As shown in Fig. 4.1, now we consider the coupling of the k^{th} emitter to an arbitrary q^{th} channel radiating at an angle ϕ where the channel spectrum is given by:

$$\langle P_{t,\phi}^q \rangle = \sum_{i=1}^N \sum_{k=1}^N \langle a_i^*(t) D_{i,q}^T D_{q,k} a_k(t) \rangle. \quad (4.24)$$

The value of $\sum_{i=1}^N \sum_{k=1}^N D_{i,q}^T D_{q,k}$ can be readily found using (4.16) as follows:

$$\sum_{i=1}^N \sum_{k=1}^N D_{i,q}^T D_{q,k} = \sum_{i=1}^N \sum_{k=1}^N \frac{\sqrt{\gamma_{c_i} \gamma_{c_k}}}{2m\pi \cos \phi}. \quad (4.25)$$

Assuming that the thermal source has a stationary characteristic given by:

$$\frac{d}{dt} |n(t)|^2 = 0, \quad (4.26)$$

it is possible to obtain the following relationship [68,70]:

$$\langle a_i^*(t) a_k(t) \rangle = \int_0^\infty d\omega \int_0^\infty d\omega' A_{i,k}, \quad (4.27)$$

where:

$$A_{i,k} = e^{-j(\omega - \omega')t} \langle a_i^*(\omega) a_k(\omega') \rangle. \quad (4.28)$$

Applying the results from (4.25) and (4.27) in (4.24) yields the following expression:

$$\langle P_{t,\phi}^q \rangle = \sum_{i=1}^N \sum_{k=1}^N \int_0^\infty d\omega \int_0^\infty \frac{\sqrt{\gamma_{c_i} \gamma_{c_k}}}{2m\pi \cos \phi} A_{i,k} d\omega'. \quad (4.29)$$

Now it is possible to compare the frequency spectrum relation for the total

emission power of the channel:

$$\langle P_{t,\phi}^q \rangle = \int_0^\infty P_{\omega,\phi}^q d\omega, \quad (4.30)$$

with the result obtained in (4.29). This allows us to explicitly obtain an expression for the channel power spectral density as follows:

$$P_{\omega,\phi}^q = \sum_{i=1}^N \sum_{k=1}^N \int_0^\infty \frac{\sqrt{\gamma_{c_i} \gamma_{c_k}}}{2m\pi \cos \phi} A_{i,k} d\omega'. \quad (4.31)$$

In order to solve (4.31) analytically, it is necessary to obtain an expression for $\langle a_i^*(\omega) a_k(\omega') \rangle$ using the relationship given for thermal sources in (4.31). This is readily achieved by considering the time-varying dependence of emitter amplitudes given by $\mathbf{a}_{\omega,t} \approx \mathbf{a}_0 e^{j\omega t}$ where matrix:

$$\mathbf{a}_0 = [a_{0_1}, a_{0_2}, \dots, a_{0_N}]^T, \quad (4.32)$$

denotes the absolute amplitudes of the emitters. Note that this is a valid assumption in the weakly coupled regime for lossless emitter assemblies [63]. This allows us to formulate the following relationship:

$$\frac{\partial}{\partial t} \mathbf{a}_{\omega,t} \approx j\omega \mathbf{a}_{\omega,t}. \quad (4.33)$$

Substituting the result from (4.1) in (4.33) yields the following for $\mathbf{a}_{\omega,t}$:

$$\mathbf{a}_{\omega,t} \approx \left[\left(j(\omega - \omega_0) + \frac{\gamma_a}{2} \right) \mathbf{I} + \mathbf{\Gamma}_c + \mathbf{\Gamma}_{int} \right]^{-1} \sqrt{\gamma_a} \mathbf{n}. \quad (4.34)$$

The inverse portion of (4.34) can be obtained using the Ken-Miller lemma [77]. Application of this mathematical lemma is subject to the necessary condition:

$$\text{rank}(\mathbf{\Gamma}_c + \mathbf{\Gamma}_{int}) = 1. \quad (4.35)$$

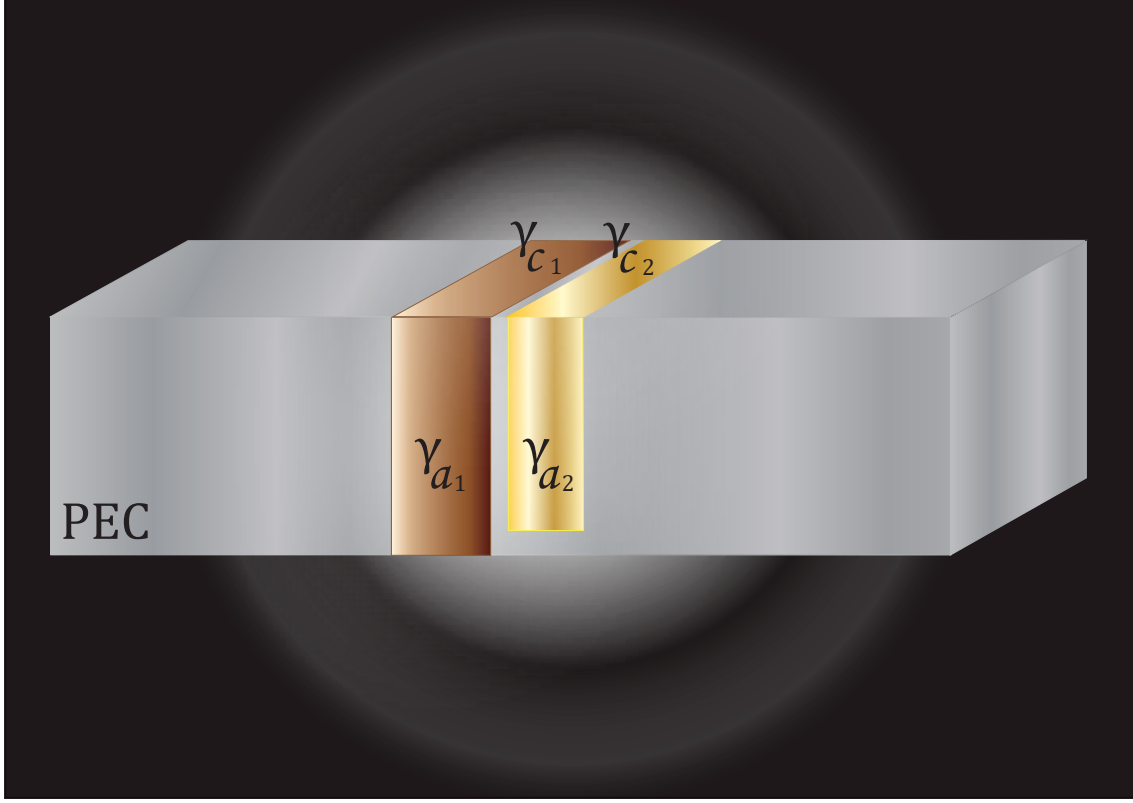


Figure 4.3: Simplified configuration shown comprises only two thermal emitters, each with a unique material and a dimensional configuration. This emitter assembly is best realized using PEC deep subwavelength slits. The emitters have unique absorption rates (γ_{a_1} and γ_{a_2}) and a far-field coupling rates (γ_{c_1} and γ_{c_2}). The various dimensions aid in retaining a unique resonance frequency across the emitters. The emission cross section for this assembly is analytically derived in eq. (4.70).

Combining the general result in (4.19) with (4.16) allows us to obtain an expression for $\mathbf{\Gamma}_C + \mathbf{\Gamma}_{int}$, on which we can perform Gaussian-Jordan eliminations to obtain the row-canonical form of the matrix, which readily proves that the necessary *rank* condition is satisfied [78] and therefore, the lemma is applicable.

Defining the inverse portion of (4.34) as $\mathcal{R}$ and applying the lemma yields the following:

$$\mathbf{K} = \left[\left(j(\omega - \omega_0) + \frac{\gamma_a}{2} \right) \mathbf{I} + \mathbf{\Gamma}_c + \mathbf{\Gamma}_{int} \right]^{-1}, \quad (4.36)$$

and applying the lemma derives the following result,

$$\mathcal{R} = \frac{1}{2(\mathcal{A} + j\Omega)(\mathcal{X} + j\Omega)} \left[(\mathcal{X} + j\Omega)\mathbf{I} - \mathbf{D}^T \mathbf{D} \right], \quad (4.37)$$

where Ω is the detuning parameter given by $\omega - \omega_0$ and,

$$\mathcal{A} = \frac{\gamma_a}{2}, \quad (4.38a)$$

$$\mathcal{X} = \mathcal{A} + \sum_{i=1}^N \frac{\gamma_{c_i}}{2}. \quad (4.38b)$$

The result in (4.37) can now be applied to:

$$\mathbf{a}_{\omega,t} \approx \sqrt{\gamma_a} \mathcal{R} \mathbf{n}, \quad (4.39)$$

and substituted in (4.31) to obtain the following result:

$$P_{\omega,\phi}^q \approx \sum_{i=1}^N \sum_{k=1}^N \int_0^\infty \frac{\sqrt{\gamma_{c_i} \gamma_{c_k}}}{2m\pi \cos \phi} \gamma_a \mathcal{R}_{i,u}^* \mathcal{R}_{k,v} N_{i,k} d\omega', \quad (4.40)$$

where:

$$N_{i,k} = e^{-j(\omega - \omega')t} \langle n_i^*(\omega) n_k(\omega') \rangle. \quad (4.41)$$

Substituting the values from (4.31) now yields the following:

$$P_{\omega,\phi}^q \approx \frac{\Theta_{\omega T}}{2\pi} \frac{\gamma_a}{2m\pi \cos \phi} \sum_{i=1}^N \sum_{k=1}^N \sum_{u=1}^N \sum_{v=1}^N \int_0^\infty \mathcal{R}_{i,u}^* \mathcal{R}_{k,v} \times \delta(\omega - \omega') e^{-j(\omega - \omega')t} \sqrt{\gamma_{c_i} \gamma_{c_k}} \delta_{u,v} d\omega'. \quad (4.42)$$

In order to obtain an expression for the power spectrum of all channels, it is necessary to decouple the channel dependency in (4.42). This can be done by normalizing the total emitted power in the channel by the flux density of a single channel. This is a valid assumption for an isotropic radiator and the channel flux

density is given by [34, 69]:

$$\mathcal{CFD} = \frac{\Theta_{\omega T}}{2\pi L \cos \phi}. \quad (4.43)$$

The total emission cross section for the emitter assembly has the following form:

$$\sigma_{\omega} = \int_0^{2\pi} \frac{P_{\omega, \phi}^q}{\frac{\Theta_{\omega T}}{2\pi} / L \cos \phi} d\phi, \quad (4.44)$$

which allows us to obtain the following relation for σ_{ω} :

$$\sigma_{\omega} \approx \frac{2\pi c}{\omega_0} \gamma_a \sum_{i=1}^N \sum_{k=1}^N \sum_{u=1}^N \mathcal{R}_{i,u}^* \mathcal{R}_{k,u} \sqrt{\gamma_{c_i} \gamma_{c_k}}, \quad (4.45)$$

where c is the velocity of light in free-space.

By substituting the values for $\mathcal{R}_{k,u}$ and its *c.c.* from (4.37), it is possible to derive the final equation for the emission cross section of the novel emitter assembly as follows:

$$\begin{aligned} \sigma_{\omega} \approx & \frac{\pi c}{\omega_0} \left[\frac{\gamma_a}{2} \sum_{i=1}^N \gamma_{c_i} \right] \frac{4 \left(\left[\frac{\gamma_a}{2} + \sum_{i=1}^N \frac{\gamma_{c_i}}{2} \right]^2 + (\omega - \omega_0)^2 \right)}{\left(\left[\frac{\gamma_a}{2} \right]^2 + (\omega - \omega_0)^2 \right) \left(\left[\frac{\gamma_a}{2} + \sum_{i=1}^N \frac{\gamma_{c_i}}{2} \right]^2 + (\omega - \omega_0)^2 \right)} \\ & + \frac{\sum_{i=1}^N \left[\gamma_{c_i}^2 - 4\gamma_{c_i} \left[\frac{\gamma_a}{2} + \sum_{j=1}^N \frac{\gamma_{c_j}}{2} \right] \right] + 2 \sum_{i=1}^{N-1} \sum_{j=i+1}^N [\gamma_{c_i} \gamma_{c_j}]}{\left(\left[\frac{\gamma_a}{2} \right]^2 + (\omega - \omega_0)^2 \right) \left(\left[\frac{\gamma_a}{2} + \sum_{i=1}^N \frac{\gamma_{c_i}}{2} \right]^2 + (\omega - \omega_0)^2 \right)}. \end{aligned} \quad (4.46)$$

The total emission power of the emitter assembly can now be obtained by multiplying the emission cross section in (4.46) with the blackbody spectral den-

sity $\mathcal{BB}_{psd}$ [79], which yields:

$$P_\omega = \mathcal{BB}_{psd} \sigma_\omega = \left(\frac{\omega^4}{8\pi^3 c^3} \Theta_{\omega,T} \right) \sigma_\omega. \quad (4.47)$$

4.2.2 Emission cross section for a conventional emitter assembly

Now we focus on proving that our generalized results are valid for the special case of emitter assemblies comprising of identical emitters, such as the conventional model discussed in [34]. For this setting, all emitters have the same coupling constant value which we denote by γ_c .

This assumption leads to the simplification of the expression:

$$\Gamma_c = \frac{\gamma_c}{2} \mathbf{I}. \quad (4.48)$$

Based on this simplification, we can obtain the following mathematical expressions that are required to reenforce this assumption to our emitter assembly,

$$\sum_{i=1}^N \gamma_{c_i} = N\gamma_c, \quad (4.49a)$$

$$2 \sum_{i=1}^{N-1} \sum_{j=i+1}^N [\gamma_{c_i} \gamma_{c_j}] = N(N-1)\gamma_c^2. \quad (4.49b)$$

Now these two simplifications are applied to (4.46), which yields the cross section for the conventional model as follows: This yields the cross section for the conventional model as follows:

$$\sigma_\omega \approx \frac{2\pi c}{\omega_0} \frac{N\gamma_a \gamma_c}{(\omega - \omega_0)^2 + \left(\frac{N\gamma_c + \gamma_a}{2} \right)^2}. \quad (4.50)$$

The result obtained in (4.50) is identical to the cross section given in [34],

which proves the mathematical consistency of our calculations. This result can be substituted in (4.47) in order to obtain the total emission power relation given in [34].

The most significant result derived in [34] shows the contrast between non-resonant and conventional resonant emitter assemblies. For non-resonant and superradiant emitter assemblies of N emitters each, the total power emission at resonance ($\omega = \omega_0$) can be obtained by applying the non-resonant condition ($\gamma_a \gg \gamma_c$) and the superradiant condition ($\gamma_c \gg \gamma_a$) in (4.50), which yield the following results:

$$\mathcal{N}on-Resonant P_\omega \approx \frac{2}{\pi} \Theta_{\omega,T} \frac{\gamma_c}{\gamma_a} N, \quad (4.51a)$$

$$\mathcal{S}uperradiant P_\omega \approx \frac{2}{\pi} \Theta_{\omega,T} \frac{\gamma_a}{\gamma_c} \frac{1}{N}. \quad (4.51b)$$

It is observed that non resonant emitter assemblies, as expected, show a linear power scaling at resonance whereas conventional superradiant emitter assemblies show an anomalous power scaling by a factor of $1/N$.

4.2.3 Emission cross section of a fully generalized emitter assembly

In order to obtain an expression for the emission cross section of our proposed design, it is required to observe spectral variations of the system. We begin by considering the same coupling between the arbitrary k th emitter and the radiation channel at angle ϕ as depicted in fig. 4.1. The power spectrum of the channel ($P_{\omega,\phi}$) can be defined using the expected value of time-varying energy relationship for the model given in eq. (4.51). For sake of simplicity, we denote the arbitrary p th element of $\mathbf{a}_{\omega,t}$ by $a_p(\omega, t)$, its expected value in the time domain by $\langle a_p(t) \rangle$ and its expected value in the angular frequency domain by $\langle a_p(\omega) \rangle$. This

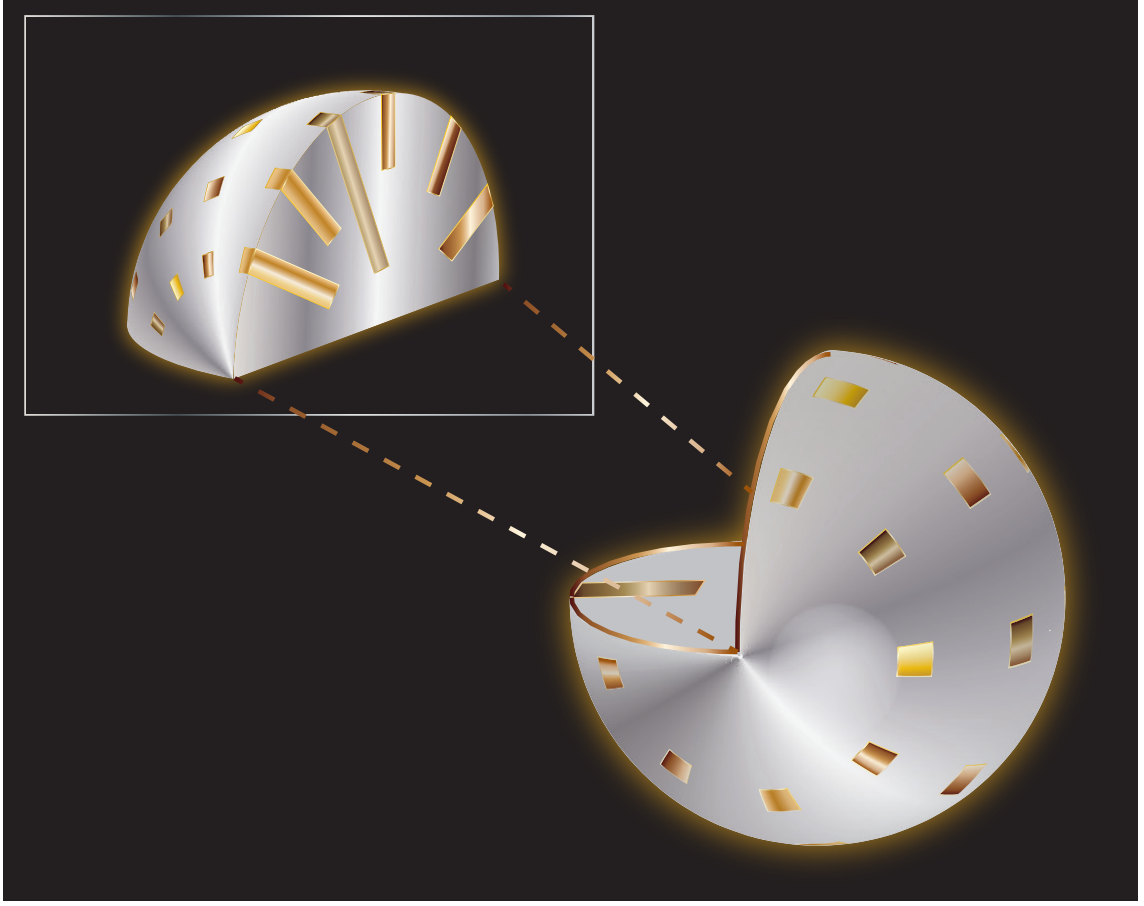


Figure 4.4: The configuration shown comprises many nanophotonic thermal emitters engulfed in a spherical, near-PEC body. Each emitter can be uniquely characterized using a specific absorptive material and dimensions, thus giving the ability to uniquely define (γ_a) and (γ_c) for each emitter. The cross-section shown in the inset shows how the emitters unevenly penetrate towards the centre of the sphere, thus capturing the dimensional variability between the emitters. It is noted that the emitters are arranged symmetrically on the surface of the sphere such that a given emitter always forms a part of a hypothetical ring that spans the surface of the sphere. Symmetry akin to this nature is vital to attain the effects of superradiance when non-identical emitters are used.

notation allows us to formulate as follows:

$$\int_0^\infty P_{\omega,\phi} d\omega = \langle P_{t,\phi} \rangle = \sum_{p=1}^N \sum_{q=1}^N \langle a_p^*(t) [\mathcal{C}_{p,\phi}^* \mathcal{C}_{\phi,q}] a_q(t) \rangle. \quad (4.52)$$

Substituting values from eq. (4.51) yields:

$$\int_0^\infty P_{\omega,\phi} d\omega = \langle a_p^*(t) a_q(t) \rangle \sqrt{\tilde{\gamma}_{c_p} \tilde{\gamma}_{c_q}} \sec \phi. \quad (4.53)$$

An expression for the expected value of electromagnetic amplitudes:

$$\langle a_p^*(t) a_q(t) \rangle, \quad (4.54)$$

can be obtained from the literature on fluctuation dissipation theorem when the used noise sources illustrate wide sense stationary characteristics [70]:

$$\langle a_p^*(t) a_q(t) \rangle = \int_0^\infty d\omega \times \left(\int_0^\infty e^{-j(\omega-\omega')t} \langle a_p^*(\omega) a_q(\omega') \rangle d\omega' \right). \quad (4.55)$$

Direct association of eq. (4.53) and eq. (4.55) yields the following expression for the channel power spectral density:

$$P_{\omega,\phi} = \sum_{p=1}^N \sum_{q=1}^N \int_0^\infty \sqrt{\tilde{\gamma}_{c_p} \tilde{\gamma}_{c_q}} \sec \phi \times e^{-j(\omega-\omega')t} \langle a_p^*(\omega) a_q(\omega') \rangle d\omega'. \quad (4.56)$$

Obtaining an expression for $\langle a_p^*(\omega) a_q(\omega') \rangle$ is possible by using eq. (4.31). It is noted that it is possible to apply CMT to analyze systems that are both in the weakly-coupled and strongly-coupled regimes [80]. However, our proposed model is exclusively studied in the weakly-coupled regime. Therefore, without losing generality, for an ideal emitter assembly operating within the weakly coupled regime, it is possible to assume that the electromagnetic amplitude $\mathbf{a}_{\omega,t} \approx \tilde{\mathbf{a}} e^{j\omega t}$, where $\tilde{\mathbf{a}}$ denotes the slow varying component of the emitter amplitudes. Using the partial differential operator yields the expression:

$$\frac{\partial}{\partial t} \mathbf{a}_{\omega,t} \approx j\omega \mathbf{a}_{\omega,t}. \quad (4.57)$$

Under this assumption, the following can be obtained using eq. (4.1):

$$\mathbf{a}_{\omega,t} \approx \mathcal{R} \sqrt{\Gamma_{\mathbf{a}}} \mathbf{n}, \quad (4.58)$$

where:

$$\mathcal{R} = [j(\omega - \omega_0)\mathbf{I} + \Gamma_{\mathbf{a}} + \Gamma_{\mathbf{c}} + \Gamma_{\text{int}}]^{-1}. \quad (4.59)$$

Now it is possible to rewrite eq. (4.56) as follows:

$$P_{\omega,\phi} \approx \sum_{p=1}^N \sum_{q=1}^N \int_0^\infty \frac{\sqrt{\tilde{\gamma}_{c_p} \tilde{\gamma}_{c_q}}}{e^{j(\omega - \omega')t} \cos \phi} \times \sum_{u=1}^N \sum_{v=1}^N \left(\mathcal{R} \sqrt{\Gamma_{\mathbf{a}}} \right)_{p,u}^* \left(\mathcal{R} \sqrt{\Gamma_{\mathbf{a}}} \right)_{q,v} \times \langle n_p^*(\omega) n_q(\omega') \rangle d\omega'. \quad (4.60)$$

Mathematically, since $\{\Gamma_{\mathbf{c}}, \Gamma_{\text{int}}\} \in \mathbb{R}^+$ by definition, $P_{\omega,\phi} \in \mathbb{R}^+$. When substituting from eq. (4.31), the effect of the $e^{j(\omega - \omega')t}$ component is nullified due to the presence of $\delta(\omega - \omega')$.

Further simplification yields the following expression:

$$P_{\omega,\phi} \approx \frac{\Theta_{\omega T}}{2\pi L \cos \phi} \frac{\lambda_0}{2\pi} \sum_{p=1}^N \sum_{q=1}^N \sum_{u=1}^N (\mathcal{R} \sqrt{\Gamma_{\mathbf{a}}})_{p,u}^* \times (\mathcal{R} \sqrt{\Gamma_{\mathbf{a}}})_{q,u} \sqrt{\gamma_{c_p} \gamma_{c_q}} \quad (4.61)$$

The expression in eq. (4.42) depicts the power spectrum of the assembly for a single channel. In order to obtain an expression for the power spectrum of the assembly for free-space, we need to find the emission cross section of the assembly. This is readily achieved by normalizing eq. (4.42) with the channel flux density [69], which yields:

$$N_{\sigma}(\omega) \approx \frac{c}{\omega_0} \sum_{p=1}^N \sum_{q=1}^N \sum_{u=1}^N \tilde{\mathcal{R}}_{p,u}^*(\omega) \tilde{\mathcal{R}}_{q,u}(\omega) \sqrt{\gamma_{c_p} \gamma_{c_q}}, \quad (4.62)$$

where ω_0 denotes the resonance frequency of the assembly. The matrix $\tilde{\mathcal{R}}(\omega)$ is

of the form:

$$\tilde{\mathcal{R}}(\omega) = [j(\omega - \omega_0)\mathbf{I} + \mathbf{\Gamma}_a + \mathbf{\Gamma}_c + \mathbf{\Gamma}_{int}]^{-1} \sqrt{\mathbf{\Gamma}_a}, \quad (4.63)$$

where $\mathbf{I}$ denotes the $N \times N$ identity matrix.

Here,

$$\mathbf{\Gamma}_c = \frac{1}{2} \text{diag} [\gamma_{c_1} \dots \gamma_{c_N}]_{N \times N}, \quad (4.64)$$

denotes the photon far-field coupling rates and

$$\mathbf{\Gamma}_a = \frac{1}{2} \text{diag} [\gamma_{a_1} \dots \gamma_{a_N}]_{N \times N}, \quad (4.65)$$

denotes the photon absorption rates for all emitters in the assembly.

The inter-resonant emitter indirect coupling rates ($\mathbf{\Gamma}_{int}$) essentially capture the presence of superradiance in the assembly and is of the form:

$$[\mathbf{\Gamma}_{int}]_{p,q} = \frac{1}{2} \sqrt{\gamma_{c_p} \gamma_{c_q}} (1 - \delta_{pq}), \quad (4.66)$$

where $\{2 \leq (p, q) \leq N \cap (p, q) \in \mathbb{Z}^+\}$, as derived earlier in this chapter.

Assuming that the assembly resembles the characteristics of a blackbody during emission to the far field, the emission power spectral characteristics of the model can now be obtained using (4.45) and the blackbody spectral density: $\mathcal{BB}_{psd}$ [81]:

$$P_\omega = \mathcal{BB}_{psd} \sigma_\omega = \left(\frac{\omega^4}{8\pi^3 c^3} \Theta_{\omega, T} \right) \sigma_\omega. \quad (4.67)$$

In order to obtain an analytical expression for the $N = 2$ case shown in fig. 4.3, we begin by defining the parameters:

$$\mathbf{\Gamma}_c = \frac{1}{2} \text{diag} [\gamma_{c_1}, \gamma_{c_2}], \quad (4.68a)$$

$$\mathbf{\Gamma}_a = \frac{1}{2} \text{diag} [\gamma_{a_1}, \gamma_{a_2}], \quad (4.68b)$$

$$\mathbf{\Gamma}_{int} = \frac{1}{2} \begin{bmatrix} 0 & \sqrt{\gamma_{c_2}\gamma_{c_1}} \\ \sqrt{\gamma_{c_1}\gamma_{c_2}} & 0 \end{bmatrix}, \quad (4.68c)$$

$$\mathbf{R} = \frac{2}{\mathcal{K}} \begin{bmatrix} -2j\Omega - (\gamma_{a_2} + \gamma_{c_2}) & \sqrt{\gamma_{c_2}\gamma_{c_1}} \\ \sqrt{\gamma_{c_1}\gamma_{c_2}} & -2j\Omega - (\gamma_{a_1} + \gamma_{c_1}) \end{bmatrix}, \quad (4.68d)$$

where:

$$\mathcal{K} = 4\Omega^2 - 2j(\gamma_{a_1} + \gamma_{c_1} + \gamma_{a_2} + \gamma_{c_2})\Omega - (\gamma_{a_1}\gamma_{c_2} + \gamma_{c_1}\gamma_{a_2} + \gamma_{a_1}\gamma_{a_2}). \quad (4.69)$$

Substituting values from eq. (4.68a - 4.68d) in eq. (4.45) and simplifying generates the following recipe:

$$^2\sigma(\omega) \approx \frac{4c}{\omega_0} \frac{\gamma_{a_1}\gamma_{c_1}(4\Omega^2 + \gamma_{a_2}^2) + \gamma_{a_2}\gamma_{c_2}(4\Omega^2 + \gamma_{a_1}^2)}{16\Omega^4 + 4\Omega^2\mathcal{F}_1 + \mathcal{F}_2}, \quad (4.70)$$

where:

$$\Omega = \omega - \omega_0, \quad (4.71)$$

is the detuning parameter and the factors:

$$\mathcal{F}_1 = [(\gamma_{a_1} + \gamma_{c_1})^2 + (\gamma_{a_2} + \gamma_{c_2})^2 + 2\gamma_{c_1}\gamma_{c_2}], \quad (4.72)$$

$$\mathcal{F}_2 = (\gamma_{a_1}\gamma_{a_2} + \gamma_{c_1}\gamma_{a_2} + \gamma_{a_1}\gamma_{c_2})^2. \quad (4.73)$$

and

$$^2\sigma_\omega \approx \frac{c}{\omega_0} \times \frac{4\gamma_{a_1}\gamma_{c_1}(4\Omega^2 + \gamma_{a_2}^2) + 4\gamma_{a_2}\gamma_{c_2}(4\Omega^2 + \gamma_{a_1}^2)}{16\Omega^4 + 4\Omega^2[(\gamma_{a_1} + \gamma_{c_1})^2 + (\gamma_{a_2} + \gamma_{c_2})^2 + 2\gamma_{c_1}\gamma_{c_2}] + [\gamma_{a_1}\gamma_{a_2} + \gamma_{c_1}\gamma_{a_2} + \gamma_{a_1}\gamma_{c_2}]^2} \quad (4.74)$$

Following a similar approach, it is possible to obtain the following lengthy recipe for the $N = 3$ case:

$${}^3\sigma_\omega \approx \frac{4c}{\omega_0} \frac{\gamma_{a_1}\gamma_{c_1}\mathcal{G}_1 + \gamma_{a_2}\gamma_{c_2}\mathcal{G}_2 + \gamma_{a_3}\gamma_{c_3}\mathcal{G}_3}{64\Omega^6 + 16\Omega^4\mathcal{G}_4 + 4\Omega^2\mathcal{G}_5 + \mathcal{G}_6}, \quad (4.75)$$

where,

$$\mathcal{G}_1 = 16\Omega^4 + 4(\gamma_{a_2} + \gamma_{a_3})\Omega^2 + \gamma_{a_2}^2\gamma_{a_3}^2, \quad (4.76a)$$

$$\mathcal{G}_2 = 16\Omega^4 + 4(\gamma_{a_1} + \gamma_{a_3})\Omega^2 + \gamma_{a_1}^2\gamma_{a_3}^2, \quad (4.76b)$$

$$\mathcal{G}_3 = 16\Omega^4 + 4(\gamma_{a_1} + \gamma_{a_2})\Omega^2 + \gamma_{a_1}^2\gamma_{a_2}^2, \quad (4.76c)$$

$$\begin{aligned} \mathcal{G}_4 = & (\gamma_{a_1} + \gamma_{c_1})^2 + (\gamma_{a_2} + \gamma_{c_2})^2 + (\gamma_{a_3} + \gamma_{c_3})^2 + \\ & 2(\gamma_{c_1}\gamma_{c_2} + \gamma_{c_1}\gamma_{c_3} + \gamma_{c_2}\gamma_{c_3}), \end{aligned} \quad (4.76d)$$

$$\mathcal{G}_5 = (\gamma_{a_1}\gamma_{a_2} + \gamma_{c_1}\gamma_{a_2} + \gamma_{a_1}\gamma_{c_2})^2 + (\gamma_{a_1}\gamma_{a_3} + \gamma_{c_1}\gamma_{a_3} + \gamma_{a_1}\gamma_{c_3})^2 + \mathcal{G}_7, \quad (4.76e)$$

$$\mathcal{G}_6 = [\gamma_{a_1}\gamma_{a_2}\gamma_{a_3} + \gamma_{c_1}\gamma_{a_2}\gamma_{a_3} + \gamma_{a_1}\gamma_{c_2}\gamma_{a_3} + \gamma_{a_1}\gamma_{a_2}\gamma_{c_3}]^2, \quad (4.76f)$$

$$\mathcal{G}_7 = (\gamma_{a_2}\gamma_{a_3} + \gamma_{c_2}\gamma_{a_3} + \gamma_{a_2}\gamma_{c_3})^2 + 2(\gamma_{a_1}^2\gamma_{c_2}\gamma_{c_3} + \gamma_{c_1}\gamma_{a_2}^2\gamma_{c_3} + \gamma_{c_1}\gamma_{c_2}\gamma_{a_3}^2). \quad (4.76g)$$

Complete analytical solution

Next we obtain a complete analytical solution to the emission cross section for the case of N emitters. Simple substitution yields the following result for ${}^N\sigma_\omega$:

$$\begin{aligned} {}^N\sigma_\omega \approx & 2\tilde{\Omega} \sum_{p=1}^N \sum_{k=1}^N \sum_{q=1}^N \left[\frac{\delta_{pq}}{j\Omega - \sqrt{\mathcal{A}_p\mathcal{A}_q}} - \frac{\mathcal{K}\sqrt{\mathcal{C}_p\mathcal{C}_q}}{(j\Omega - \mathcal{A}_p)(j\Omega - \mathcal{A}_q)} \right] \\ & \times \left[\frac{\delta_{kq}}{j\Omega + \sqrt{\mathcal{A}_k\mathcal{A}_q}} - \frac{\mathcal{K}\sqrt{\mathcal{C}_k\mathcal{C}_q}}{(j\Omega + \mathcal{A}_k)(j\Omega + \mathcal{A}_q)} \right] \sqrt{\mathcal{C}_p\mathcal{C}_k\mathcal{A}_p\mathcal{A}_k}, \end{aligned} \quad (4.77)$$

where:

$$\mathcal{K} = \frac{1}{1 + \sum_{r=1}^N \frac{\mathcal{C}_r}{j\Omega + \mathcal{A}_r}}. \quad (4.78)$$

In order to simplify the mathematical complexity of the final recipe, we introduce two mathematical operators for positive integers. The index operator: ${}_x\mathbf{Y}^{\{X\}}$ where $(x \leq |X|)$, represents a matrix of dimensions $x \times \binom{|X|}{x}$, where each column uniquely represents a combination of x non-repeating elements from $\{X\}$, ordered in ascending order, such that all possible combinations are exhausted and the overall matrix follows an ascending pattern. The operator can be defined as:

$${}_x\mathbf{Y}^{\{X\}} = \prod_{k=1}^x \{\aleph_{i_k=i_{k-1}+1}^{|X|-x+k}\} [X_{i_1}, X_{i_2}, \dots, X_{i_x}]^T, \quad (4.79)$$

where $\aleph_{i=a}^b$ is the iterating operator from a to b where $a \leq b$ and $(a, b) \in \mathbb{Z}^+$. Furthermore, $\forall k \leq 0, i_k = 0$.

To illustrate this further, consider the following:

$${}_2\mathbf{Y}^{\{1,2,3,4\}} = \begin{bmatrix} 1 & 1 & 1 & 2 & 2 & 3 \\ 2 & 3 & 4 & 3 & 4 & 4 \end{bmatrix}, \quad (4.80a)$$

$${}_3\mathbf{Y}^{\{1,2,4,5,6\}} = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 & 2 & 2 & 2 & 4 \\ 2 & 2 & 2 & 4 & 4 & 5 & 4 & 4 & 5 & 5 \\ 4 & 5 & 6 & 5 & 6 & 6 & 5 & 6 & 6 & 6 \end{bmatrix}. \quad (4.80b)$$

Next we define the combination summation operator: ${}_{\star p_n} \Xi_{q_m}^{\{X\}}$ where $n + m \leq |X|$, which denotes all possible summations of patterns between n elements of p_* and m elements of q_* , ordered using the wildcard $\star$. The values of $\star$ are defined in the same order as $\{X\}$.

In order to formally define the combination summation operator for the general case, we begin by segmenting X into $\binom{|X|}{m}$ unique segments. Let the k th

unique segment be denoted by:

$$\{M_k\} = {}_m Y_{:,k}^{\{X\}}, \quad (4.81)$$

and by defining:

$$\{N_k\} = \{X \setminus M_k\}, \quad (4.82)$$

the following expression defines the operator:

$$\star p_n \Xi_{q_m}^{\{X\}} = \sum_{i=1}^{\binom{|X|}{m}} \left(\prod_{j=1}^m (q_{{}_m Y_{j,i}^{\{X\}}}) \left[\sum_{k=1}^{\binom{|X|}{n}} \prod_{l=1}^n p_{{}_n Y_{l,k}^{\{N_i\}}} \right] \right) \quad (4.83)$$

To illustrate its operation, consider the special case $n + m = |X|$:

$$\begin{aligned} \star p_3 \Xi_{q_2}^{\{1,2,3,4,5\}} &= q_1 q_2 p_3 p_4 p_5 + q_1 p_2 q_3 p_4 p_5 \\ &\quad + q_1 p_2 p_3 q_4 p_5 + q_1 p_2 p_3 p_4 q_5. \end{aligned} \quad (4.84)$$

and the more general case $n + m < |X|$

$$\begin{aligned} \star p_2 \Xi_{q_2}^{\{1,2,3,4,5\}} &= q_1 q_2 (p_3 p_4 + p_3 p_5 + p_4 p_5) \\ &\quad + q_1 q_3 (p_2 p_4 + p_2 p_5 + p_4 p_5) + q_1 q_4 (p_2 p_3 + p_2 p_5 + p_3 p_5) \\ &\quad + q_1 q_5 (p_2 p_3 + p_2 p_4 + p_3 p_4) + q_2 q_3 (p_1 p_4 + p_1 p_5 + p_4 p_5) \\ &\quad + q_2 q_4 (p_1 p_3 + p_1 p_5 + p_3 p_5) + q_2 q_5 (p_1 p_3 + p_1 p_4 + p_3 p_4) \\ &\quad + q_3 q_4 (p_1 p_2 + p_1 p_5 + p_2 p_5) + q_3 q_5 (p_1 p_2 + p_1 p_4 + p_2 p_4) \\ &\quad + q_4 q_5 (p_1 p_2 + p_1 p_3 + p_2 p_3) \end{aligned} \quad (4.85)$$

It is now possible to express the final form of the emission cross section as follows:

$${}^N\sigma_\omega \approx \tilde{\Omega} \frac{\sum_{n=1}^N \{ \mathcal{A}_n \mathcal{C}_n [\sum_{m=0}^{\mathcal{M}} (\omega - \omega_0)^{2m} (\sum_{p=1}^{\mathcal{P}} (\prod_{q=1}^{\mathcal{Q}} \mathcal{A}_{\mathcal{Q}Y_{q,p}^{\{\mathcal{N} \setminus n\}}}))] \} }{\sum_{n=1}^N \{ \Omega^{2n} (\sum_{r=1}^{\mathcal{R}} [\prod_{s=1}^{\mathcal{S}} \mathcal{A}_{\mathcal{S}Y_{s,r}^{\{\mathcal{N}\}}} + {}_{\star}\mathcal{A}_{\mathcal{S}-1} \Xi_{\mathcal{C}}^{\{\mathcal{S}Y_{:,r}^{\{\mathcal{N}\}}\}}])^2 + 2 {}_{\star}\mathcal{C}_2 \Xi_{\mathcal{A}_{\mathcal{S}-1}^2}^{\{\mathcal{N}\}} \} \} + \mathcal{Z}^2}, \quad (4.86)$$

where

$$\mathcal{Z} = \prod_{n=1}^N \mathcal{A}_n + {}_{\star}\mathcal{A}_{N-1} \Xi_{\mathcal{C}}^{\{\mathcal{N}\}} \quad (4.87)$$

and $\{\mathcal{N}\} = \{1, 2, \dots, N\}$, $\mathcal{M} = N - 1$, $\mathcal{Q} = \mathcal{M} - m$, $\mathcal{S} = N - n$, $\mathcal{P} = \binom{\mathcal{M}}{\mathcal{Q}}$ and $\mathcal{R} = \binom{N}{\mathcal{S}}$.

4.2.4 Analytical validation of derivations

The derived analytical result shown in eq. (4.70) and (4.75) are in complete agreement with existing work [34, 82] and it is possible to reproduce existing results by applying restrictive assumptions. As a proof of mathematical validity of our model, we now apply the restrictive assumptions into eq. (4.70) and eq. (4.75) and replicate the published analytical solutions in [82] and [34].

We begin by applying the material restriction where:

$$\gamma_a = \gamma_{a_1} = \gamma_{a_2}. \quad (4.88)$$

Our recipe for $N = 2$ resolves to:

$${}^2\sigma_{\omega,1} \approx \frac{4\gamma_a c}{\omega_0} \times \frac{(4\Omega^2 + \gamma_{a_2}^2)(\gamma_{c_1} + \gamma_{c_2})}{16\Omega^4 + 4\Omega^2[(\gamma_a + \gamma_{c_1})^2 + (\gamma_a + \gamma_{c_2})^2 + 2\gamma_{c_1}\gamma_{c_2}] + \gamma_a^2[\gamma_a + \gamma_{c_1} + \gamma_{c_2}]^2}, \quad (4.89a)$$

$${}^2\sigma_{\omega,1} \approx \frac{4\gamma_a c}{\omega_0} \frac{(4\Omega^2 + \gamma_a^2)(\gamma_{c_1} + \gamma_{c_2})}{(4\Omega^2 + \gamma_a^2)(4\Omega^2 + (\gamma_a + \gamma_{c_1} + \gamma_{c_2})^2)}, \quad (4.89b)$$

$${}^2\sigma_{\omega,1} \approx \frac{c}{\omega_0} \frac{4\gamma_a(\gamma_{c_1} + \gamma_{c_2})}{(4\Omega^2 + (\gamma_a + \gamma_{c_1} + \gamma_{c_2})^2)}. \quad (4.89c)$$

which is in agreement with the observation in [82].

Next we apply the dimensional restriction where:

$$\gamma_c = \gamma_{c_1} = \gamma_{c_2}. \quad (4.90)$$

The recipe in eq. (4.89c) further resolves to:

$${}^2\sigma_{\omega,2} \approx \frac{c}{\omega_0} \frac{8\gamma_a\gamma_c}{(4\Omega^2 + (\gamma_a + 2\gamma_c)^2)}, \quad (4.91)$$

which is in agreement with the observation in [34].

A similar approach proves the validity of the model for the $N = 3$ case as well where the obtained results are:

$${}^3\sigma_{\omega,1} \approx \frac{c}{\omega_0} \frac{4\gamma_a(\gamma_{c_1} + \gamma_{c_2} + \gamma_{c_3})}{(4\Omega^2 + (\gamma_a + \gamma_{c_1} + \gamma_{c_2} + \gamma_{c_3})^2)}, \quad (4.92a)$$

$${}^3\sigma_{\omega,2} \approx \frac{c}{\omega_0} \frac{12\gamma_a\gamma_c}{(4\Omega^2 + (\gamma_a + 3\gamma_c)^2)}. \quad (4.92b)$$

4.3 Tuning the Resonance Frequency

It is observed that in order to deliver power optimally, it is necessary to tune the resonant frequency of the emitter. There is a vast availability of literature in the study of deep subwavelength slits that addresses the resonant frequency tuning problem and equation (4.95) is one of the major results.

Our emitter assembly assumes that all emitters within it have the same absorption constant γ_a . Within the weak field regime, this can be achieved by using an emitter array made using the same emissive material, which ensures that all

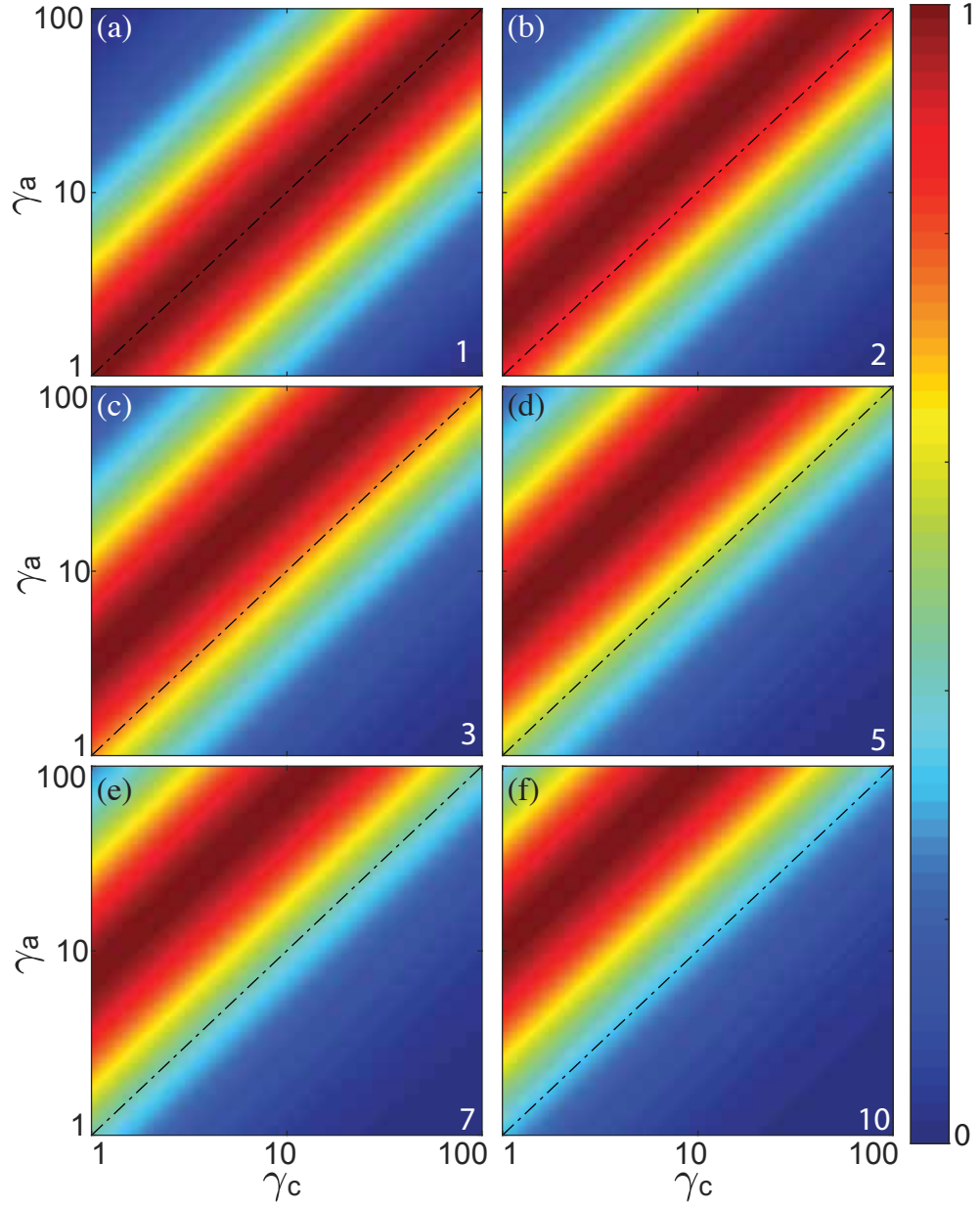


Figure 4.5: Emission power of resonant emitter assemblies of (a) 1, (b) 2, (c) 3, (d) 5, (e) 7, and (f) 10 emitters in subwavelength proximity. The dashed lines represent the maximum power delivery point a non-resonant emitter assembly with the same number of emitters. It is observed that a higher number of emitters deliver less power at resonance when the principles of superradiance are taken into account.

emitters will have the same dielectric constant [83]. However, the superradiant phenomena, as observed in (4.20), depends solely on the far field coupling rates of the emitters. Therefore, the success of our proposed assembly depends on the individual tunability of this parameter in each emitter. Furthermore, it is vital to maintain the emitter assembly in the over-coupling regime, defined by:

$$\left(\frac{1}{N} \sum_{i=1}^N \gamma_{c_k} \gg \gamma_a \right), \quad (4.93)$$

in order to sustain the superradiant phenomena [80]. While these might appear as straightforward requirements, several design challenges need to be overcome to achieve a successful realization.

The main issue is with the design of the individual emitter unit. For the 2-dimensional case, it can be assumed that an assembly of ideal Fabry-Perot cavity emitters with a constant height will have γ_c values proportional to their widths [34]. However, if the slit width is varied, it is no longer possible to assume that the resonant frequency ω_0 is constant to all emitters [84]. This detuning effect needs to be addressed in order to maintain the functionality of our emitter assembly.

For their emitter model, Zhou *et al.* [34] have used an ideal Fabry-Perot resonant cavity. As shown in Fig. 4.6(a) and Fig. 4.6(c), for an ideal Fabry-Perot cavity with 1-dimensional control, resonant frequencies are governed by the equation:

$$f_m = \frac{mc}{2nL}, \quad (4.94)$$

where c is the velocity of light in free-space, L is the cavity length, n is the refractive index of the cavity and m is the resonant mode. The cavity resonant mode can directly be tuned by changing the length of the cavity. This means that for an ensemble of emitters with varying far-field coupling parameters, a unique resonant frequency cannot be maintained with 1-dimensional control. Therefore, this simplified emitter model fails to facilitate our design criteria.

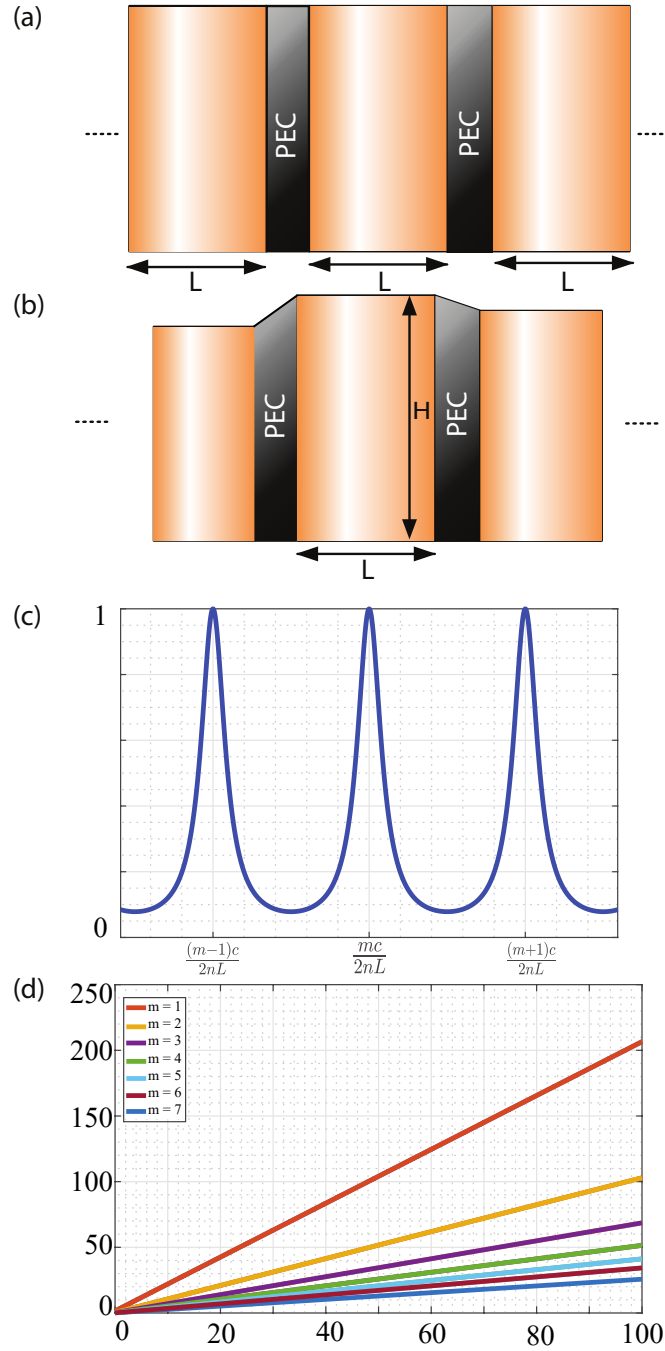


Figure 4.6: (a) Emitter model used to design the conventional superradiant thermal emitter in [34]. (b) Emitter model used to design the flexible emitter where ω_0 is controlled by tunable PEC dimensions as shown. (c) Periodic resonant modes observed in a 1-dimensional Fabry-Perot emitter cavity. (d) Resonant modes of a tuneable cavity. Note the existence of multiple modes for the same PEC thickness and the linear relationship between PEC thickness and ω_0 .

Control of resonant frequency in metallic slits has been an area of interest for several decades [84,85]. While the problem itself is highly complex and analytical, within the weak field regime, several simple relationships have been proposed to design cavities with arbitrary resonant frequencies. One such study [86] gives the following 2-dimensional control relationship for the resonant wavelength of a slit:

$$\lambda_m \approx \left(\frac{2.05}{m} H + \frac{a}{m} L \right), \quad (4.95)$$

where:

$$a \approx 1.8 + \pi \delta_{m,1}, \quad (4.96)$$

and L, H correspond to the cavity dimensions shown in Fig. 4.6(b). This relationship directly solves our design problem. The far-field coupling constant can now be tuned by changing the L parameter and the resonant frequency can be tuned by changing the H parameter as required. This clearly enables the retention of a unique resonant frequency by varying cavity dimensions. For a given thickness (H), several resonant modes will exist as shown in Fig. 4.6(d).

Another challenge is associated with designing emitter arrays in such a manner as to avoid radiating into various far-field non-superradiant modes. Such a model suppresses the effects of superradiance and aids various uncontrollable phenomena. This problem has been studied both analytically and numerically by Verslegers *et al.* [80] for the case of 2 emitters. However, through their findings, it can be assumed that in general terms, maintaining the inter-emitter distances of the assembly within subwavelength dimensions is the sufficient criteria to avoid radiating into sub-radiant modes [34].

Chapter 5

Characterisation of the Generated Thermal Superradiance Pulse

THis chapter presents the numerical simulation results for all emitter assembly configurations discussed in chapter 4. Insights into thermal superradiance are highlighted based on these results. Two superradiant emitter assemblies based on quantum dots are presented along with spectral and numerical simulation results.

5.1 Generating a Thermal Superradiant Pulse based on Nanophotonic PEC Systems

5.1.1 Simulation details

We performed a numerical simulation on the model to examine and validate the capabilities of the system. We solved eq. (4.21) by using the ECS derived in eq. (4.45) to obtain the emission variation of the assembly against angular frequency. We limited the simulation to an assembly with $N = 5$ emitters. The temperature of the assembly was maintained at 300K. We assumed that the velocity of light within each emitter is $0.013c, 0.017c, 0.01c, 0.017c$ and $0.013c$, where c is the velocity of light in free space. For the sake of simplicity, the widths of the emitters were predetermined to be 30, 50, 100, 50 and 30nm. We set an arbitrary resonant

wavelength of $8.3\text{ }\mu\text{m}$ for this simulation. By solving eq. (4.95) iteratively, we were able to obtain the depths of the emitters as 78.9, 162.6, 74.1, 162.6 and 78.9nm for the resonant modes $m = 2, 3, 4, 3, 2$ within each emitter, respectively. The calculated maximum wavelength of Planck's emission spectrum for this temperature is $8.28\text{ }\mu\text{m}$, which is clearly in the same order of magnitude as the resonant wavelength of the coupled emitter system. These simulation results are presented in detail within the following sections.

5.1.2 Semi generalized model

Fig. 5.1(a) shows the power spectra obtained for the sample emitter assembly designs shown to the right. The design corresponding to figure 5.1(b) is the non-resonant emitter assembly case where the emitters are placed at a greater than subwavelength distance apart. The design shown in Fig. 5.1(f) corresponds to the conventional emitter model proposed in [34]. By comparing the spectrum curves obtained for the two designs, it is clear that the non-resonant emitter assembly result is in agreement with the linear power scaling proven in (4.51a) and the conventional superradiant configuration is in agreement with the anomalous power scaling proven in (4.51b).

Figures 5.1(c, d, e) depict three possible novel emitter assembly configurations. The power spectral density curves based on (4.46) and (4.47) in this case show the expected enhancement by a factor better than $1/N$. The power spectral density curves based on (4.46) and (4.47) in this case show the expected enhancement by a factor greater than $1/N$. This is evident by the resonant peaks of these configurations being above the reference spectral value P . As described previously, because of the contribution of the temporal superradiant phenomena, the overall potential power gain at the resonant frequency is now apparent. Note that all emitter dimensions are selected according to (4.95), a crucial criterion required

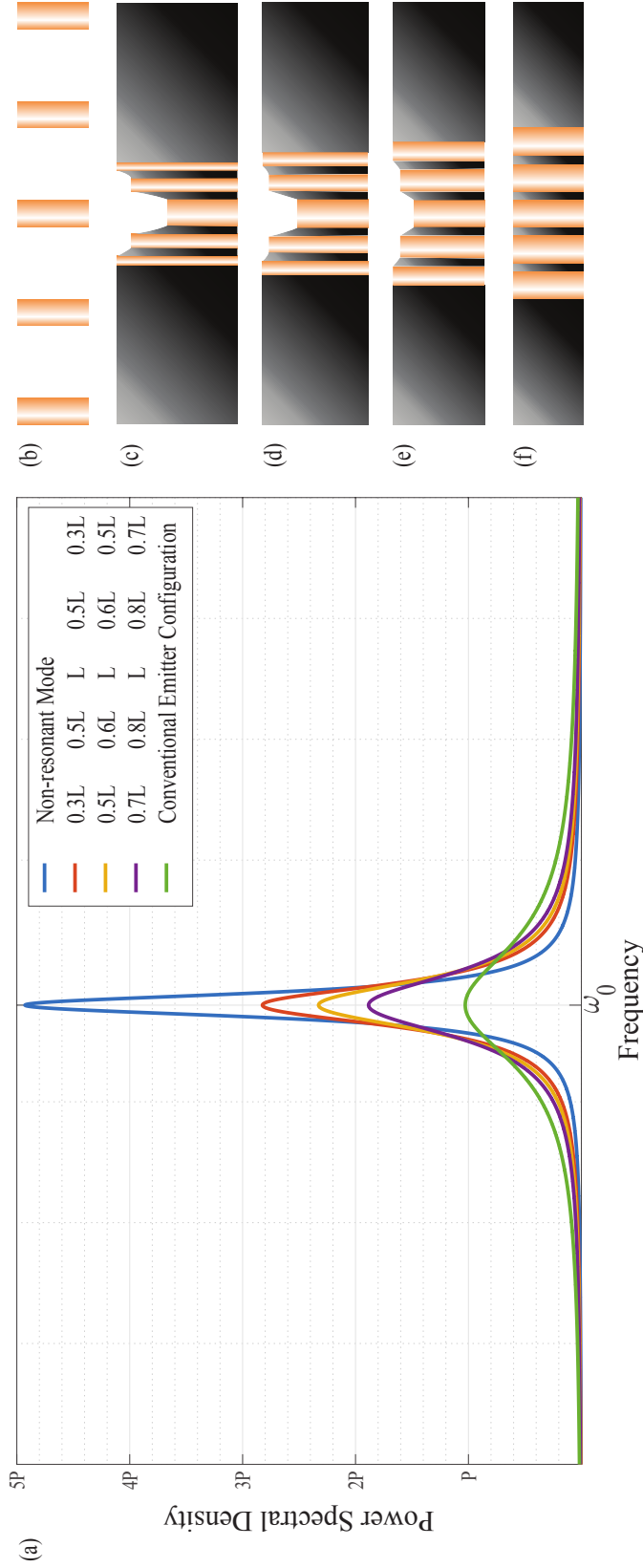


Figure 5.1: (a) Power spectral density as a function of frequency for different emitter designs utilizing 5 emitters. Note how the power at ω_0 varies for the different emitter configurations. The power scaling relationships for non-resonant and conventional superradiant modes agree with the direct results in (4.51a) and (4.51b). P is the power spectral density of a non-coupled emitter with a width of L and a thickness of H, corresponding to the dimensions of the middle emitter in each case. These symbolic values are chosen to represent typical values and exact, numerical values can be calculated using (4.47) for a given design. (b) Illustration of geometric distribution of non-resonant emitters. Note that the emitters are not in subwavelength proximity. (c,d,e) Configurations of tunable superradiant emitters where the emitters are positioned in subwavelength proximity. (f) Emitter configuration for the conventional superradiant emitter case where the emitters are positioned in subwavelength proximity. All emitter dimensions are shown to scale.

to maintain the same mode at the same resonant frequency.

Through these results, it is evident that by relaxing the common coupling constant assumption for the emitter ensemble, it is possible to considerably alleviate the spectral broadening issues highlighted in [34]. As expected, the characteristic Lorentzian curve for the non-resonant mode still shows significantly less spectral broadening than all superradiant modes. However, this issue is not a theoretical concern. The goal of the emitter is to deliver the most amount of power at resonance. Upon achieving steady-state operation, the effects of superradiance will quadratically improve the emission efficiency as discussed previously. While there are other losses that need to be considered when designing an operational emitter, any spectral suppression better than a factor of $1/N$ should theoretically enhance the overall emission at resonance.

We have shown that by choosing a set of tunable coupling constants, it is indeed possible to control the spectral suppression. In order to perform a fair comparison, we have scaled the coupling constants by tuning the dimensions with respect to the widest emitter in the array. In all of the emitter assembly designs illustrated in figures 5.1(b-f), we have chosen the central emitter to be at the same, largest chosen width. Relative width scales of the other emitters are shown in 5.1(a). For the case of figures 5.1(b) and (f), all of the emitters have the same height and width. In other words, we have maintained symmetry around the central emitter in order to facilitate direct comparisons.

Note that none of these are theoretical limitations and these emitters can be tuned according to the design requirements. All emitters are filled with an emissive material with the same permittivity and γ_{c_i} is taken as a quantity proportional to the emitter length as well as the permittivity of the emissive material [34].

5.1.3 Fully generalized model

Performance enhancement

The main advantage in being able to arbitrarily tune each emitter with complete flexibility is that it allows the assembly to be fine tuned to deliver optimal thermal power at an application specific frequency. While the number of emitter configuration possibilities are countless, a few are demonstrated here to illustrate how the flexibility can be used to enhance total power delivery. Note that all results are obtained through numerically solving eq. (4.21) for a five emitter assembly ($N = 5$) configuration. Four interesting power emission spectra are depicted in fig. 5.2. The γ_a and γ_c values of the centre emitter is maintained the same across each assembly for the ease of comparison. Each assembly is designed to be symmetric around the centre emitter for simplicity. It is worth noting that these are not actual design requirements and the versatility of the proposed emitter model enables us to adopt extremely liberal (application specific) design conditions.

The first configuration (see fig. 5.2(a)) represents the uniform emitter configuration. Note that the power emission at resonance is anomalously curtailed by a factor of $1/5$ as expected and this mode fails to emit beyond its non-resonant counterpart [34]. By extending the assembly to comprise three types of emitters, it is possible to circumvent this hurdle (see fig. 5.2(b)). The outermost emitters tend to absorb photons at a slightly higher rate than the inner ones and the observable power gain is apparent in the spectra. The third configuration (see fig. 5.2(c)) corresponds to a setup without material flexibility. Each emitter has the same photon absorption rate but the photon far-field coupling rates are varied such that the outermost emitters couple at a lower rate than the inner emitters. The obtained gain in this case is approximately similar to the previous assembly, but involves a more complex design due to high shape perturbation dependencies [82]. The final configuration (see fig. 5.2(d)) combines both effects. The ma-

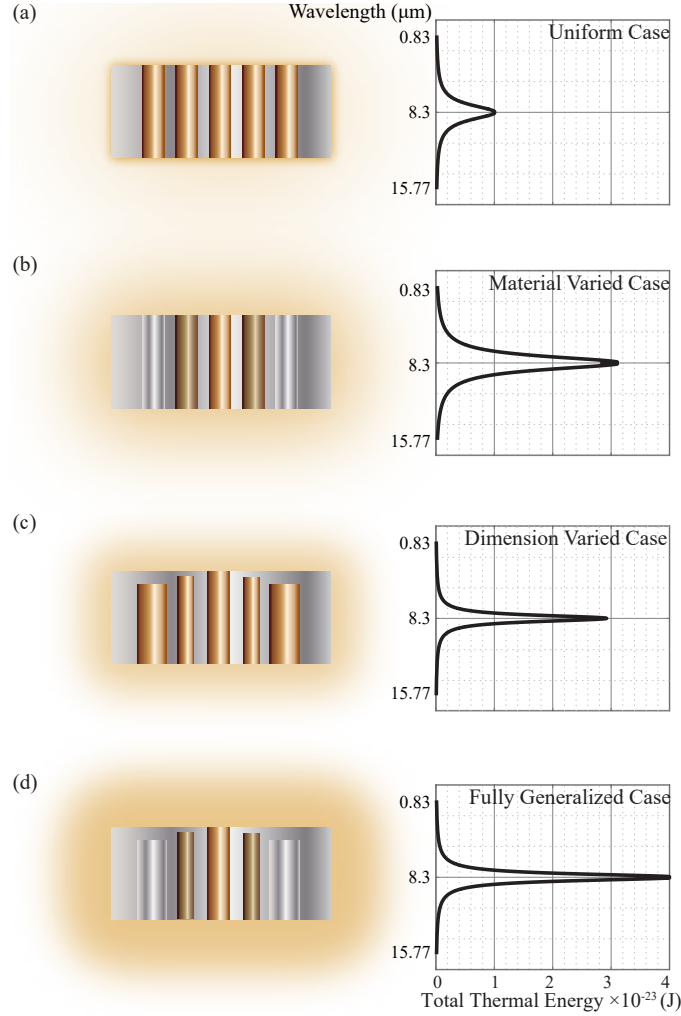


Figure 5.2: Numerically obtained emission spectra by solving eq. (4.21), together with eq. (4.45) for different assemblies comprising 5 emitters in each. Primary emitter parameters used for the simulation are $\gamma_a = 10^{12}\text{s}^{-1}$ and $\gamma_c = 10^{13}\text{s}^{-1}$. **(a)** The uniform emitter assembly case depicts the expected anomalous power scaling discussed elsewhere [34]. Each emitter within this assembly uses the parameters γ_a and $2\gamma_c$. **(b)** The material varied emitter configuration shows a higher power emission at resonance. All emitters have the same γ_c parameter and the photon absorption rate parameters are $[1.7\gamma_a, 1.6\gamma_a, \gamma_a, 1.6\gamma_a, 1.7\gamma_a]$. **(c)** The dimensionally varied emitter configuration achieves a similar power emission at resonance. All emitters have the same γ_a value and the photon far-field coupling rate parameters are $[0.5\gamma_c, 0.7\gamma_c, \gamma_c, 0.7\gamma_c, 0.5\gamma_c]$. **(d)** The final assembly is completely generalized using emitter parameters for photon absorption rates from **(b)** and photon far-field coupling rates from **(c)**. The energy values computed at the point of resonance are $[1.051, 3.043, 2.975 \text{ and } 4.155] \times 10^{-23}\text{J}$, respectively.

terials are used in the same order as configuration (b) and the far-field coupling constants are varied in the same order as configuration (c). It is noted that the gain obtained for this configuration outperforms all other configurations shown.

Startling phenomena

Depending on the configurations of the individual emitters, power spectra of various emitter assemblies tend to depict startling emission spectra similar to those observed in the transmission spectra of EIT and scattering spectra of super-scattering systems respectively. To elucidate this observation: Traditionally, EIT occurs in atomic systems when two states are coupled through possible alternative processes where quantum constructive or destructive interference can modify the total transition probability between energy levels [87,88]. By artificially inducing such interferences, it has been possible to practically demonstrate the effects of EIT [89]. Theoretically in a three level system, EIT is modelled through electromagnetically coupling the two upper energy levels at a Rabi frequency higher than the inhomogeneous width of the two lower levels, thus rendering the generally opaque transition transparent. This interesting phenomenon has led to many attractive discoveries such as, slow light [90] and stopped light [91] and has given rise to many applications in fields such as quantum computing [92] and optics [93]. Superscattering is a relatively new construct proposed by Fan's group that explains the significant increase of the scattering cross-section of sub-wavelength nano structures [94]. Several efforts have focussed on obtaining the effects of superscattering since, ranging from the use of nano-spheres [95] to single metallic nano-disks [96]. Both these phenomena result due to the presence of quantum interference.

However, the analysis of our model (ECS) is completely different to the models on which these phenomena are defined and therefore, we refrain from labelling these observations as EIT and superscattering. Instead, due to the similar

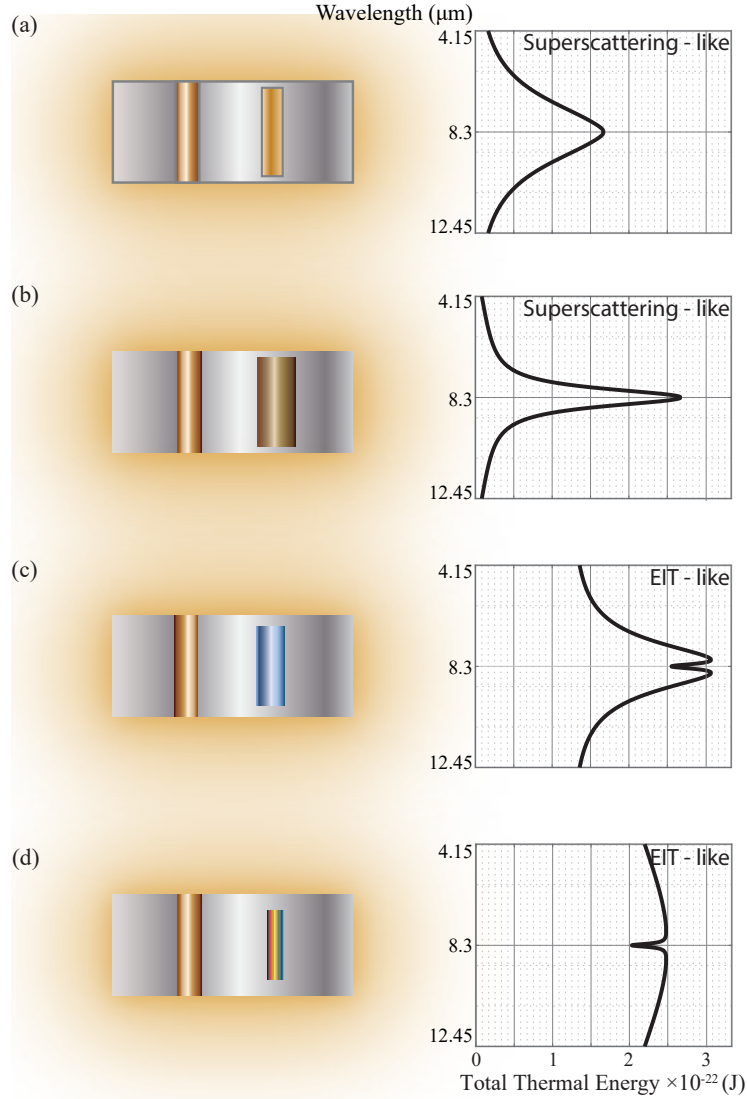


Figure 5.3: Analytically obtained emission spectra by solving eq. (4.21), together with eq. (4.70) for different assemblies comprising 2 emitters in each. Primary emitter parameters used for the computation are $\gamma_a = 10^{12}\text{s}^{-1}$, $\gamma_c = 10^{13}\text{s}^{-1}$. The left emitter in each assembly has the parameters γ_a and $0.1\gamma_c$. **(a)** Superscattering-like emission phenomenon observed when the right emitter has parameters $[1.5\gamma_a, 0.5\gamma_c]$. **(b)** Superscattering-like emission phenomenon observed with a higher disparity when the right emitter has parameters $[15\gamma_a, 0.15\gamma_c]$. **(c)** EIT-like emission phenomenon observed when the right emitter has parameters $[30\gamma_a, 4\gamma_c]$. **(d)** EIT-like emission phenomenon observed with a higher disparity when the right emitter has parameters $[250\gamma_a, 70\gamma_c]$. The energy values computed at the point of resonance are $[1.633 \ 2.673 \ 2.545 \ 2.063] \times 10^{-22}$ J, respectively.

characteristics observed, we refer to these observations as EIT-like and super-scattering-like respectively.

In order to better investigate the emitter assembly and observe the presence of interference, (4.45) can be analytically solved for the case of two emitters ($N = 2$, as shown in Fig. 4.3) and simplified as follows:

$$^2\sigma(\omega) \approx \frac{4c}{\omega_0} \frac{\gamma_{a_1}\gamma_{c_1}(4\Omega^2 + \gamma_{a_2}^2) + \gamma_{a_2}\gamma_{c_2}(4\Omega^2 + \gamma_{a_1}^2)}{16\Omega^4 + 4\Omega^2\mathcal{F}_1 + \mathcal{F}_2}, \quad (5.1)$$

where $\Omega = \omega - \omega_0$ is the detuning parameter and the factors:

$$\mathcal{F}_1 = [(\gamma_{a_1} + \gamma_{c_1})^2 + (\gamma_{a_2} + \gamma_{c_2})^2 + 2\gamma_{c_1}\gamma_{c_2}] \quad (5.2)$$

$$\mathcal{F}_2 = (\gamma_{a_1}\gamma_{a_2} + \gamma_{c_1}\gamma_{a_2} + \gamma_{a_1}\gamma_{c_2})^2 \quad (5.3)$$

In order to better investigate the emitter assembly and observe the presence of interference, we need to carefully analyse the mathematical behavior of the equations. It is clearly noted that both eq. (4.70) and eq. (4.75) have resolved to the form of a division between two polynomials of the detuning parameter. The majority of the coefficients of both the numerator and denominator polynomials are self terms where components of the same emitter are associated together. However, the presence of cross terms is evident in the constant-part of the denominator polynomial, which is a clear predictor of the presence of interference effects in the system. When the two emitters are dissimilar in character, these interferences will intensify, giving rise to EIT-like and superscattering-like observations.

These vital phenomena are unobserved in previously proposed thermal superradiant systems [34, 82] due to their aforementioned inadequacies.

Figure 5.3 depicts four possible emitter configurations for the $N = 2$ case where startling emission phenomena are predicted by our model (note that these observations are not limited to the $N = 2$ case). Furthermore, the left emitter characteristics are unchanged on all designs for the ease of comparison. The right

emitter is changed in each case to observe the superscattering-like and EIT-like phenomena. The right emitter in the first assembly (see fig. 5.3 (a)) is only slightly different in character to the left emitter. It has a higher photon absorption rate and an identical photon far-field coupling rate. The curve shows a characteristic superscattering-like behaviour but due to the approximate similarity of the emitters, it almost maintains the shape of a perfect Lorentzian. The right emitter of the second assembly (see fig. 5.3 (b)) has a photon absorption rate and a photon far-field coupling rate that is fifteen times larger than its corresponding left emitter. Due to the prominent dissimilarity in this case, a much sharper superscattering-like behaviour is observed in this scenario. These superscattering-like predictions are completely novel and unobserved in practical or analytical studies of thermal superradiant emitter systems.

The third assembly (see fig. 5.3 (c)) has a right emitter that has a much higher photon absorption rate and a much lower photon far-field coupling rate when compared with its left emitter counterpart. Due to the higher absorption intensity, a marked destructive interference is observed in this case, which gives rise to an EIT-like characteristic to the corresponding power spectrum. The final assembly (see fig. 5.3 (d)) is an extension of the previous case. The photon absorption rate of the right assembly is much larger than the left assembly and the photon far-field coupling rate is unchanged from the previous right emitter counterpart. The EIT-like characteristic dip is highly prominent in this setup and the power emission at resonance is at the lowest when compared with the other demonstrated assemblies. These predicted observations are in agreement with the recent practical observation involving the superradiant decay of qubits [97].

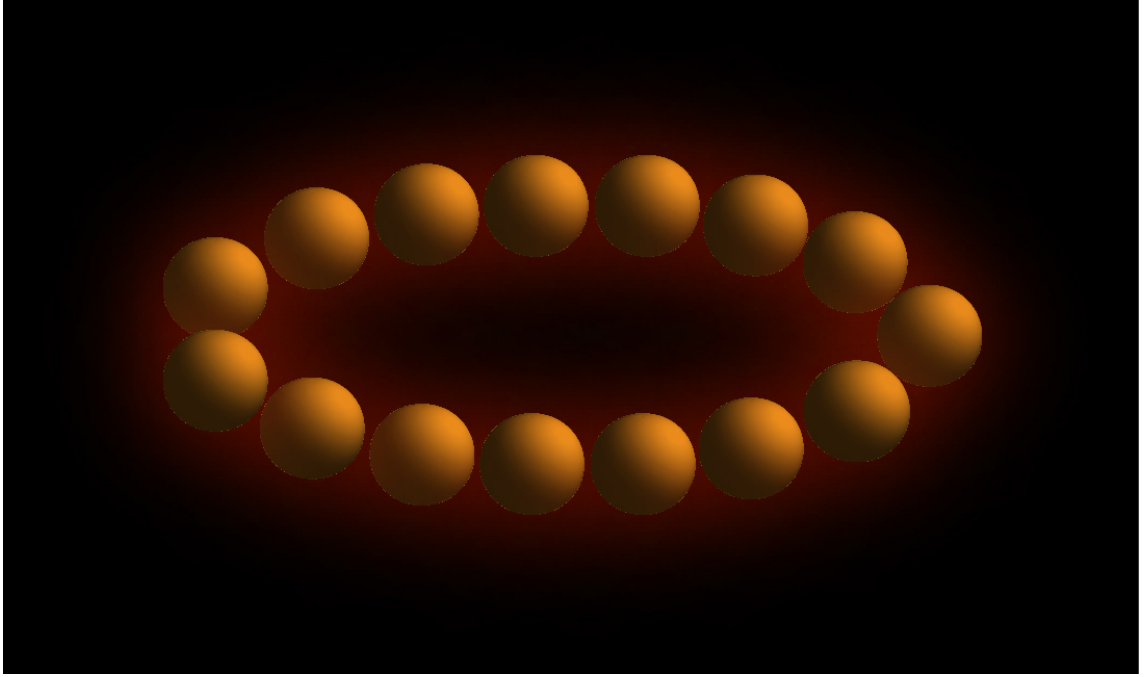


Figure 5.4: Proposed emitter assembly has 15 quantum dots (QDs) in the shape of a ring. Each QD operates in the thermal regime at a resonance frequency of 2.27×10^{14} Hz. The QDs are modeled after measured dimension and dipole moment parameters for CdSe QDs.

5.2 Ring Arrangement of Quantum Dots

Superradiance is observed in a system of N interacting, identical sources when confined within dimensions smaller than the resonant wavelength of the emitter assembly. It is often an effect ignored in many recent studies involving nanoparticle systems [98–102]. Furthermore, it is essential that all the emitters in the assembly maintain permutation symmetry. This means that, in a purely mathematical sense, while the emitters should be geometrically distributed in an entirely symmetrical shape, any two emitters of the assembly should be interchangeable without disrupting the integrity of the assembly. Although this requirement appears simplistic, due to the vectorized nature of the intrinsic dipole moments of the emitters, very few geometries can actually fulfil the permutation symmetry criteria. As shown in Fig 5.4, a ring structure of quantum dots (QDs) can be de-

signed to adhere to these criteria and emit superradiant pulses [29]. Given the recent advances in QD design and experimental realization, such a ring structure is certainly within the scope of experimentation [103, 104].

5.2.1 Simulation parameters

Each QD has a radius of 5.5nm and an inter-source separation of 4nm. A conventional system is realized when the QDs are placed far apart and the system dimensions are larger than the interacting wavelength.

5.2.2 Emission characteristics

Superradiant and conventional emission characteristics are shown in Figs. 5.5(a) and (b), respectively. These results are obtained using coupled mode analysis and quantum electrodynamics [82, 105]. The log of emitted thermal energy (J) by the system in Fig. 5.4 is shown. The parameter $\tau = 1/\Gamma_a$, where Γ_a is the spontaneous emission rate of a QD [29]. The superradiant pulse is initially $\approx 10^3$ more intense than the conventional.

The energy emission spectra are shown in Fig. 5.6 (a) and (b) for a superradiant and conventional system, respectively. Throughout the pulse duration ($t \gg \tau$), the conventional system delivers $\approx 3 \times$ the power of the superradiant counterpart at a much slower rate.

5.3 Buckyball Arrangement of Quantum Dots

The emitter assembly proposed in Fig. 5.7 comprises 60 QDs that are placed at the vertices of a truncated icosahedron. The QDs are positioned according to Cartesian coordinates that are computed using the well-known even permutations method. Furthermore, the QDs are labeled using the numbering scheme

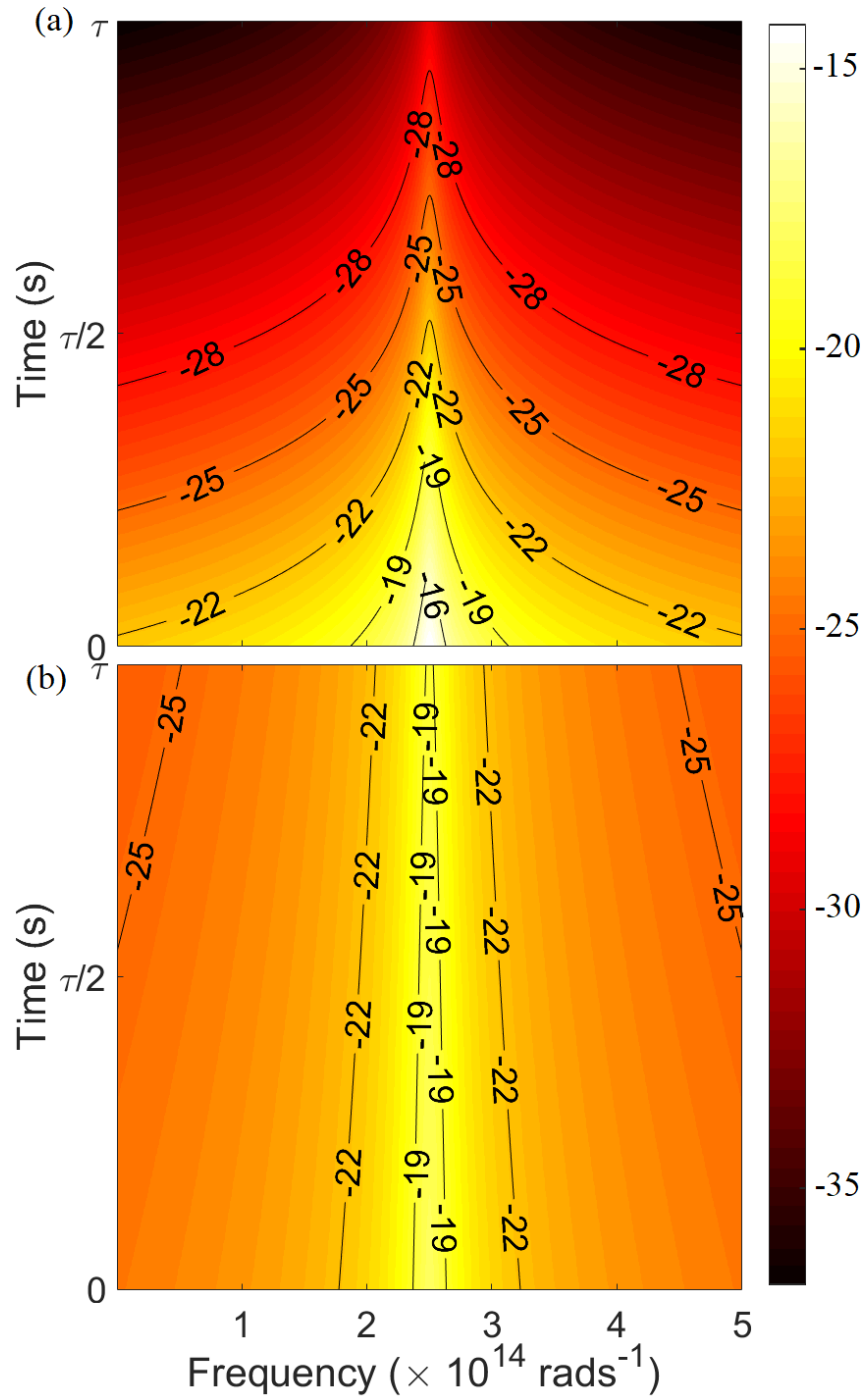


Figure 5.5: The emission dynamics of the (a) superradiant and (b) conventional system are shown. The photon generation rate is $1.0278 \times 10^{14} \text{ s}^{-1}$. The numbers show the log of emitted thermal energy. The superradiant pulse is clearly a high intensity and an ephemeral pulse compared to the conventional counterpart.

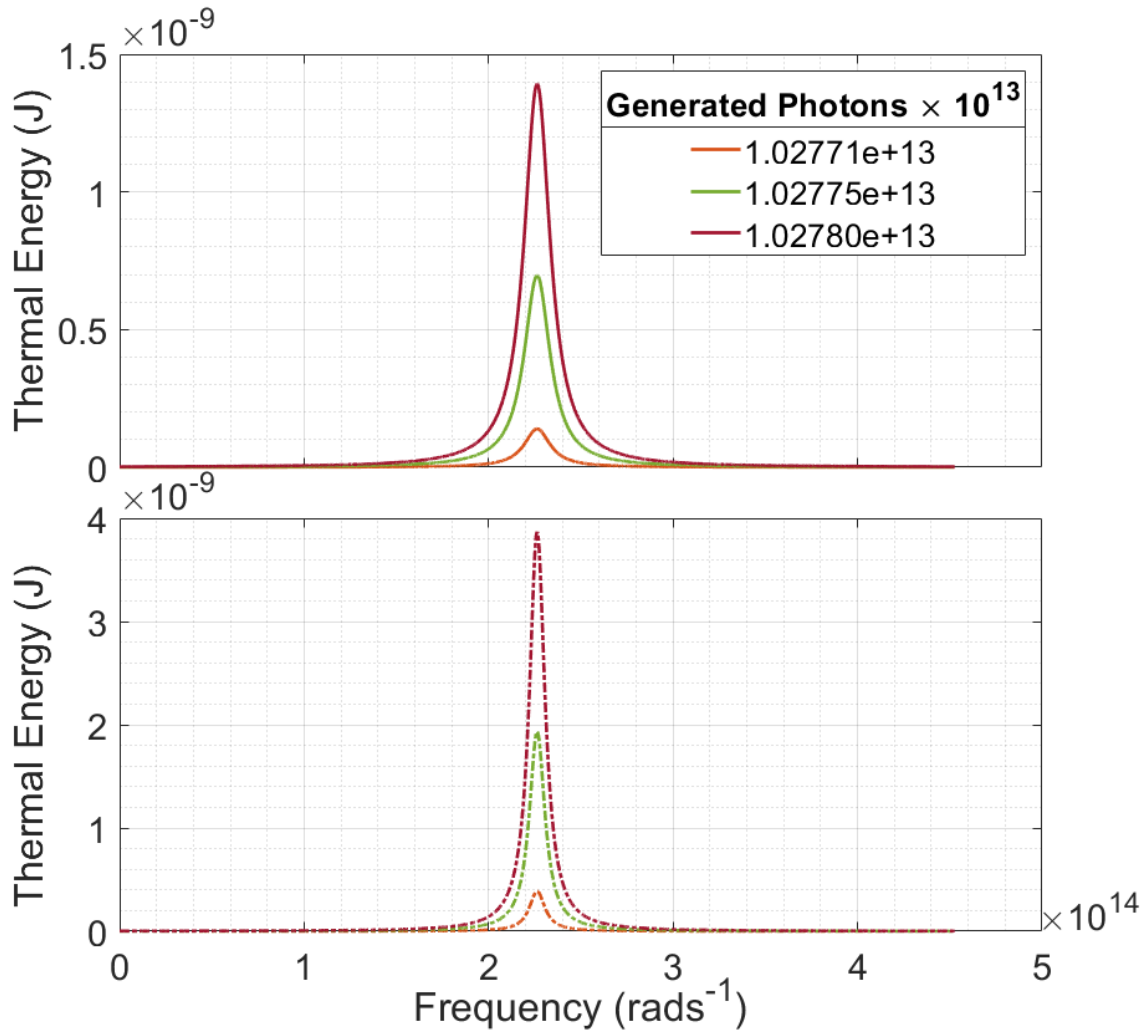


Figure 5.6: The emission spectra of the (a) superradiant and (b) conventional system are shown for a single pulse. The conventional system emits $\approx 3 \times$ intractable energy compared to the superradiant system.

based on permutations in the alternating group A_5 [106].

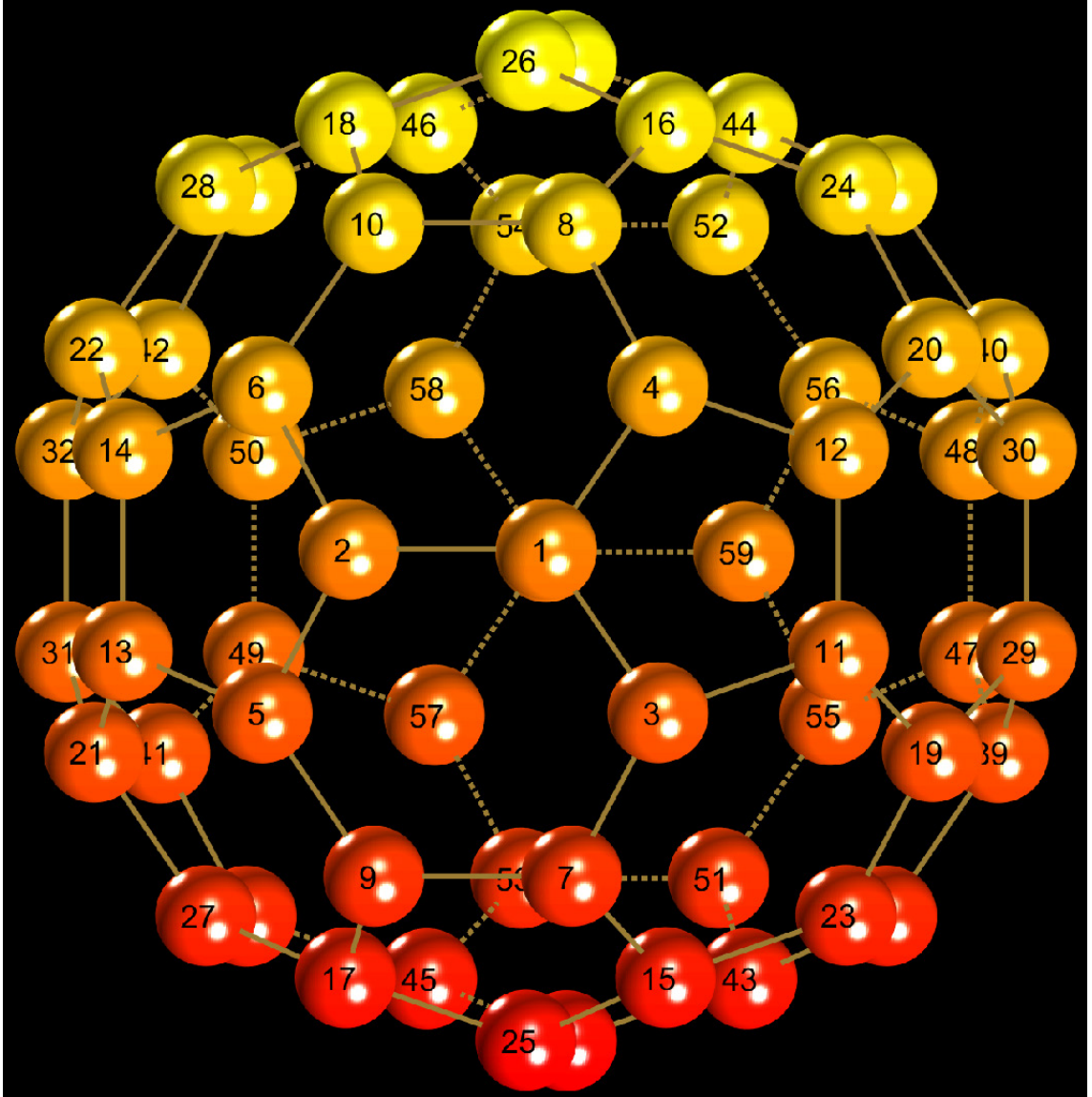


Figure 5.7: Proposed superradiant emitter assembly configuration where 60 QDs are placed at the vertices of a truncated icosahedron. The assembly is designed nanoscopically to fulfil the size SRC, where the emitter dimensions (r) are much smaller than the wavelength with which it interacts (λ). Furthermore, each QD is assumed to comprise a permanent dipole and an external control field is used to manipulate the behaviour of the resultant dipole of each QD. Under the correct conditions, this assembly fulfils the symmetry SRC as well.

Maintaining size SRC

When constructed using a scaling dimension (say r), all vertices of the truncated icosahedron will lie on a sphere with radius:

$$\mathcal{R} = r\sqrt{9\psi + 10}, \quad (5.4)$$

where ψ is the golden mean and the edge length will be $2r$ [106]. This implies that $r_{pb} = 2r$, where QD b is a nearest neighbour of p . As shown in Fig. 2.3a, we assume that each QD will have exactly 3 nearest neighbours, each at a distance of $2r$. This is a fair assumption, given that the hopping-interaction strength is proportional to $1/r_{pq}^3$ and within nanoscopic dimensions, interactions beyond the nearest neighbour limit are usually safely ignored [2, 29, 32].

It is noted that by choosing r appropriately, our model can be designed to fulfil the size SRC. Furthermore, by varying the QD dimensions while maintaining the size SRC, it will be possible to tune the emission characteristics due to the resulting changes to QD parameters such as absorption and emission.

Maintaining symmetry SRC

Assuming that it is possible to obtain a geometrical QD assembly where the resultant polarization vector of each QD intersects with the origin of the truncated icosahedron and the centre of the respective QD, it is possible to maintain the SRC. In this configuration, each QD will have a unique unit polarization vector. As shown in Fig. 2.3b, under this assumption, the angle β then determines the value of the hopping-interaction strength in equation (2.17). Simple trigonometry shows that:

$$\Lambda_{int}(p, b) = 0.8778616 \frac{|\mathbf{q}_p|^2}{4\pi\epsilon_b\hbar r^3}, \quad (5.5)$$

is a constant where b is a nearest neighbour of p . Maintaining a constant value for $\Lambda_{int}(p, b)$, $\forall(p, b)$, where b is a nearest neighbour of p , is clearly in agreement with the symmetry SRC. It is noted that since only the nearest neighbour interactions are used in our model, Λ_{int} is a sparse matrix as shown in Fig. 5.8.

Therefore, our proposed emitter assembly should be superradiant under these conditions. However, it is noted that by using non-spherical QDs, the symmetry SRC is potentially challenged due to anisotropy and the existence of multiple

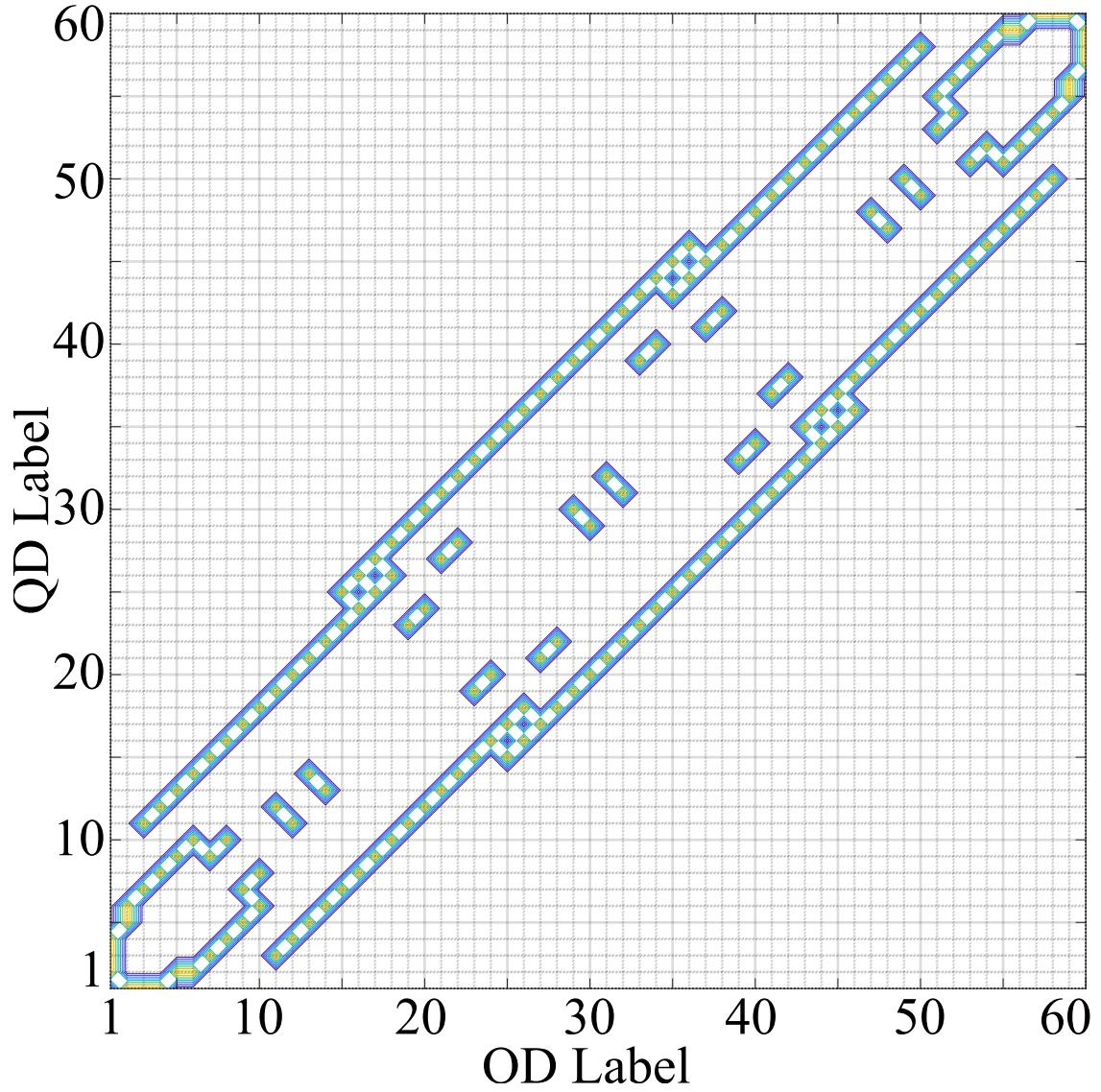


Figure 5.8: In this grid arrangement, an interacting pair of QDs is highlighted using a coloured box. The presence of these interactions is critical in achieving superradiant emission. The assembly QDs are labelled using the same technique used in Fig 5.7. Only the neighbouring QDs that are highlighted in coloured boxes are considered to interact with each other. This implies that the interaction rate matrix Λ_{int} is sparse and occupies the same rate value at each of the highlighted places. Incorporating interactions beyond this limit will break the symmetry SRC, thus rendering the system sub-radiant.

resonant modes.

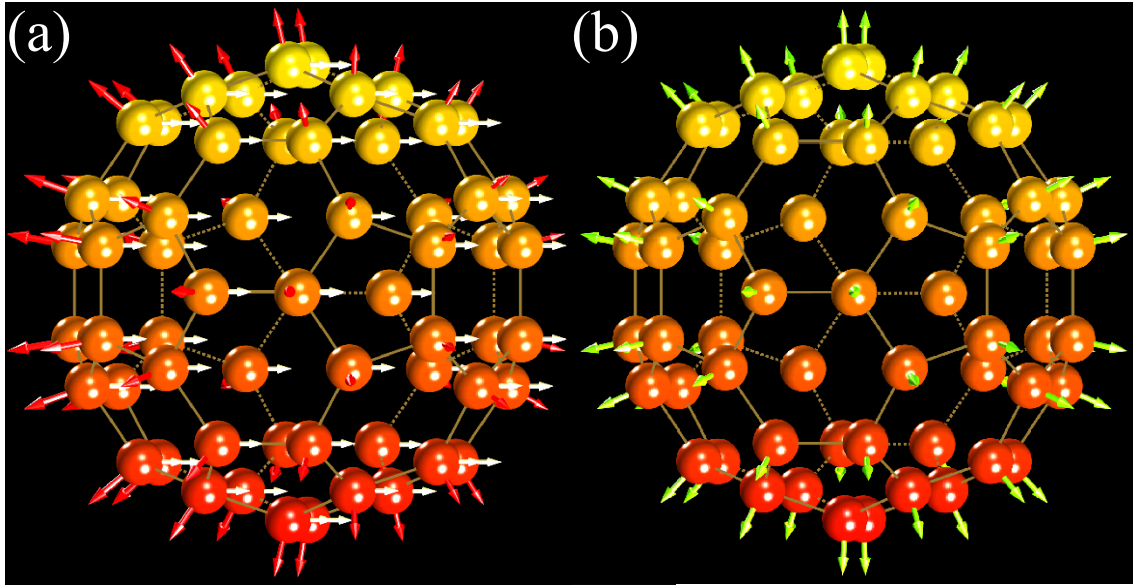


Figure 5.9: (a) An assembly of QDs with large, intrinsic, permanent dipoles (white arrows) placed at the vertices of a truncated icosahedron. When the assembly is exposed to an electric field, a temporary dipole is induced (red arrows) within the QDs. Since the assembly is minuscule, it is assumed that each QD interacts with the incident field in the same manner. Therefore, the induced dipoles have the same magnitude and direction. (b) The resultant dipole moments for the two dipoles are shown by arrows. When the permanent and induced dipoles are perfectly matched, it is possible to have all resultant dipoles originating from the center of the buckyball and traverse through the center of the corresponding QD.

5.3.1 Superradiant controllability

Now we propose a method to realistically achieve the resultant dipole arrangement to maintain the symmetry SRC. It is known that QDs built using specific material and size criteria possess intrinsic, permanent dipoles of high potentials [107–109]. Furthermore, it is well-known that the application of an external electric field will induce a dipole moment due to the polarizability of the exciton. Fig. 5.9a depicts this scenario. Here we have shown the field-induced dipoles using red arrows. Given the nanoscopic dimensions of the emitter assembly, we have assumed that all QDs uniformly interact with the incident electric field. The white arrows denote the direction of the permanent dipoles of each QD. When

two dipole moments are present within the same QD, the resultant dipole moment is the pivotal parameter controlling inter-emitter couplings. The resultant dipole moment for each QD is shown by the green arrows in Fig. 5.9b.

Under this configuration, superradiant emission of the proposed assembly can clearly be controlled by alternating the control field. When the control field is OFF, the resultant dipole will be the permanent dipole for each QD and the symmetry SRC is clearly violated. Emission will still exist but without superradiant enhancement. When the control field is ON, the induced dipoles will change the resultant dipole to the configuration in Fig. 5.9b. This will reinstate the superradiant behaviour to the assembly. When superradiant, the system will emit a high burst of energy within a very short time. Alternating the control field will result in pulsated emission of high energy, ephemeral pulses. It is noted that this controllable pulsating behaviour is not present in conventional, non-interacting emitter assemblies.

However, in a purely practical perspective, arranging a QD assembly to perfectly meet the presented mathematical requirements will be challenging. However, recent advancements in QD technology strongly suggests the possibility of realizing the structure [103, 104]. Furthermore, it is noted that the value of $\Lambda_{int}(p, b)$ beyond the nearest neighbour condition is not a constant under our configuration. However, in a practical setting, the presence of such trivial interactions will not hinder the effects of superradiance [110].

Assuming all these criteria are maintained, the next step is to study the spectral characteristics of the QD emitter assembly. Once the spectra are known, it is possible to use equations (2.19) and (2.20) to conduct simulations and study the dynamics of the proposed assembly.

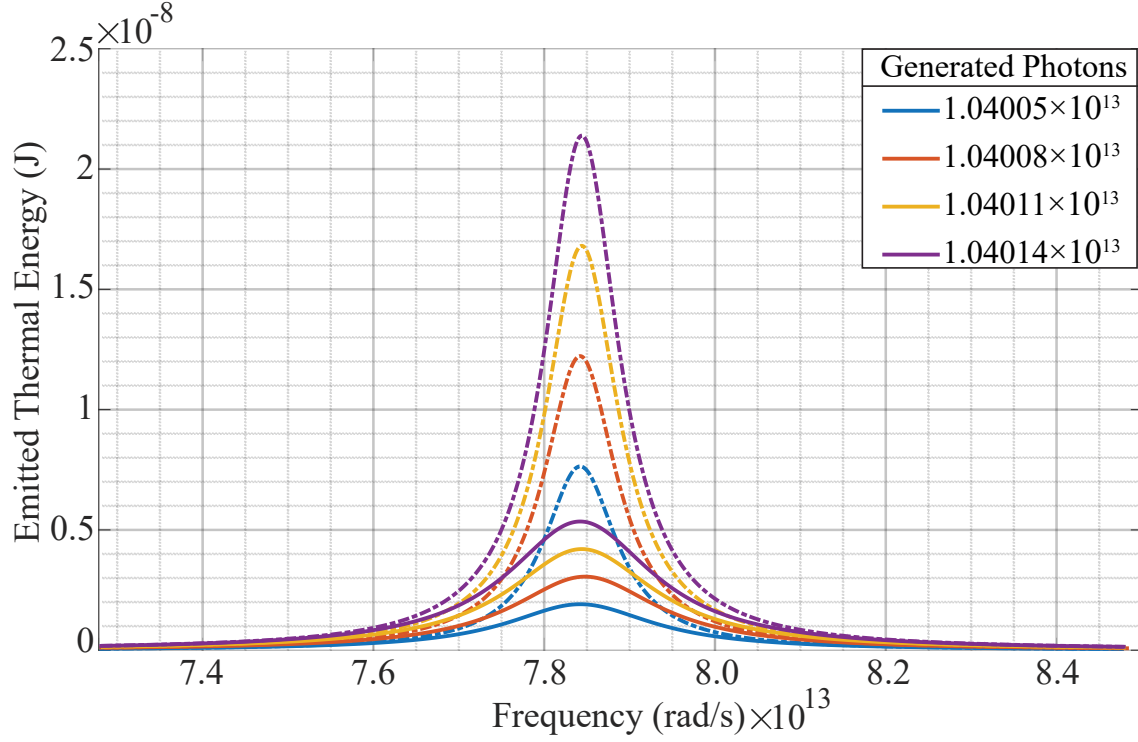


Figure 5.10: The emission spectra are plotted using the derived analytical spectrum. Continuous and dashed lines correspond to the superradiant and non-interacting assemblies, respectively. The curves correspond to different QD configurations with varying photon generation rates. The non-interacting assembly always outperforms the superradiant counterpart, which is the expected behaviour.

5.3.2 Simulation parameters

First we analyzed the non-interacting emitter assembly, the dynamics of which are described by equation (2.19). Here, we assumed that $\Lambda_{int}(p, b) = 0, \forall p, b$ and solved. A similar approach was used to solve equation (2.20), where we assumed that $\Lambda_{int}(p, b)$ takes the form of the sparse matrix shown in Fig. 5.8.

We assumed that the initial temperature value of both emitter assemblies is at 323.15K, which is well suited for cancer hyperthermia. This corresponds to a resonance frequency of $7.84 \times 10^{13} \text{ rads}^{-1}$ for thermal photons. In the simulation, we assume that all emitters are engulfed within cancerous tissue. Therefore, we assumed the bath permittivity: $\epsilon_b = 7.17$, which is in agreement with pub-

lished results [111]. For simplicity, we ignored the temperature and frequency dependencies of ϵ_b for this simulation. In order to perform cancer hyperthermia effectively, we assumed the resultant dipole matrix element of each QD as 3.3336×10^{-28} Cm. This value is in agreement with measured values for CdSe QDs, each with a diameter of 5.6nm [107]. The separation between two adjacent QDs was maintained at 4.4nm, which corresponds to $r = 2.2$ nm.

5.3.3 Emission characteristics

For the superradiant assembly, these parameters correspond to a total inter emitter coupling rate of $1.309 \times 10^{13} \text{ s}^{-1}$ for all interacting QDs. Since we are assuming a 3-nearest neighbour scenario, this value has to be scaled appropriately to find the effective coupling rate Λ_{int} .

This results in a far-field coupling rate of:

$$\Lambda_E = 2.618 \times 10^{13} \text{ Hz}, \quad (5.6)$$

which can be directly calculated. Due to the lack of experimental data for photon emissivity and absorptivity, we picked a variety of photon generation rates for our simulations. Finally, the value for Λ_A is found using the principle of energy conservation.

The heat transfer simulation for the tissue model is solved for both transient and steady-states. At $t = 0$, the emitters, tumour and healthy tissue regions are assumed to be at 323.15K, 311.15K and 310.5K, respectively. For both tissue types, thermal conductivity and mass density were maintained at $0.48 \text{ Wm}^{-1}\text{K}^{-1}$ and 1080 kgm^{-3} , respectively. Furthermore, specific heat capacities of 3000 and 3500 $\text{Jkg}^{-1}\text{K}^{-1}$ were maintained for healthy and tumour tissue types, respectively. For the simulation, we placed 8 emitter bundles close to the edge of the tissue region. Each emitter bundle was assumed to comprise 1000 buckyball assemblies.

The size of a single emitter assembly was assumed to be negligibly small compared to the size of the tumour and the specific heat capacity of each assembly was assumed to be negligibly small. All emitter parameters were obtained by solving equations (2.19) and (2.20) for the non-resonant and superradiant emitter assembly simulations, respectively [112].

However, it is noted that different QD parameters are required for a realistic simulation because the biocompatibility of TOPO-coated CdSe QDs is unestablished. Furthermore, it is necessary to functionalize the surface of each QD prior to in-vivo applications, which will result in further modified QD parameters. Furthermore, we have ignored the temperature dependent variations to these parameters. When simulating the tissue model, we have assumed that the effects of blood perfusion and metabolic heat generation are negligible. We have also assumed that the simulated region is free from blood vessels. Moreover, it is known that the application of hyperthermia will alter the thermal tissue characteristics. We have ignored such changes as well. In fact, the simulated tissue model is a crude approximation, that is presented to juxtapose and validate the superior cancer hyperthermia characteristics of the superradiant assembly in comparison with its conventional counterpart.

As shown in Fig. 5.10, for the time independent emission, a Lorentzian pattern is observed for both non-resonant and superradiant assemblies. This is the expected result for emissions of this nature [34]. These results are obtained by solving the set of equations for four photon generation rates. For each instance, it is evident that the non-resonant assembly emits a larger number of photons, compared to its superradiant counterpart. This is expected, given that the resonant emitter assembly suffers losses due to inter-emitter interactions.

Fig. 5.11 shows the complete dynamics for the two assemblies. Colour values depict the log of generated energy, at each frequency and time point. These results are for the highest photon generation rate shown in Fig. 5.10. It is noted

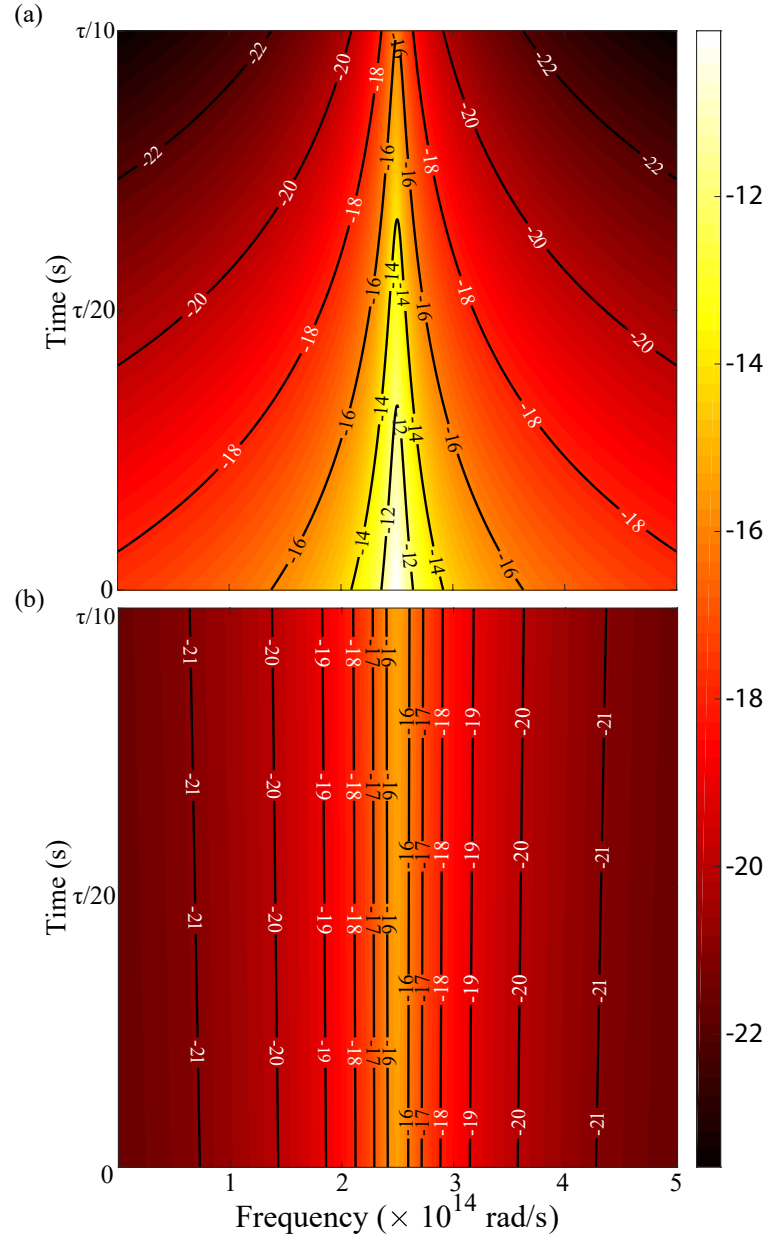


Figure 5.11: These plots depict the emission dynamics for two analogous (a) superradiant, (b) non-interacting, emitter assemblies. The photon generation rate is assumed to be $1.04014 \times 10^{13} \text{s}^{-1}$. These results are obtained by solving (a) equation (2.20) and (b) equation (2.19), respectively. Colour values correspond to the $\log$ of generated energy at each time and frequency point. The negative values suggest that a single assembly only generates a minute amount of heat. The black contour lines highlight the contrasting emission dynamic patterns of the two emitter assemblies.

that, although the superradiant assembly generates a lower number of photons for a given frequency as shown in Fig. 5.10, the emission power at resonance at the beginning of the superradiant pulse is much higher ($\sim \times 10^4$) than the power generated by the non-resonant counterpart at any frequency and time point.

Chapter 6

Superradiance as a Viable Method for Biomedical Applications

This chapter briefly discusses the literature pertaining to the field of nanomedicine and provides insights on novel cancer treatment modalities that stem from it. Simulation details and results from tissue model simulations for two tumour types are presented as a case study.

6.1 Overview of Nanomedicine

Nanomedicine is best described as the use of nanotechnology for the enhancement of medicine and has been around for nearly 3 decades. It is a relatively new field in medicine and holds a profound amount of potential towards improving various techniques and eradicating diseases. Currently, nanomedicine has clinical applications in various fields such as drug delivery and active implants.

As with any new invention in medicine, it takes considerable effort to commercialize a nanomedicine product, regardless of its application or potential benefits. A review published in 2006 reported more than 150 start-ups with a predicted sales value of nearly US\$7 billion [113].

6.1.1 Enhanced permeability and retention effect

The enhanced permeability and retention effect (EPR) is one of the major drivers of nanomedicine [114]. However, in recent years, the concept has undergone serious scrutiny over possible ineffectiveness for human subjects [115]. The idea is simple; utilizing the weaknesses of tumours to improve upon treatment techniques. Tumour vasculature, on the gross level, comprises poorly aligned cells and lacks innervations. This leaves openings in the vasculature, this leads to a scenario referred to as a 'leaky vasculature', which allows nanoparticles to easily reach for tumour cells.

In fact, in early EPR studies in animal subjects, it was confirmed that due to EPR, almost 100% of the nanoparticles gather and retain within the tumour after a certain amount of time. Early drug delivery trials on animal subjects showed significant promise due to this fact. However, clinical replication of the same level of success was not possible. The failure to replicate the same success levels in human subjects is potentially due to the differences in tumours between humans and smaller animals (such as mice), specifically the tumour to body weight ratio. It is hypothesized that due to the high tumour to body weight ratio in mice, their tumours tend to depict much higher leaky vasculature characteristics than the tumours in human counterparts.

Improving the EPR effect

Currently, researchers are extensively studying the microenvironment of human tumours in order to propose a method to enhance the EPR effect and to mitigate anti-EPR properties [116]. Certain schemes have in fact, proven be successful in achieving this requirement. Biochemical components, known as vascular mediators, involved in producing the EPR effect in animal tumours have been identified and are currently been investigates as EPR enhancing agents [115].

6.1.2 Drug delivery

Nanotechnology enhanced drug delivery is one of the earliest aspects of nanomedicine that dates back to the 1960's [115]. Through its utilisation, studies have shown that it's possible to achieve higher localization of drugs within tumours. Furthermore, the coupling of drugs and nanoparticles has shown to reduce the toxicity of the drugs themselves. This is a significant achievement, given the fact that most anti-cancer drugs are highly cytotoxic. Through the use of nanoparticles, it has also been possible to design drugs that release slowly (retarded-release). One major issue with conventional drugs is their inability to penetrate the blood-brain barrier. Through the utilization of nanomedicine, this too has become a possibility. Furthermore, nanomedicine holds the key to designing personalised medications that can be highly effective compared to the generic counterparts [117].

6.1.3 Imaging of tumours

Imaging tumours and their environment is a key component in battling cancer. Not only does it allow experts to understand the state of the disease, it also plays a major role in determining the proper treatment approach. Even though there is doubt whether the EPR technique is as applicable to humans as once expected, there have been clinically proven instances of nanotechnology based biomarkers that have shown higher concentrations in human tumours, compared to healthy tissues. Once the nanoparticles gather within tumours, it is possible to use various optical techniques to exactly recreate their structure within the tumour, which eventually allows the recreation of the tumour structure itself. However, the newest trend, known as *nanotheranostics* is the combination of therapeutic and contrast agents together to form a single drug. When used, this fused drug not only facilitates tumour imaging, but also provides means to predict the drug de-

livering efficiency [117].

6.1.4 Nanomedicine and cancer therapy

Cancer ablation

Ablation of tumours is a comparatively more invasive technique than hyperthermia. It is possible to either increase (more than 60°C) or decrease (less than -40°C) the tumour temperature to successfully achieve ablation [118]. As expected, depending on the type of tumour treated, the temperature requirement is varied. The main advantage of cancer ablation is the ability to treat the tumour at a much shorter time period with a higher efficiency. In fact, if the temperature of 60°C is exceeded, even for a short duration, tumour necrosis can be 'almost' guaranteed.

These larger temperatures are achieved in one of many ways. Radiofrequency waves can be used to create a current through the body, the ionic flow of current will heat the body temperature due to friction. However, as dehydration occurs due to heating, so does the resistance to ionic current. This cycle often time leads to charring of the entire current flowing pathway and its applicability is limited to small sized tumours. Pulsation techniques are used to mitigate the damage to healthy tissues. Microwave based ablation generates a propagating electromagnetic wave, which eventually increases the tumour temperature due to vibrational heating. Once again, there is minimal control of the heat propagation once a certain temperature is reached. Furthermore, there are multiple reports of skin burns with MW heating. Optical laser based heating and focussed ultrasound based heating are also techniques that are used for cancer ablation. Out of all heating modalities, HIFU holds most promise as a relatively non-invasive option. The drawback in using HIFU is its inability to penetrate into deeper tumours [118].

In contrast, tumour cooling is achieved using the Joule-Thomson effect to rapidly cool down the tumour to the required temperatures. In fact, clinical studies demonstrate that cryoablation appears to have a quicker recovery time than the heating approach. In contrast, certain studies have shown that cryoablation can trigger potential adverse and fatal systemic reactions. Furthermore, due to the lack of coagulation during cryoablation, there is a potential to induce further risks [119–121].

Based on these observations, it is sufficient to conclude that cancer hyperthermia is a much safer treatment modality when compared with cancer ablation.

Cancer hyperthermia

Hyperthermia refers to the cytotoxic increase of body temperature beyond its normal value. The effects of hyperthermic cytotoxicity are known to intensify in de-oxygenated and acidic cellular environments, establishing its potency as an ideal cancer treatment modality. The clinical appeal of hyperthermia is further enhanced by the fact that it does not involve using harmful chemicals or radiation. However, high temperature exposure causes protein denaturation in healthy and malignant cells alike. Therefore, caution must be exercised in clinical hyperthermia to minimize potential harm to healthy cells [122].

Early in-vivo studies based on hyperthermia have reported a high degree of success in treating a variety of tumour types. Based on early analysis, it can be concluded that hyperthermia can be used to directly shrink or at least enhance the susceptibility of tumours to other treatment modalities such as radiation therapy [123]. Furthermore, it is worth noting that these early studies have been conducted by delivering thermal energy to tumours through external mechanisms [124]. Due to the intractable heat propagation in such techniques, there have also been reports of damage to healthy tissues [125–127]. Furthermore, most clinical data available in the literature studies the combined effect of traditional

hyperthermia and another treatment modality such as chemotherapy or radiotherapy. Therefore, it is difficult to quantify the effects of hyperthermia alone.

Due to the potential of heating tumours from within the tumour itself, nanomedicine has recently reawakened the interest towards cancer hyperthermia [26, 128–131]. However, exact temperature and exposure duration requirements to successfully achieve hyperthermia in specific tumours are still unknown. It is reported that achieving temperatures above 40°C and maintaining it for around 30 minutes is sometimes sufficient. Whereas some studies report temperatures higher than 44°C are required for the same task [122]. Due to the novelty of these techniques, human trials are still several years away from being attempted.

Requirement for a controlled thermal pulse

As discussed in the previous sections, even targeted heating of cancer cells will generate undesired results due to intractable heat propagation, which leads to induced hyperthermia of neighbouring healthy cells. The only way to achieve the desired temperature gradient at the tumour as well as in the surrounding healthy tissue, is to control the emission dynamics of the system itself.

The emergence of nanobiophotonics, which is the combination of medicine and nanophotonics, presents us with a myriad of control techniques for heat propagation and delivery. The rest of this chapter shows how the utilization of superradiance provides us with means to tackle this complicated issue and aid us in obtaining a controllable temperature gradient across the tumour and its boundary.

It is our intention that this technique will prompt the clinical establishment of cancer hyperthermia as a first-line treatment modality.

6.2 Optimally Tuning the Superradiant Pulse

Recently, there has been a keen interest in utilizing nanoscopic structures in cancer therapy. The minute nature of these devices theoretically enables minimally-invasive treatment of cancers [132]. Therefore, various attempts are being made at understanding light, heat and matter interactions [133], energy transportation [20,21,134,135] as well as the modelling of such nanoscopic devices targeting this application, ranging from gold nanoshells [136] to nanoscopic lasers known as spasers [137–139].

There are several key areas of significance when designing devices for photothermal cancer therapy. Increased power delivery is one such area that has been in the spotlight for several years [132]. As a solution, we propose the use of steady-state thermal superradiance as an enhanced power delivery tool for photothermal cancer therapy. To the best of our knowledge, this is the first theoretical formulation targeting the utilization of the principles of superradiance into enhancing this application. Another key area of interest with such devices is bio-compatibility. Due to the tunable nature of our emitter assembly, it has the capability to be designed using a wide array of materials as suited for the application. Furthermore, applications of this nature require the maintenance of specific wavelengths to achieve optimal cellular ablation. Once again, the tunable nature of our design can be utilized for this purpose efficiently.

It is noted that the wavelength $1.45\mu m$ in the near infra-red spectrum is optimal for electromagnetic absorption by water, which will in turn be ideal for an application like photothermal cancer therapy [140]. Therefore, this value can be set as the resonant wavelength for a model superradiant emitter assembly.

As an example, consider using a PEC substrate with a slit thickness of $6\mu m$ for the first emitter of the assembly. The resonant frequency spectrum shown in Fig. 6.1 can now be obtained for the various possible modes for this thickness. From the spectrum, it is observed that the resonant wavelength is proximal to the

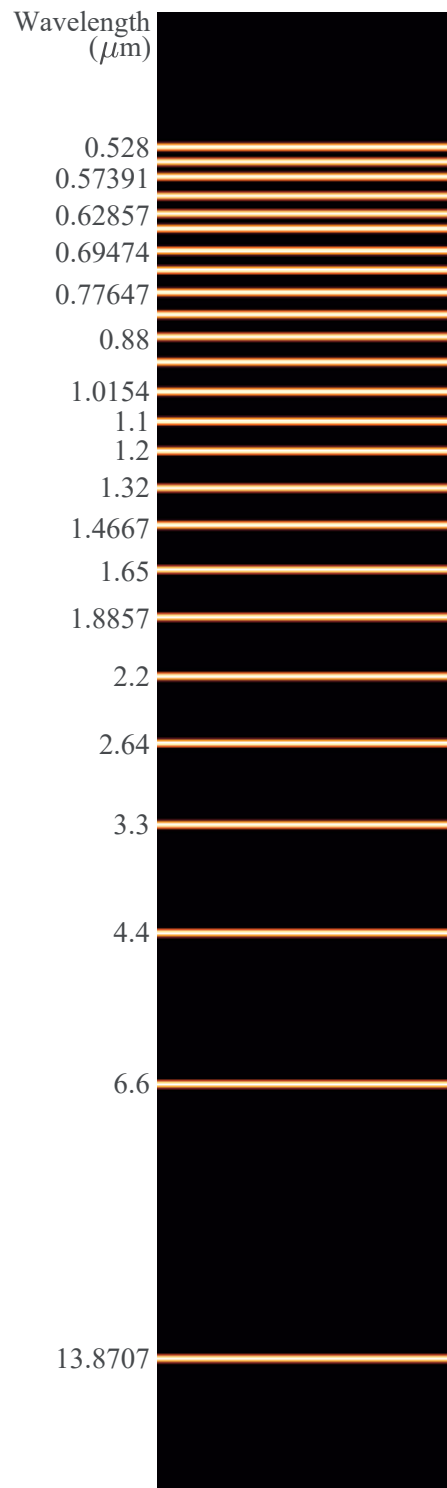


Figure 6.1: Resonant spectrum of the first 25 modes for a single emitter designed using a PEC thickness of $6\mu\text{m}$.

17th mode. By using (4.95), now it is possible to solve for the other dimension of the cavity. In order to maintain superradiance without undergoing anomalous power suppression at resonance as shown in Fig. 5.1, now it is required to vary the thickness of the PEC substrate for the second emitter. This process can be followed iteratively until an emitter with suitable design characteristics is realized.

Design and realization of structures of nanoscopic magnitudes have been a subject of interest in recent times. Owing to the discovery of interesting characteristics of deep subwavelength slits, many attempts have been made towards studying them experimentally. Since our design is entirely based on these deep subwavelength slits, the experimental feasibility is quite high. For instance, lithographic techniques utilizing focused-ion beams may be used with suitable masks to fabricate structures of this nature experimentally [141]. Furthermore, recent improvements in nanoscopic realization makes it possible to prototype nanoscale devices of a few orders of nanometres [142]. This further strengthens the experimental feasibility of our novel emitters, thus paving the way to assess the accuracy and relevance of the analytical results provided herein.

6.3 Tissue Model Simulations

6.3.1 QD ring assembly

The energy emission spectra are shown in Fig. 5.6 (a) and (b) for a superradiant and conventional system, respectively. Throughout the pulse duration ($t \gg \tau$), the conventional system delivers $\approx 3 \times$ the power of the superradiant counterpart at a much slower rate. This results in the leakage of thermal photons beyond the tumour boundary, resulting in the undesired temperature increase in adjacent healthy tissues.

Superradiant pulse effectiveness was validated by comparing it with its con-

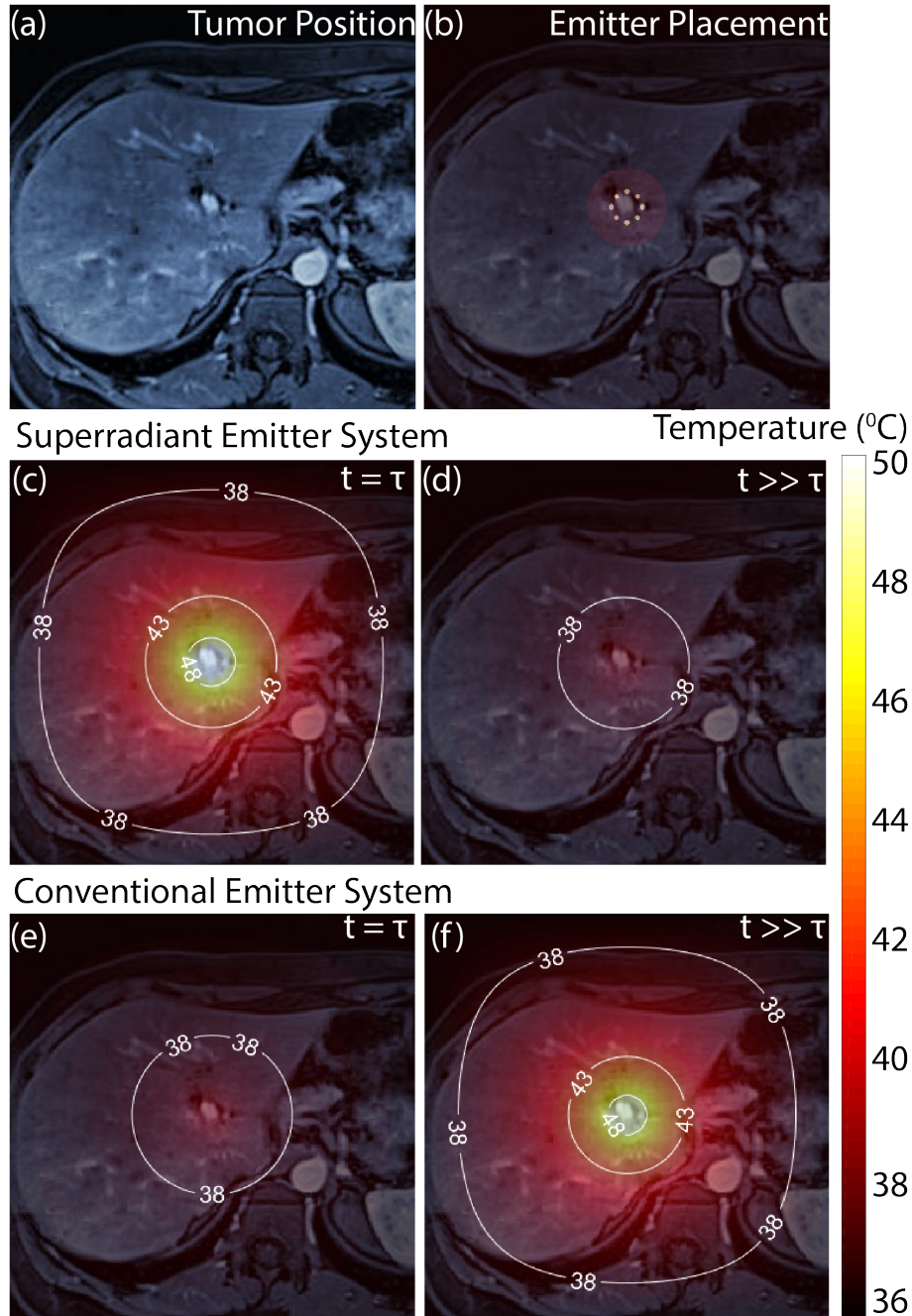


Figure 6.2: Artistic illustration of the thermal characteristics. (a) Modelled liver tumour and (b) emitter placement. Illustration of the thermal performance of the (c,d) superradiant and (e,f) conventional systems.

ventional counterpart. Finite-element and finite-difference techniques were used and the bio-heat equation was solved using values for a liver tumour engulfed within liver tissue as shown in Fig. 6.2(a) [143]. For both studies, 1000 systems were placed within each white spot shown in Fig 6.2(b). As shown in Fig 6.2(c), before $t = \tau$, the superradiant system successfully increases tumour temperature. Although healthy tissue temperature is elevated in this period, as seen in Fig 6.2(d), they return to body temperature at $t \gg \tau$. The situation is reversed for the conventional system and the healthy tissues remain harmfully heated as seen in Figs. 6.2(e) and (f).

This demonstrates that the superradiant QD system enhances cancer hyperthermia compared to a conventional system.

6.3.2 QD buckyball assembly

Initially, it is assumed that there are 3 clearly disparate temperature zones as shown in Fig. 6.3(a). The tumour region is confined to a circle of radius $15\mu\text{m}$ and initially maintains a temperature of 311.15K. The sub-figures throughout Fig. 6.3 maintain the same scale and the human breast outline is overlaid as a purely artistic illustration and its dimensions are ignored.

At $t = 0$, both emitter assemblies have the same conditions as shown in Fig. 6.3(a). During transience, it is observed that the superradiant assembly is capable of effectively increasing the temperature of the tumour region to around 317K as shown in Fig. 6.3(b). This increase is sufficient for successful cancer hyperthermia. Comparatively, the non-resonant assembly barely increases the tumour temperature beyond its starting value during transience as shown in Fig. 6.3(d). This result is clearly in agreement with Fig. 5.11. The high powered and ephemeral superradiant pulse clearly increase the temperature of tumour tissue at the commencement of emission, compared to the conventional counterpart. At

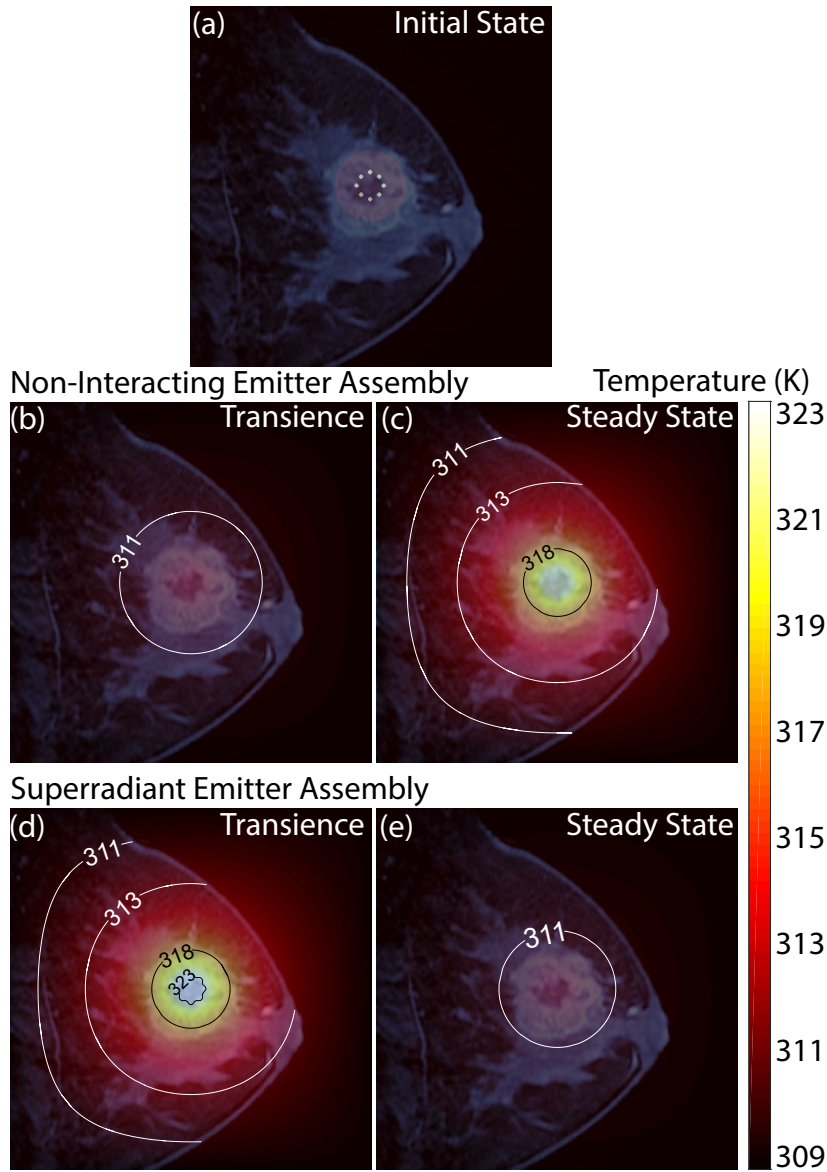


Figure 6.3: Artistically illustrated thermal characterization of the non-interacting and superradiant assemblies. (a) The simulation state at $t = 0$. Three clearly disparate temperature regions are visualized. The emitters, tumour and healthy tissue are at 323.15, 311.15 and 310.15K, respectively. (b,d) During transience, the superradiant assembly has clearly obtained a sufficiently high temperature within the tumor tissue for cancer hyperthermia. The non-resonant assembly has a lower power emission and the temperature increase is marginal. (c,e) At the steady-state, the superradiant assembly maintains the tumour temperature at around 312K while maintaining the healthy tissues at harmless temperatures. In contrast, the non-resonant assembly has increased the temperature of healthy tissues adjacent to the tumour to dangerously high values.

steady-state, it is observed that the superradiant assembly barely increases the normal tissue temperature beyond its starting value as shown in Fig. 6.3(c). Conversely, at steady-state, the conventional assembly has increased the temperature of the adjacent healthy tissue beyond 320K as shown in Fig. 6.3(e). Since the conventional assembly emits a higher number of photons compared to the superradiant counterpart, this result is expected. It is noted that both these results are recorded for the decay of a single energy pulse.

These observations imply that, when using a conventional emitter assembly for cancer hyperthermia, inadvertent hyperthermia of healthy tissues is unavoidable. This is attributed to the slow release of a large number of thermal photons, which leads to undesired temperature rises in adjacent healthy tissues. However, our results suggest that by utilizing a superradiant pulse instead, it is possible to significantly reduce the hyperthermia of adjacent healthy tissues. Therefore, using our proposed assembly to repeatedly deliver a superradiant pulse of heat from within the tumour environment will enhance cancer hyperthermia over using its conventional counterpart.

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Chapter 7

Summary of Contributions and Suggestions for Future Work

THis chapter summarizes the contributions made to the scientific literature through the work presented in this thesis, followed by recommendations for future research work.

7.1 Summary of Contributions

The focus of this thesis has been to design a high density nanoparticle assembly that can generate a controlled superradiant thermal pulse, which can be used to enhance cancer hyperthermia by reducing the damage to healthy tissues around the tumour. A sound theoretical understanding of superradiance and nanophotonic thermal emission was extremely important towards achieving the goal.

Presenting a tuneable model for superradiant nanophotonic thermal emission

In chapter 4, we obtained analytical characterisations for a semi and a fully generalized assembly of nanophotonic emission. The framework for the mathematical characterisation was based on coupled mode theory, which was presented in 3.

The semi-generalized model analysed a scenario where multiple nanoparticles, each comprising the same type of material, were emitting superradiantly.

Each nanophotonic emitter was modelled based on a Fabry-Perot like cavity, which allows the resonant frequencies to be perturbed, without coupling with modes which are sufficiently further away from the main resonant mode. This analytical characterisation and the resulting simulation proved that thermal superradiance, when controlled by perturbing the resonance frequency within individual emitters, can emit a sufficient amount of energy to fulfil hyperthermia requirements. These results were published in *Physical Review B* [82].

Observing startling phenomena in superradiant nanophotonic thermal emission

The fully-generalized model analysed a similar scenario involving multiple nanoparticles, however, the theoretical limitation of adhering to the same material type assumption was lifted. This allowed superior flexibility over the previous model and depicted EIT-like and superscattering like characteristics, which were never before observed in a similar analysis. These results were published in *Physical Review B* [105].

Analysing the suitability of a superradiant pulse generated by a QD-ring type emitter assembly

Unlike the previous PEC based hypothetical simulation scenarios, this model used experimentally measured QD parameters to generate a controlled superradiant thermal pulse. The actual number of photons generated by a single pulse was statistically calculated to obtain the amount of thermal energy generation. While the ring structure was previously proven to be superradiant in the literature, its application for cancer hyperthermia was novel. The results were validated using a simple tissue model simulation to analyse the temperature variation across the tumour and the healthy tissue around it. These results were presented at the *IEEE Nanomed Conference* conference in Hawaii, USA [144].

Presenting a buckyball type quantum dot emitter assembly as a high density superradiant thermal emitter suitable for cancer hyperthermia

In the literature, due to the complexities associated with fulfilling stringent criteria, very few structures are known to be capable of depicting superradiance. We presented a complete mathematical characterisation for the proposed emitter assembly model, which can pack thermal emitters at a much higher density in a 3-dimensional configuration, in contrast to the famous ring structure. The photon statistics were calculated to obtain the amount of energy within a single superradiant pulse and a breast tissue model was used to validate its superiority over conventional emitter assemblies. These results were published in the *IEEE Journal of Selected Topics in Quantum Electronics* [26].

The study of absorption, emission and scattering phenomena of various nanoparticle systems

Throughout this thesis, various mathematical studies were conducted to investigate the absorption, emission and scattering spectra of various nanoparticle systems. An absorption spectra study was conducted on PNIPAM capped gold nanorod particles and the results were experimentally verified. These results were published in the *Journal of Materials Chemistry C* [100]. Results pertaining to the emission characteristics of quantum dot structures and generalized perfect electric conductor structures were presented previously. Results pertaining to the scattering spectra of a metal nanoparticle - quantum dot hybrid structure were presented in *The Journal of Physics - Condensed Matter* [99]. A similar scattering study involving a hybrid graphene nanoflake and a carbon nanotube were presented in the same journal [145].

7.2 Recommendations for Future Research

The work presented in this thesis can be further extended in both theoretical and experimental areas. As a guideline, a number of possible suggestions are listed as follows:

Formulation of a fully quantum mechanical analysis for coupled mode theory

This is a completely abstract area of significant potential towards the advancement of nanophotonics in general. Until two decades ago, the applicability of coupled mode theory towards modelling nanophotonic emission was not conceptualized. Since then, the versatile nature of coupled mode theory has been well established in the literature. However, a fully quantum mechanical derivation of the basic coupled mode theory does not exist.

Study other potential geometric structures suitable for thermal superradiance

The 2-D ring structure is the only established superradiant structure presented in the literature. Identifying newer superradiant structures can significantly improve the domain knowledge in the field of nanophotonics. Furthermore, these new structures may have the tendency to depict characteristics beyond the conventional understanding of superradiance.

Modelling the electronic structure of the proposed assemblies using DFT

Using a DFT approach, it is possible to model the electronic structure of the proposed buckyball shaped quantum dot based emitter assembly. Due to the high symmetry of the assembly, studying its electronic structure may unveil information to further the field of nanophotonics.

Developing advanced tissue models for cancer thermal simulations

The main scope of this research was in designing a device capable of generating a controlled pulse of thermal energy. Although tissue models developed based on experimentally obtained data were used to validate the flow of heat photons across the tissues, there is a requirement for an advanced model with higher accuracy and complexity to further validate these results.

Experimental validation of the proposed superradiant structure

The 2D ring structure and the buckyball structure are both yet to be validated by experimental observations. Recent developments in the area of quantum dot research shows significant potential towards the possibility of validating these two structures. In fact, the current state of technology should technically be sufficient to validate the 2D ring structure and investigate its superradiant capabilities.

Experimental study of nanoparticle based cancer hyperthermia

Several human trials utilising nanoparticles have been undertaken in the past with satisfactory levels of success. In fact, several nanomedicine drugs have already been approved and have proven clinical success over their conventional counterparts. Hyperthermia, without the use of nanoparticles, has also been attempted on humans numerous times with strong positive outcomes. However, as of now, cancer hyperthermia involving nanoparticles has only been attempted on animal subjects a handful of times. Continuing in its positive trajectory, these studies too have reported highly favourable outcomes. Although not a direct extension of this thesis, an investigation of nanoparticle based cancer hyperthermia on human patients will have a high impact on this area of research.

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