Quantum Electrodynamics of a Driven Three-level Atom Near the Edge of a Photonic Band Gap

by

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A thesis submitted in conformity with the requirements for the Degree of Doctor of Philosophy
Graduate Department of Physics
University of Toronto

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God said, "Let there be light," and there was light. God saw the light, and saw that it was good. God divided the light from the darkness. God called the light Day, and the darkness he called Night. There was evening and there was morning, one day.

Genesis 1.3 - 1.5
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Abstract

In the first part of this thesis, the coherent control of spontaneous emission for a three level atom located within a photonic band gap (PBG) material is demonstrated. Spontaneous emission from the three level atom can be totally suppressed or strongly enhanced depending on the relative phase between the steady-state control laser coupling the two upper levels and the pump laser pulse used to create an excited state of the atom in the form of a coherent superposition of the two upper levels. Unlike the free space case, the steady-state inversion of the atomic system is strongly dependent on the externally prescribed initial conditions. This non-zero steady state population is robust to decoherence effects provided that the Rabi frequency of the control laser field exceeds the rate of dephasing interactions. As a result, such a system may be relevant for a single-atom, phase sensitive, optical memory device on the atomic scale. Provided that coherence can be maintained between the two upper atomic levels, the model system can also act as a qubit to encode information for quantum computation.

In the second part of this thesis, the resonance Raman scattering of light from a three-level atom in the $\Lambda$ configuration embedded in a photonic band gap material is studied as a direct experimental probe for the photon-atom bound state discussed in the first part. The one particle spectrum of the system is demonstrated to consist of either a continuous part with energy lying outside the gap or a single discrete mode with energy lying inside the gap. The discrete mode, which occurs when both of the allowed atomic transitions lie inside the...
gap, can be treated as a photon-atom bound state in which the radiation is localized in the vicinity of the atom. In the case of the continuous spectrum, the Rayleigh and Stokes lines are shifted as well as narrowed (or broadened) as the corresponding transition frequencies are shifted relative to the upper band edge, providing a distinctive experimental signature of atom-photon interactions near a photonic band edge.
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4.9 V system coherence $n_c(t) = |c_2(t)c_1^*(t)|$ between levels $|2\rangle$ and $|1\rangle$ near the isotropic photonic band edge as a function of the scaled time $\beta t$ for $\theta = \pi/4$, $\phi = -\pi/2$, and for different values of $\Omega$, in the presence of dipolar dephasing Gaussian random Stark shifts $\delta \omega_{20}(t)$ and $\delta \omega_{10}(t)$ (each of zero mean and $0.5\beta$ variance) of the transition frequencies $\omega_{20}$ and $\omega_{10}$. In the absence of the random Stark shifts, the band edge is assumed to be midway between the two upper levels with detuning $\delta_{20} = -\delta_{10} = 0.5\beta$. Compare this figure with the corresponding figure (Fig. 4.7) in the absence of phonon mediated dephasing.

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5.4 The spectrum of spontaneous emission \( \sigma_R(\omega) \) for the Rayleigh transition \( |2\rangle \rightarrow |0\rangle \) in the case of the PBG described by the isotropic dispersion relation (5.74). We take \( r = 15\% \), \( \omega_{20} = 1.2\omega_c \), and \( \gamma_{20} = \gamma_{21} = 0.01\omega_{20} \). Thus \( \omega_{20} \) is removed from the band edge \( \omega_c \) by more than the width of the gap. We plot \( \sigma_R(\omega) \) for different values of the Stokes frequency \( \omega_{21} \). The dot-dashed curve represents both equations (5.73) and (5.64) when \( \omega_{21} = 1.15\omega_c \). Thus the effect of the gap is negligible for transition frequencies removed from the band edge by at least the width \( \Delta \) of the gap.
5.5 The photon density of states in a dispersive medium described by the dispersion relation (5.84). The dashed curve represents the free space density of photon states. We see that \( \rho(\omega) \) is highly singular at the lower band-edge \( \omega_v \) behaving like \( \rho(\omega) \sim (\omega_v - \omega)^{-4} \), but is identically zero at the upper band edge, \( \rho(\omega_c) = 0 \). This should be contrasted with the PBG case shown in Fig. 5.2 where \( \rho(\omega) \) exhibits square-root singularities at both band edges. In the inset is shown the photon spectrum for a dispersive medium described by the dispersion relation (5.84).

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5.7 The spectrum of spontaneous emission \( \sigma_R(\omega) \) for the Rayleigh transition \( |2\rangle \rightarrow |0\rangle \) in the case of FDM described by the isotropic dispersion relation (5.84). The parameters \( r = 15\%, \omega_{20}, \gamma_{20}, \) and \( \gamma_{21} \) have the same values as in Fig. 5.4. We plot \( \sigma_R(\omega) \) for different values of \( \omega_{21} \) (\( \omega_{21} = 1.15\omega_c, 1.05\omega_c, \omega_c, \) and \( \omega_o \)) of the Stokes frequency. On the scale of this figure, all cases give the same spectral distribution cut off at \( \omega = \omega_v \) and a peak value which is less than the free space case (shown in the inset) by about three orders of magnitude.

A.1 Schematic representations of a three-level atom (a) in the \( \Xi \) or cascade, (b) in the \( \Lambda \), and (c) in the \( \Gamma \) configurations. Dashed lines with arrows denote dipole allowed transitions.
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Chapter 1

Introduction

Photonic Band Gap (PBG) materials are periodic dielectric structures that exhibit a range of frequencies for which electromagnetic wave propagation is classically forbidden. These systems lead to strong localization of light at the classical level, suppression of spontaneous emission, and formation of a photon-atom bound state, a strongly coupled eigenstate of the electronic degrees of freedom of an atom and the electromagnetic modes of the dielectric. Near a photonic band edge, spontaneous emission is anomalous and leads to fractionalized steady-state inversion for a single atom.

The suppression of spontaneous emission combined with the coherent localization of light in a PBG material leads to interesting phenomena in quantum optics as well as important technological applications. In the visible and near-infrared wavelength regimes, PBG materials have numerous applications in the telecommunications industry. These applications include the design of zero threshold, highly efficient microlasers and light emitting diodes, low threshold optical switches, novel resonators and cavities, highly efficient microwave antennas, and all-optical transistors.

The absence of single photon spontaneous emission for an isolated atom in a PBG guarantees that the emitted photon remains partially localized in the vicinity of the emitting atom leading to the formation of a photon-atom bound state. On the other hand, driving a multi-level atom with a sufficiently strong resonant field alters the radiative dynamics
in a fundamental way, even in ordinary vacuum. It leads to such interesting effects as the enhancement of the index of refraction with greatly reduced absorption, electromagnetically induced transparency and optical amplification without population inversion. In view of these results, it would be interesting to investigate the combined effects of coherent control by an external driving field and photon localization facilitated by a PBG on spontaneous emission from a three-level atom embedded in a PBG material. This is precisely what I have done in this thesis. I have demonstrated that storage of quantum information in a single three-level atom is facilitated by the localization of light in the vicinity of the atom when one of the atomic transitions lies within a photonic band gap. The nature of this stored information is controlled by the combination of quantum interference between different radiative pathways within the atom, mediated by the external laser field, and photon localization effects, mediated by the PBG. This suggests an application of the model system as a single-atom optical memory device. Provided that coherence can be maintained between the upper atomic levels, the model system can also act as a qubit to encode information for quantum computation.

In Chapter (2) an extensive literature survey is presented to provide context for this research. Here I review the nature, fabrication and applications of PBG materials.

Chapter (3) introduces the model system in the leading approximation, whereby a number of spontaneous emission effects and non-radiative interactions are neglected. I compare my results with the known results for the case of vacuum in order to isolate the effects of the PBG, and the resulting new physics and applications. Chief among these is the application of the model system as a memory device on the atomic scale.

The effects of higher order terms on the basic results found in Chapter (3) are discussed in Chapter (4). This includes a semi-quantitative analysis of phonon-relaxation mechanisms for the model system. I have demonstrated that such dephasing effects can be offset by intense driving fields. Different ways of realization of the model system are also discussed in Chapter (4).

The resonance Raman scattering of light from a three-level atom embedded in a photonic
band gap material is studied in Chapter (5). I have demonstrated that the spectrum of the Raman scattered light contains features that can be used as a direct experimental probe for the photon-atom bound state. I also studied the same Raman scattering problem, in the context of a frequency dispersive medium (FDM) such as NaCl whose photon spectrum exhibits a gap due to photon coupling to medium excitations such as excitons and optical phonons. The results are compared and contrasted with the results in a PBG, where the photonic band gap results from multiple photon scattering by a periodic array of scatterers. I have demonstrated that, owing to the difference in the behavior of the photon density of states near a band edge, the Raman spectrum in the FDM case is highly distinct from that in the PBG case.

The Appendices contain supplementary material. The Hamiltonian of a three-level atom interacting with a quantized radiation field is derived in Appendix (A). The various expressions for the Green’s functions used in the main body of the thesis are calculated in Appendix (B). Solution of the relevant equations of motion by means of Laplace transformation and the complex inversion formula is carried out in Appendix (C). Details of the quantum beats problem for both the free space and PBG cases are discussed in Appendix (D). The two-level atom is discussed in Appendix (E) as a special case of a three-level atom. The density of photon states is derived in Appendix (F) for both a PBG material and a frequency dispersive medium.
Chapter 2

Photonic Band Gap Materials: Introductory survey

2.1 Introduction

A new paradigm has emerged in which the band structure concepts of solid-state physics are applied to electromagnetism, leading to the invention of artificial electromagnetic crystal structures. These structures are called photonic crystals (PC’s) and were originally proposed by S. John and E. Yablonovitch as a means to realize two fundamentally new optical principles - the localization and trapping of light in bulk material[1, 2], and the complete inhibition of spontaneous emission[3, 4] over a broad frequency range.

A photonic crystal is the photonic analog of an electronic crystal. Rather than a periodic array of atoms which scatters and modifies the energy-momentum relation of electrons, a PC consists of a three-dimensionally ordered dielectric structure having spatially periodic dielectric constant, with a lattice parameter comparable to the wavelength of the electromagnetic wave. The band structure of a photonic crystal is referred to as a Photonic Band Structure (PBS). Provided that the conditions of sufficiently high index-contrast between the high- and low-index regions, appropriate spatial structure, and dielectric filling ratio are met, photonic states inside a PC will be classified into bands separated by band gaps. These frequency gaps are termed photonic band gaps (PBG’s) and a photonic crystal with
Partially infiltrated Si inverted Opal. $n_{Si}=3.45$. $f_{Si}=0.25$

\[
\frac{\omega a}{2\pi c} = \begin{cases} 
0.1 & L \\
0.2 & U \\
0.3 & X \\
0.4 & K \\
0.5 & W \\
0.6 & L \\
0.7 & U \\
0.8 & X \\
0.9 & 0.9
\end{cases}
\]

Bloch vector

Figure 2.1: The photonic band structure for the lowest ten bands of a PBG material made of Silicon ($n = 3.45$) inverse opal. The filling ratio of Silicon is 25%. In units of $\omega a/2\pi c$, where $a$ is the lattice constant, the width of the gap is 0.822 whereas the gap-midgap ratio is 8.94%. (Courtesy of Ovidiu Toader, Dept. of Physics, University of Toronto)

A PBG is often referred to as a **PBG material**. Fig. 2.1 shows the photonic band structure for the lowest ten bands of a PBG material made of Silicon ($n = 3.45$) inverse opal. The photonic band gap occurs between the eight and ninth bands.

The concepts of reciprocal space, Brillouin zones, dispersion relations, Bloch wave functions, Van Hove singularities etc. of solid state physics are now being applied to PBG materials[5, 6, 7, 8, 9]. Unlike electrons in a semiconductor crystal which are constrained by Fermi statistics and therefore have to be excited from the valence band to the conduction band to become mobile, photons are bosons which propagate freely at frequencies both above and below the photonic bandgap. Thus the terms "valence band" and "conduction band" may not be appropriate in the context of a photonic crystal. Instead, the bands above and below a photonic band gap can be distinguished by applying the electromagnetic variational theorem[10]. According to this theorem, for modes in the lower photonic band, the power of modes lies primarily in the high index regions, whereas for modes in
the upper photonic band the power lies in the low index regions. In photonic crystals, the low index regions are often air regions. For this reason it is more meaningful to refer to the band above a photonic band gap as the "air" band, and the one below the gap as the "dielectric" band.

Most of the promising applications of PBG materials depend on the widths and locations of their photonic band gaps. The gap size in a PBG material is determined by the refractive index contrast of the two materials that constitute the 3-D structure, and by the filling ratio of the higher index material\[11\]. The location of the gap is determined by the lattice constant of the photonic crystal. For a face centered cubic (fcc) lattice, the gap is centered at roughly twice the index modulation wavelength.

The extent of a PBG is characterized by a dimensionless parameter called the \textit{gap-midgap ratio}. It is the ratio \( r \equiv \Delta \omega / \omega_o \) of the width \( \Delta \omega = \omega_c - \omega_o \) of the gap to the \textit{mid-gap} frequency \( \omega_o \), where \( \omega_o \) and \( \omega_c \) are the lower and upper band edge frequencies of the gap, respectively. This characterization of the extent of a PBG is independent of the scale of the crystal. If the system is compressed (expanded) by a factor \( s \), all the relevant frequencies (\( \omega_o, \omega_o, \) and \( \omega_c \)) will increase(decrease) by the same factor so that the \( r \) stays the same. For example, a gap-midgap ratio of \( r = 10\% \), which is readily achievable in present day PBG materials\[12\], translates to a gap width of about 0.2 eV at an optical mid-gap frequency\[1\]. By contrast the electronic band gap of Germanium at room temperature is 0.67 eV, while that of GaAs is 1.43 eV\[13\].

For the frequency range spanned by the gap, a PBG material is completely free of propagating electromagnetic modes. Put another way, \textit{the density of propagating photon modes is absolutely zero within a photonic band gap} [see Fig. (2.2)]. The effect is analogous to the existence of gaps in the electron density of states in crystalline solids. By contrast, in free space (or a cavity of infinite volume), the density of photon modes \( \rho(\omega) \) varies as \( \omega^2 \) and exhibits no gap. In a cavity of finite volume, the density of states is substantially modified for frequencies close to the cavity cutoff. Below cutoff the cavity sustains no modes at all\[2\],

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\[1\] The energy \( h\omega \) of an optical transition \( \omega \sim 2\pi \times 10^{15} \text{ Hz} \) is of the order \( E \sim 2 \text{ eV} \)

\[2\] A laser will oscillate in a cavity only if the round trip phase shift \( \Delta \phi \) is an integral multiple of \( 2\pi \) or the round-trip optical path length \( l \) is an integral number of wavelengths. For a linear bare-cavity of length \( L \), we have \( \Delta \phi = 2\pi (2L/c)\nu \), and \( l = 2L \).
and near and above cutoff, the density of states can be increased relative to the continuum case[14].

The absence of propagating electromagnetic modes, in a refractive medium without dissipation, is due to a synergetic interplay between the microcavity resonances of individual dielectric particles of the photonic crystal and the Bragg scattering resonance of the array. One of the conditions for the appearance of a PBG is that the density of dielectric scatterers be chosen such that the microscopic (Mie) scattering resonance of a single unit cell of the photonic crystal occur at the same frequency as the macroscopic (Bragg) resonance of the periodic array[15]. This is a highly restrictive condition in three dimensions. It may be illustrated [see Fig. 2.3] by a simple example of one-dimensional wave propagation through a periodic array of square wells of width $a$ and spaced by a distance $L$. Suppose the refractive index is $n$ inside each well and is unity outside. Then the Bragg scattering condition is given by $\lambda = 2L$ where $\lambda$ is the vacuum wavelength of light. The analog of a Mie resonance in one dimension is a maximum in the reflection coefficient from a single well and this occurs

The axial resonant modes of the cavity are thus $\nu_m = mc/2L$; and the cavity cut-off is $\nu_c = c/2L$. 

Figure 2.2: Summary of photon density of states in different cases. For free space or independent point scatterers (dotted curve) there is no gap in the photon density of states. For a PBG material (solid curve), there is a complete photonic band gap between the band edge frequencies $\omega_1$ and $\omega_2$. A cavity of finite volume sustains no modes below cutoff, whereas near but above cutoff, the density of states can be increased relative to the free space case.
Figure 2.3: The photonic band gap arises from a synergetic interplay between macroscopic and microscopic resonances. This effect is maximized when the lattice constant $L$ and the sphere radius $a$ are chosen in such a way that the two resonance conditions coincide.

when a quarter wavelength fits into the well: $\lambda/(4n) = 2a$. Combining these two conditions yields the optimal volume filling fraction $f \equiv 2a/L = 1/(2n)$. The generic form of the magnitude of the photonic band gap with volume filling fraction $f$ of dielectric material is shown in Fig. 2.4.

The vanishing of the density propagating photon modes within a PBG means that, for the frequency range spanned by the gap, *linear propagation of electromagnetic waves is forbidden in any direction* in the PBG material. Thus, light incident on a PBG material with a frequency in the gap region will be backscattered from the material, independent of the angle of incidence. In other words, a PBG material acts as a $4\pi$ steradian stop band for the frequency range spanned by the PBG. This effect is again analogous to the strict prohibition of propagating electron waves in conventional crystals for a frequency range spanned by an electronic band gap. Strong suppression of transmission, with an associated peak in the reflectivity at the characteristic frequencies, is then an experimental signature of a photonic band gap[3].

Although photonic band structures are analogous to electronic band structures, there
Figure 2.4: The gap-midgap ratio $r = \Delta \omega/\omega_o$ of a photonic crystal as a function of the volume filling fraction $f$ of the solid material. The maximum gap occurs at $1/(2n)$ where $n$ is the refractive index of the solid.

are major differences between them[16]:

- Electrons are massive, and, therefore, the underlying dispersion relation for electrons in crystals is parabolic. Photons have no mass, so the underlying dispersion relation is linear.
- Conventional crystals occur naturally whereas photonic crystals are artificial and should be designed using ingenuity.
- Electronic band structure is obtained by solving the appropriate Schrödinger equation. On the other hand, photonic band structure is obtained through Maxwell’s equations.
- Electrons have spin $1/2$ and are fermions. However, frequently, this spin is ignored in band structure calculations, and Schrödinger’s equation is treated in the scalar wave approximation[13, 17]. In contrast photons are spin 1 bosons and it is generally never a good approximation to neglect the vector (polarization) character of the electromagnetic field (that is to treat the two polarizations of the field independently) in band structure calculations[18, 19]. One consequence of the vector nature of the electromagnetic field is that the band structures for \textit{transverse-electric} (TE) and
\textit{transverse-magnetic} (TM) modes can be completely different. In particular, there can be photonic band gaps for one and not for the other\cite{10}. To have a \textit{complete band gap} for all polarizations, a photonic crystal should not only have TM and TE band gaps, but these band gaps should also overlap. While there are a number of techniques for band structure calculations of electronic crystals\cite{17}, photonic band structure calculations are largely based on the plane-wave expansion of the electromagnetic fields and use of Bloch's theorem to reduce the problem to the solution of a set of linear equations\cite{9, 10, 12}.

- Electronic band structure is based on the description of a single non-interacting electron. The effect of all other electrons is combined in the potential energy and effective mass. Thus, the band theory of electrons is only an approximation owing to electron-electron interactions. When there are strong correlations between electrons, as in high $T_c$ super-conductors, band theory is not even a good zeroth order approximation. In contrast, photonic band theory is essentially exact, since interactions between photons are negligible (unless the medium is non-linear or the electromagnetic fields are very strong). Thus, photonic band structure calculations provide an accurate description of experimental data.

PBG materials have been attracting considerable attention of the scientific and engineering community due to their potential capabilities which are of immense practical and commercial importance. There have already been a number of books\cite{10}, special journal issues\cite{20, 21, 22}, conference proceedings\cite{23, 24, 25, 26}, bibliographies\cite{27}, and surveys\cite{28, 29, 30}. A lot of applications have been proposed\cite{10, 24, 25, 31}, including those which would considerably enhance the performances of quantum electronic devices such as semiconductor lasers, and those which would drastically reduce the sizes of devices such as couplers, beam splitters, filters, cavities and lenses, paving the way for the integration of a large number of highly compact optical components onto an all-optical micro-chip.
2.2 Quantum electrodynamics in vacuum and in cavities

Quantum electrodynamics (QED) deals with the interaction of electromagnetic radiation with atomic (and molecular) matter, and with interactions between atoms[32]. Its key feature is that the electromagnetic field, as well as the atomic matter, is quantized, the particles associated with field quantization being photons. QED is, perhaps, the most successful theory known at present and, in applications so far made, its results are in excellent agreement with experiment. In this section we briefly consider QED in vacuum and in microcavities.

2.2.1 Vacuum Fluctuations

The electromagnetic vacuum is characterized by the absence of photons - the mean value of the electric (or magnetic) field at any given point in vacuum is identically zero. However, due to the quantum mechanical nature of the field, the root-mean-square deviation of the electric (or magnetic) field in vacuum is different from zero. This means, for example, that if we perform one measurement of the field in vacuum, it is possible to find randomly varying non-zero results. We say that the “vacuum state” of photons is subject to vacuum fluctuations or zero-point fluctuations (ZPF’s).

One effect of ZPF’s is that, even in the absence of incident photons, they can perturb an excited atom to fall back to a lower energy state by emitting a photon, the energy of the global system being conserved in the process. This is the well known phenomenon of spontaneous emission[33, 34, 35, 36]. Most of the light around us is ultimately the result of spontaneous emission. The phenomenon goes by various names, depending on the context. The term luminescence, for instance, is often used to describe spontaneous emission from atoms or molecules excited by some means other than by heat. If excitation occurs in an electric discharge such as a spark, the term electroluminescence is used. If the excited states are produced as a by-product of a chemical reaction, the emission is called chemiluminescence, or if this occurs in a living organism, bioluminescence, a good example being the luminescence of fireflies. Photoluminescence or fluorescence refers to spontaneous emission
from an excited state produced by the absorption of light. *Phosphorescence* describes the situation in which the spontaneous emission persists long after the exciting light is shutoff, and is associated with a meta-stable (long-lived) level[37].

Another effect of ZPF's is to impart to the atomic electrons an erratic motion which slightly modifies the energy of the atomic levels. This modification in the energy levels due to the coupling of the atom to the electromagnetic vacuum is known as the *Lamb shift* and was of utmost importance for the development of QED. To explain the Lamb shift, it was necessary to assume that the electrons in an atom were continually emitting and re-absorbing photons. The emission and absorption processes are virtual in that the associated energies are not subject to energy conservation. Such photons are called virtual photons and energy shifts caused by virtual processes are termed self energy.

### 2.2.2 QED in Markovian and non-Markovian photon reservoirs

Atom-photon reservoir interaction depends crucially on the behavior of the density of photon modes of the reservoir, $\rho(\omega)$, near the relevant atomic transition frequency $\omega_0$. If, near $\omega_0$, $\rho(\omega)$ is a smoothly varying function of frequency, the atom-reservoir interaction will be characteristically *Markovian*[14, 39]. In such an interaction, the future of the atomic system is entirely determined by the present and not by the past, that is *the atom loses all memory of its past*. Moreover, the spontaneous emission from the atom may be described by the well known Wigner-Weisskopf formalism[40]. In this formalism, spontaneous decay is characteristically *exponential* (with a decay rate $\gamma$), the spectrum of the spontaneously emitted photons has a *Lorentzian* shape of half width $\gamma$ centered at the radiatively shifted frequency $\omega_0 + \delta\omega_0$ (where $\omega_0$ is the bare atomic transition frequency); and both the $\gamma$ and $\delta\omega_0$ depend only on the density of modes in the photon reservoir. As an example, atom-photon interaction in free space is Markovian, free space being an infinitely broad featureless photon reservoir.

On the other hand, if, near $\omega_0$, $\rho(\omega)$ changes on a frequency scale comparable to the

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3According to the uncertainty relation between energy and time[38] energy is sharply defined only when a measurement is performed over a sufficiently long period of time. It is thus completely consistent with energy conservation that an electron can emit a quantum even without having the necessary energy, as long as the quantum is re-absorbed quickly enough.
spontaneous-emission rate (estimated on the basis of the local photon mode density), the atom-reservoir interaction may be *Non-Markovian*. In this case, the Wigner-Weisskopf approximation can no longer be used, and the atomic decay necessarily becomes *non-exponential* and the emission spectrum *non-Lorentzian*.

There are several situations in which the Wigner-Weisskopf approach breaks down[41, 42]. One such situation occurs in a microcavity where the cavity decay rate is much less than the spontaneous emission decay rate of the atomic system so that there will be an oscillatory exchange of the energy between the atomic and photonic degrees of freedom before the spontaneously emitted photon leaks out of the cavity. The atom-photon interaction is then sufficiently strong to split the atomic level into a doublet which, in turn, leads to the splitting of the spontaneous emission spectrum. Such a splitting is known as the *vacuum Rabi splitting*[43]. The qualitative change of the spectrum from a single Lorentzian peak into a two-peaked structure is a clear indication of the onset of a non-Markovian (non-exponential) decay.

Non-Markovian system-reservoir interaction is also expected to occur in the case when the density of photon modes of the reservoir exhibits a threshold-like behavior, that is, when $\rho(\omega)$ exhibits a sudden jump or some weaker kind of singular non-analytic behavior. Such a behavior occurs, for example, in a waveguide close to its fundamental frequency. If the atomic transition frequency lies close to the threshold, non-exponential (usually algebraic) temporal behavior dominates the whole decay process, leading to strong modifications in the shape of the spontaneous-emission spectrum. Specifically, the emission spectrum becomes non-Lorentzian (non-Markovian) and may even exhibit additional peaks or valleys[41, 42].

In this thesis, we consider a non-Markovian interaction caused by photonic crystal possessing gaps in its photonic density of modes. Quantum electrodynamics in such a reservoir is the subject of the next section.
2.3 Quantum electrodynamics in a PBG

The abrupt vanishing of the photon density of states inside a PBG leads to fundamental modifications in the QED properties of atoms and molecules with transition frequencies in the band gap or near a band edge. The most important of these modifications are the inhibition of single-photon spontaneous emission and the localization and trapping of light in a bulk matter. In this section, these fundamentally new optical principles are discussed in some detail.

2.3.1 Inhibition of single-photon spontaneous emission

Spontaneous emission rate was, at one time, regarded as an intrinsic property of a material over which we have no control[40]. In spectroscopy, it gave rise to the term natural line width. However, in 1946 Purcell[44] already suggested that the spontaneous emission rate of radiating dipoles can be tailored by using a cavity to modify the dipole-field coupling and the density of available photon modes. If the modal density in the vicinity of the frequency of interest is greater than that of free space, the spontaneous emission will be enhanced (Purcell effect); if it is less spontaneous emission will be inhibited. This important concept is now well established thanks to the experimental and theoretical development of cavity quantum electrodynamics[26, 45, 46, 47].

The challenging application of Purcell’s concept to optoelectronics has been intensively pursued. A number of interesting experimental[45, 48] and theoretical[49] studies with metallic cavities have shown the basic soundness of the idea of engineering atomic spontaneous emission by imposing boundary conditions on the electromagnetic field other than those of free space. A microcavity with perfectly reflecting walls can considerably inhibit or enhance spontaneous emission of atoms placed inside it, depending on whether the cavity is tuned to or detuned from the relevant atomic transition frequency[25, 46, 50]. Spontaneous emission is predicted[48] to be eliminated altogether from an atom placed in a waveguide, provided the atomic transition frequency is below the fundamental frequency of the waveguide.
Although extremely interesting in their own right, metallic cavities are less important in practice, because they do not scale well into optical frequencies. At high frequencies metallic cavities become more and more lossy (because metals are transparent in the ultraviolet[51]). Moreover simple geometries for the boundaries, such as single- and parallel-plane mirrors do not lead to suppression of spontaneous emission in all directions and, therefore, entail only minor modifications of the spontaneous emission rate[52, 53, 54]. On the other hand, photonic crystals made of positive-dielectric-constant materials (such as glasses and insulators) can be almost free of dissipative losses at any prescribed frequency. Moreover, 3-D PBG materials are able to confine optical waves in all three dimensions.

A rough picture of spontaneous emission in a PBG material follows from Fermi's Golden rule. Suppose that we have a single two-level atom in an initial excited state written as $|i, 0_k\rangle$, where $0_k$ indicates the absence of photons of wave vector $k$. Let the final state of the system consist of the atom in the final state $|f\rangle$, after the emission of a single photon of wave vector $k$. The final state of the system is then $|f, 1_k\rangle$. In the weak atom-field coupling regime, the atomic spontaneous emission rate is given by Fermi's Golden rule[55]:

$$ W_{fi} = \frac{2\pi}{\hbar} |\langle f, 1_k|d.E(\omega_0, r_0)|i, 0_k\rangle|^2 \rho(\omega_0). $$(2.1)

Here $\hbar$ is Planck's constant, $d$ is the electric dipole moment operator for the atomic transition, $E(\omega_0, r_0)$ the electric field operator at the dipole frequency $\omega_0$ and position $r_0$, and $\rho(\omega_0)$ is the density of electromagnetic states at the dipole frequency $\omega_0$ available for the spontaneously emitted photon. Eq. (2.1) is valid, provided that $\rho(\omega)$ is smooth in the vicinity of $\omega_0$.

Eq. (2.1) shows that the spontaneous emission rate can be enhanced, attenuated, or even suppressed by changing $\rho(\omega_0)$ and/or the matrix element $V_{fi} = \langle f, 1_k|d.E(\omega_0, r_0)|i, 0_k\rangle$. Within a photonic band gap, $\rho(\omega) = 0$ which, in turn, means that $W_{fi} = 0$. In other words, single photon spontaneous emission is completely inhibited within a photonic band gap. This implies that zero-point fluctuations, which are present even in vacuum, are absent for frequencies inside a PBG. Thus, within the forbidden frequency band, PBG materials are
emptier than even the vacuum.

In PBG materials $\rho(\omega)$ and $V_{ji}$ can be engineered to enhance spontaneous emission as well. The existence of PBG leads to other frequency regimes where $\rho(\omega)$ is larger than in free space. Recent experimental investigations of the spontaneous emission properties of organic dye molecules\[56, 57, 58, 59, 60, 61, 62\], semiconductor nanoparticles (quantum dots)\[63\] and rare-earth ions\[64\] embedded in PBG materials made of inverted opals have reported pronounced modification of spontaneous emission spectra and noticeable changes in decay kinetics.

### 2.3.2 The photon-atom bound state

The absence of propagating modes within a PBG means that, for frequencies within the band gap, there are no extended states expressible in Bloch form - as plane waves with purely real wave vectors and modified by functions invariant under translation through any lattice vector. Instead, the wave vector is pure imaginary, which causes the modes to decay exponentially in space. Now consider a single excited two-level atom embedded in a PBG material with a transition frequency $\omega_a$ to the ground state which lies within the band gap. If this atom drops to the ground state via single-photon spontaneous emission (that is by emitting a single photon of frequency $\omega_a$) the resulting photon state will be exponentially decaying away from the atom, since the frequency $\omega_a$ of the emitted photon lies within the classically forbidden energy gap of the PBG material. In other words, the spontaneously emitted photon will tunnel through the crystal for a short length, called the localization length, before being Bragg reflected back to the emitting atom to re-excite it. The result is a strongly coupled eigenstate of the electronic degrees of freedom of the atom and the electromagnetic modes of the dielectric. This is the photon-atom bound state first predicted by John and Wang\[5, 6\] and is the optical analog of an electronic impurity level bound state in the gap of a semiconductor\[13\]. When the atomic transition frequency is at mid-gap ($\omega_a = \omega_a$) the photon tunneling distance is on the scale of few optical wavelengths, for a gap to midgap ratio of $\Delta\omega/\omega_a = 5\%$. As $\omega_a$ approaches the band edge $\omega_e$, the photon
localization length $\xi_{\text{loc}}$ grows larger and eventually diverges near $\omega_c$: $\xi_{\text{loc}} \sim c/\sqrt{\omega_c|\omega_c - \omega_a|}$.

In free space, Lamb shift of atomic levels is dominated by the emission and re-absorption of high energy virtual photons. Within a photonic band gap, this self dressing is dominated by the real, bound photon. In general this will lead to some anomalous Lamb shift. If an atomic level lies near a photonic band edge, a more striking effect is predicted to occur[65, 66, 67]. In this case the atom is resonantly coupled to photons of vanishing group velocity. The resultant self-dressing of the atom by its own localized radiation field is sufficiently strong to split the atomic level into a doublet. One member of the doublet is pulled into the gap and retains a photon bound state, whereas the other member is pushed into the continuum and exhibits resonance fluorescence. In the nearly free photon approximation to the electromagnetic band structure, the splitting of a hydrogenic $2p_\frac{1}{2}$ level is predicted to be as large as $10^{-7} - 10^{-6}\hbar\omega_a[6]$. This is the analog of the much weaker vacuum Rabi splitting ($10 - 40$ MHz) well known for atoms in micro-cavities[43]. Unlike the well known Mollow level splittings[68] observed when an atom is externally dressed by an intense laser field, the splitting in the present case is solely due to the atom's own localized radiation field and occurs even in the absence of any external driving field. As a result of the interference between the doublets, spontaneous emission from the atom displays an oscillatory behavior which is quite distinct from a simple exponential decay as described by Fermi's golden rule[65, 66, 67]. Moreover, the photon-atom bound state leads to a novel fractionalized steady-state atomic population in the excited state[65, 66, 67]. Again, this is quite different from the free-space case where the steady-state population on an excited level is always zero, since all of the excited level population eventually decays to the ground level.

2.3.3 Dynamical suppression of spontaneous emission

Atomic level splitting may be effected not only passively through micro-cavities and PBG materials, but also actively by driving the relevant atomic transition by a resonant (or nearly resonant) laser field[69]. For a two-level atom consisting of the ground state $|0\rangle$ and an excited state $|1\rangle$, with transition frequency $\omega_a$ between them, the driving field splits
the excited level into two levels having energies close to $\omega_a \pm \Omega/2$, where $\Omega$ characterizes the strength of the driving field and is called the Rabi frequency of the driving field[70]. This phenomenon of level splitting induced by a driving field is known as dynamic Stark splitting or Autler-Townes splitting[71] and has been observed experimentally by several groups[72, 73]. The resulting two lines in the spontaneous emission spectrum of the atom are known as Autler-Townes doublets.

If a driven atom resides in a region of space in which the density of photon modes varies appreciably on a frequency scale set by the Rabi frequency of the driving field (such as near a photonic band edge), the interplay between the driving field and the threshold like behavior of the photon density of states may lead to dramatic modifications of the spontaneous emission from the atom. These modifications exhibit themselves through changes in the resonance fluorescence spectrum of the atom. The widths, heights, positions, and even shapes of the peaks in the spectrum become dependent on both the intensity of the driving field and the position of the atomic transition frequency relative to the band edge. This effect, brought about the combined actions of a driving field and a photon density of modes which changes on the scale of the Rabi frequency of the driving field, has been termed in the literature as “dynamical suppression of spontaneous emission”[74].

2.3.4 Defect modes within a photonic band gap

A PBG material is a periodic dielectric structure in which photonic states are classified into bands separated by band gaps. The presence of defects (local deviations from the ideal crystalline structure) in the crystal may drastically change the optical properties of the PBG material. A defect destroys the perfect 3D translational symmetry of the PBG material and may lead to the formation of a defect mode within the band gap region, analogous to donor and acceptor defect modes in a doped semiconductor crystal⁴. If a single defect

⁴A pure semiconductor crystal such as silicon has valence and conduction bands separated by a bandgap. However if this pure semiconductor crystal is doped with donor atoms (i.e. if some of the atoms in the crystal are replaced by donor atoms such as arsenic which contribute electrons to the conduction band), a donor level will appear within the forbidden band pushed down from the conduction band. Likewise, if the crystal is doped with acceptor atoms (atoms such as boron which accept electrons from the valence band and contribute holes), an acceptor level will be formed within the forbidden gap pushed up from the valence band[13]
mode is introduced into the photonic band gap, the density of states of the system will be zero within the band gap, except for isolated peaks associated with the defect mode [see Fig. 2.5].

Defects in a photonic crystal may be introduced by adding extra dielectric material where it does not belong or by removing some of the dielectric material that should be there. The first type of defect is called a "dielectric defect" and the second an "air defect"[10]. Removing a small amount of high-index material from one unit cell (air defect) leads to the occurrence of a localized defect mode just above the top of the lower band, analogous to acceptor modes in semiconductors. On the other hand adding a small amount of high-index material to a single unit cell (dielectric defect) causes a single localized state to split off from the upper band edge, analogous to donor modes in semiconductors[75]. This is a general result which applies for one-, two-, and three-dimensional photonic crystals. Whereas in one- and two-dimensional crystals even arbitrarily small defects can localize modes, in three-dimensional crystals the defect must be larger than some critical size before its localizing power begins. This is the electromagnetic analog of the quantum mechanical result that an arbitrarily weak attractive potential can bind a state in one and two dimensions, but not in three dimensions. Significant random perturbation of a photonic crystal (by adding or removing large amount of dielectric material, for instance) may wash the gap out and result in a pseudogap where the density of states is significantly reduced from that of free space but is not absolutely zero [see Fig. 2.5].

The frequency of a defect mode is an increasing function of the volume of an air defect and a decreasing function of the volume of a dielectric defect[10, 75, 76, 16]. In other words, for air defect, the larger the volume of material removed, the farther the defect mode is pushed from the lower photonic band edge into the gap (that is, the higher the frequency of the defect mode). Conversely, for a dielectric defect, the larger the volume of high-index material added, the further the defect mode is pushed from the upper photonic band edge into the band gap (that is, the lower the frequency of the defect mode). Thus the frequency of a defect mode can be “tuned” to any desired value within the gap by adding
Figure 2.5: Photon density of states in a PBG material with defects. When a single dielectric defect is created, a localized defect mode appears within the forbidden gap (a). Further disorder in the photonic crystal gives rise to a pseudogap (b), instead of a complete gap.

(or removing) the appropriate amount of dielectric material from a unit cell.

Powerful theoretical tools have been developed for modeling electromagnetic fields within photonic crystals[9, 77, 78]. Numerical simulations[10, 79] and experiments[75, 80] have confirmed the confinement of light to local defects in a PBG material. Local defects confine photons to volumes on the order of $(\lambda/2n)^3$, where $\lambda$ is the photon wavelength and $n$ is the refractive index of the material[31]. Thus, the high-index-contrast systems that are often necessary for achieving PBG’s result in strong photon confinement at local defects[10]. Such highly confined optical systems act as *microcavities* of very high quality factor$^5$ $Q$. They can be used to reduce the size and power requirements of integrated optical components, to generate single-mode operation of light-emitting devices, to reduce the lasing threshold of semiconductor lasers, and to allow higher modulation speed of these devices, as discussed in section (2.6).

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$^5$The quality factor $Q$ of a cavity is a measure of the optical energy stored in the cavity over the total cycle-averaged power radiated out of the cavity. It is defined as $\omega/\Delta\omega$, where $\omega$ is the peak frequency of the resonance and $\Delta\omega$ is the width of the resonance. The sharper the resonance line of the cavity, the smaller the decay rate and the larger the $Q$. 

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2.4 Photonic crystals in one-, two-, and three-dimensions

Photonic crystals are formed by wavelength-scale periodic patterning of dielectric materials in one, two, and three dimensions. The unique properties displayed by such crystals depend crucially on their dimensionality. In this section we discuss one-, two-, and three-dimensional photonic crystals in some detail[10].

2.4.1 One-dimensional photonic crystals

A one-dimensional (1-D) photonic crystal is nothing other than the well known dielectric Bragg mirror consisting of alternating layers with low and high indices of refraction, see Fig.2.6. The term "one dimensional" refers to the fact that the arrangement is periodic only in one direction (chosen here as the z-direction) and homogeneous in the xy-plane.

Photonic band gaps appear in the direction of periodicity, the z-direction. A mode with frequency in the gap region and propagating in the z-direction (on-axis propagation) will be totally reflected from the photonic crystal. For off-axis propagation, however, there are no band gaps, since the off-axis direction contains no periodic dielectric regions to coherently scatter light and split open a gap. Thus off-axis propagating modes are expected to be oscillatory, with real wave vectors.

In a 1-D photonic crystal, band gaps always appear for any dielectric contrast. In other
words, there is no threshold dielectric contrast for the appearance of a PBG. The smaller the contrast, the smaller the widths of the gaps, but the gaps open up as soon as \( n_1/n_2 \neq 1 \), where \( n_1 \) and \( n_2 \) are the refractive indices of the dielectric materials.

A defect can be introduced in a 1-D photonic crystal, for example, by making one of the layers have a slightly different width than the rest. The defect mode is then localized in the \( z \)-direction but is extended in the \( x \)- and \( y \)-directions.

2.4.2 Two-dimensional photonic crystals

A two-dimensional (2-D) photonic crystal is periodic along two of its axes and homogeneous along the third. A typical specimen, consisting of an array of dielectric columns arranged on a square lattice, is shown in Fig. 2.7. This structure is periodic along the \( x \)- and \( y \)-axes and homogeneous along the \( z \)-axis. Photonic band gaps appear in the plane of periodicity (the \( xy \)-plane). Thus, unlike the multilayer film which reflects light only at normal incidence, a 2-D photonic crystal can reflect light incident from any direction in the plane of periodicity. Since the structure is homogeneous in the \( z \)-direction, modes traveling in that direction do not see a photonic band gap.

In 2-D, unlike in 1-D, there is a threshold dielectric contrast necessary for the appearance of a photonic band gap. In other words, in a 2-D structure, a photonic band gap does not open up just because the structure has a refractive index contrast, but rather requires
special design considerations, as discussed below.

A 2-D photonic band gap is defined as a gap for all EM waves propagating perpendicular to the z-axis of either polarization. A complete band gap, on the other hand, occurs only for 3-D structures. Owing to the vector nature of the electromagnetic field, photonic band structures for transverse-electric (TE) and transverse-magnetic (TM) modes can be completely different. To have a 2-D band gap for all polarizations, a photonic crystal should not only have TM and TE band gaps, but these band gaps should also overlap. TM band gaps are favored in a lattice of isolated high-index regions, as in an array of dielectric columns in air [see Fig. 2.7]. On the other hand, TE band gaps are favored in the inverse structure, as in an array of air columns (veins) drilled in a dielectric substrate[10]. A structure with dielectric veins is said to be a connected structure in that the high-index regions form a continuous path instead of discrete spots. Thus, to design a photonic crystal that has band gaps for both TM and TE polarizations, one has to somehow reconcile these seemingly contradictory conditions. A triangular lattice of low-index columns (air columns, for instance) inside a high-index medium just does that. Such a structure is shown in Fig. 2.8. If the radius of the columns is large enough, the spots between columns look like localized regions of high-index material, which are connected (through a narrow squeeze between columns) to adjacent spots [see Fig. 2.8(b)].

In a 2-D photonic crystal, made by perforating a high-index slab with a triangular or hexagonal array of air holes, a defect can be formed by removing an air hole and/or adjusting the diameters of a few neighboring air holes. A mode (or a set of modes depending on the defect geometry), which is highly localized to the defect region in the \(xy\)-plane, but extended in the \(z\)-direction may be formed. Photons can escape from the defect cavity by tunneling through the 2-D photonic crystal, or by leaking out in the \(z\)-direction. Removing rows of air holes is one way of creating line defects in a 2-D photonic crystal. Such line defects can serve as waveguides able to transmit light around sharp corners with very high efficiency.
2.4.3 Three-dimensional photonic crystals

A three-dimensional (3-D) photonic crystal is a dielectric structure that is periodic along three different axes. Provided that the conditions of sufficiently high dielectric contrast, suitable periodicity, dielectric filling ratio and network connectivity are met, a photonic band gap appears in all directions. Such a 3-D PBG material, unlike the 1-D and 2-D ones, can reflect light incident from any direction. In other words, a 3-D PBG material behaves as an omnidirectional high reflector.

Even though there are infinitely many possible geometries for a 3-D photonic crystal, to date the best structure found to support a full 3-D photonic band gap is the diamond lattice[9, 81].\(^6\) For a diamond lattice, a complete photonic band gap exists whether one embeds dielectric spheres in air or air spheres in a dielectric medium, as long as the radius is chosen appropriately. The calculated band structure of a diamond lattice of air spheres in silicon ($\varepsilon = 11.9$) substrate is shown in Fig. 2.9. Between the second and the third bands resides a band gap with a gap-midgap ratio of 27.28%, centered at $\omega_o = (2\pi c/a)(0.589)$ where $a$ is the lattice constant. The filling ratio of the air spheres is 81% indicating that the

\(^6\)Incidentally, the common semiconductors, silicon and germanium, also have diamond symmetry[13]
Figure 2.9: The photonic band structure for the lowest ten bands of a diamond lattice of overlapping air spheres in a high dielectric ($\epsilon = 11.9$) material. The filling ratio of the air spheres is 81%. The frequency is given in units of $c/a$ where $a$ is the lattice constant and $c$ is the speed of light. A full photonic band gap centered at $\omega_0 = 0.589(2\pi c/a)$ with a gap-midgap ratio of $r = \Delta\omega/\omega_0 = 27.26\%$ appears between the second and third bands.

Air spheres are overlapping (the structure is highly porous). Both the air and the dielectric regions are connected, in the sense that there are no isolated spots of either.

A systematic examination of the photonic band structures for dielectric sphere and air spheres on a diamond lattice, as a function of refractive index ratio and filling ratio, was made by Ho et al.[9]. In all cases examined the lattice constant $a$ was kept fixed and the radius $r$ of the spheres was varied to change the filling fraction $f$. They found that when the refractive index is fixed at 3.6,$^7$ complete band gaps exist over a wide range of filling ratios for both dielectric spheres and air spheres. The calculated size of the band gap normalized to the midgap frequency is plotted in Fig. 2.10(a) for both cases. For dielectric spheres on a diamond lattice, a maximum gap-midgap ratio of 15.7$\%$ was found at $f = 37\%$, whereas for the case of air spheres, the ratio can reach 28.8$\%$ at $f = 81\%$. In Fig. 2.10(b) the gap-midgap ratio $\Delta\omega/\omega_0$ is plotted as a function of refractive index for a fixed dielectric structure, with $f = 34\%$ for the case of dielectric spheres in air, and $f = 81\%$ for air spheres in a dielectric background. For both cases a complete photonic band gap exists when the

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7Crystalline silicon and other semiconductors are excellent infrared optical materials, providing refractive indices $\sim 3.5$. 

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refractive index contrast exceeds 2. This is a very important result, because in the optical region there are many transparent materials with refractive index above 2. For increasing contrasts the gap-midgap ratio saturates to a value of 21% for the case of solid spheres and to a very large value of 46% for the case of air spheres[9].

In a 3-D PBG material, just as in one- and two-dimensional ones, perturbing a single lattice site may cause the appearance of a single peak in the photon density of states at a frequency that may lie inside the band gap. The width of this peak tends to zero as the crystal size tends to infinity. Since no extended states are allowed in the crystal within the band gap, the mode in the band gap must decay exponentially away from the defect. But in this case the decay occurs in all three dimensions. Thus, in a 3-D PBG material, the defect mode is localized at a single point in the crystal. By contrast, one (two) dimensional PBG materials can only localize light on a plane (line).

2.5 Fabrication of PBG materials

There have been two main challenges in the field of PBG materials. The first was to show that a full 3-D PBG could actually exist in some type of dielectric structure. The second was to show that such a a PBG material could be created in a micro-structure amenable to practical micro-fabrication. The theoretical calculations of Ho, Chan and Soukoulis predicted that periodic dielectric materials with a diamond[9] or diamond like[82] symmetry would have a full 3-D PBG. The gap is centered at roughly twice the index modulation wavelength. Thus, for microwave control, a photonic crystal should be constructed with millimeter dimensions, for infra-red control with micron dimensions, and for optical control with submicron dimensions. Based on the Ho, Chan, Soukoulis calculations, the first experimental PBG material was fabricated by Yablonovitch and co-workers, with a band gap in the microwave region[81]. This structure, sometimes known as the Yablonovite, was made by mechanically drilling cylindrical holes through a low loss dielectric block ( of refractive index $n \sim 3.6$) so as to create a structure with diamond symmetry.

While demonstrating the existence of a photonic gap, the fabrication of the Yablonovite
Figure 2.10: (a) Gap-midgap frequency ratio ($\Delta \omega / \omega_0$) as a function of filling ratio for the case of dielectric spheres in air and air spheres in dielectric. The refractive index of the material is chosen to be 3.6. (b) $\Delta \omega / \omega_0$ as a function of refractive index contrast for a fixed dielectric structure. The dotted line is for the case of air spheres in dielectric with a filling ratio of 81%, and the solid line is for dielectric spheres in air with a filling ratio of 34%.
is a very sophisticated process that cannot be easily reproduced or extended to optical wavelengths. Several different geometries have been suggested for the fabrication of 3-D PBG materials[11, 83, 84], and 3-D PBG's have been developed in the near infra-red[85, 86, 87, 88, 89]. However, the micro-fabrication of large scale photonic crystals with full 3-D band gaps at infra-red and optical frequencies is a major challenge. To achieve band gaps for the infra-red and visible spectrum, the periodicity of the crystal should be on the scale of the wavelength of light (about 500 nm), both constituent materials of the crystal should be topologically interconnected[90], and the ratio of their refractive indices should be close to 3.0[12]. The submicron size periodic lattices required for optical frequency PBG materials limits the use of micro-lithographic fabrication techniques[87, 91, 92]. Self organizing systems such as colloidal crystals[93, 94, 95], and artificial opals[96, 97, 98] provide a template for fabricating these structures.

Colloidal particles have been synthesized from a variety of materials, such as latex and $SiO_2$, as monodispersed spheres having precisely controlled diameters (less than 5% variation in sphere diameter) in the range of a few nanometers to a few micrometers[93, 95]. A suspension of such colloidal microspheres, with a typical concentration of $10^{10}$ particles/cm$^3$, can sediment under gravity into a cubic-close-packed structure with relatively large domain size ($\sim 1 \text{ cm}^2$)[99]. These 3-D lattices have a crystalline structure similar to that of a natural opal$^8$. These artificial opals are solid but present a low mechanical stability which can be greatly improved by a sintering process[100]. Sintering or thermal treatment of these artificial opals at elevated temperatures reduces the inter-particle pore volume by changing point contacts between spheres into faceted ones, leading to the formation of a mechanically robust crystal. Thus, colloidal crystal growth produces inherently 3-D structures, a significant advantage over lithographic techniques which primarily produce 2-D patterns. However, neither colloids nor opals achieve the high refractive index ratios necessary for photonic band gap formation. These requirements are attained by using the colloidal crystals as templates to fabricate inverse opals which are close-packed lattices of air balls in a

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$^8$A natural opal is close-packed fcc lattice of $SiO_2$ spheres ($\varepsilon = 2.1$) with a filling fraction of $\sim 74\%$. 

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Figure 2.11: Computer representation of an inverse opal structure. Note the circular “windows” interconnecting the air balls. (Courtesy of Ovidiu Toader, Dept. of Physics, University of Toronto).

dielectric matrix.

Inverse opals are fabricated in a three-step process. First, a colloidal crystal of latex or silica spheres is self-assembled to generate a high quality fcc lattice template. Next, the *interstitial regions* (the void spaces among the colloidal particles of an opalline lattice) are infiltrated with high refractive index material[101, 102, 103, 104, 105, 106]. The third step involves the removal of the template material by heat or chemical treatment, immersing the sample in an appropriate chemical etching solution being one example. The end result is a membrane consisting of highly ordered 3-D arrays of air balls interconnected by circular “windows” as shown by a computer simulation[107] displayed in Fig. 2.11.

A highly porous inverse opal, with a filling ratio of 20% – 30% of the high dielectric material, is needed for optimum photonic effects[12, 84, 107]. Theoretically, it has been demonstrated[12] that inverted opal structures exhibit near visible photonic band gaps on the scale of 10% of the midgap frequency. Such structures have been experimentally realized
with TiO$_2$ (refractive index $n = 2.8$) and CdSe[101, 105, 106, 108]. However, both of these structures are below the required refractive index threshold for PBG formation.

An important application of PBG materials arises for the band gap around 1.5 $\mu$m - the wavelength most commonly used in fiber optical communication. Such a PBG structure requires colloidal spheres with a diameter of $\sim 870$nm. In this connection, an important step has been achieved[109] through the fabrication of large scale silicon inverse opals, with a complete three-dimensional photonic band gap centered near 1.5 $\mu$m.

2.6 Applications of PBG materials

The ability to control spontaneous emission using PBG materials will have profound consequences for optoelectronics devices. It can be used to dramatically enhance the light extraction efficiency (the ratio of the flux emitted into specified modes to the total emitted flux) of light emitting diodes (LEDs)[110, 111, 112, 113], which, in turn, has the effect of reducing the absorption loss and increasing the modulation (response) speed of LEDs. It also has potential applications in photocatalysis - a very large field which impacts various areas of chemical synthesis and applications in biochemistry, fuels development, and solar energy conversion[114]. PBG materials made out of a semiconducting material can be used to control the radiative recombination of electrons and holes in the semiconductor[115]. In a semiconductor laser, this would lead to a near unity quantum efficiency into the lasing mode.

A PBG material can be used as a perfect dielectric mirror which reflects light, since light incident on a PBG material with a frequency in the gap region is backscattered from the material, independent of the angle of incidence. This property lends PBG materials to numerous applications, reflectivity being the heart of so many devices such as lasers. For instance, a PBG material might make a very good, narrow band filter, by reflecting all (and only) frequencies in the gap. Photonic crystals, fabricated so as to have a photonic band gap for only one type of polarization, can be used as polarization filters or polarizers which reflect back one type of polarization while allowing the other independent polarization to
pass through. In these applications dielectric structures have an advantage over metallic mirrors which rely on the high (frequency dependent) conductivity of metals and suffer dissipative losses at higher frequencies.

The reflective property of PBG materials can also be used to advantage in the design of planar antennas[116, 117, 118] which, in integrated circuits, play the important role of radiating signals off the chip into free space[119]. By using a 3D PBG as an antenna substrate, it is possible to ensure that all of the antenna power is radiated into air rather than into the substrate, provided that the driving frequency of the antenna lies within the band gap.

The localized mode associated with a defect in an otherwise perfect PBG material can act as a microcavity of very high quality factor $Q$[see section (2.3.4)]. This high $Q$ defect mode microcavity can be considered for all the applications in which high-$Q$ optical microcavities are used. It can be used to realize the optical engineer’s dream of threshold-less lasers[120, 121]. It may also be used in experiments to illustrate the quantum properties of light, and atom-photon interactions, such as the Jaynes-Cummings model[122] which describes the interaction of two level atoms with a single quantized mode of the radiation field.

Just as point defects in a photonic crystal are used to trap light, extended defects (such as a line of point defects) can be used to guide light from one location to another. If the frequency of the guided mode lies within the photonic band gap, the mode is forbidden to escape into the crystal, regardless of the shape of the waveguide, because the confinement mechanism does not have angular dependence. Thus, a waveguide cut out of a PBG material is capable of guiding light around sharp bends with little or no leakage, even when the radius of curvature of the bend is less than the wavelength of light[123, 124, 125, 126]. Moreover, since the light is guided in a hollow waveguide surrounded by omnidirectionally reflecting PBG material, the propagation is primarily through air and will therefore experience substantially lower absorption losses[125]. These properties of a PBG waveguide are in sharp contrast to those of conventional waveguides such as optical fibers. Conventional waveguides are based on the principle of total internal reflection which confines light only
of a limited angle. If a waveguide takes a tight curve, the angle of incidence will be too large for total internal reflection to occur so that the light escapes at the corners and is lost. Moreover, in conventional waveguides, light is guided through a dense medium, and, therefore, for long distances, material absorption becomes significant even in low loss materials. To compensate for losses the fiber is doped with erbium which is used to amplify the signal. This, in turn, limits the bandwidth of the fiber to that of the narrow band erbium excitation lines\[127\].

The polarization characteristics of photonic bands in photonic crystals, even those without a photonic band gap, mean that these crystals can possess large birefringence\(^9\) in the long-wavelength limit. The birefringence of two-dimensional photonic crystals (composed of a triangular lattice of air cylinders in silicon) was recently investigated both theoretically and experimentally by H. M. van Driel and co-workers\[129\]. The measured birefringence (for electric field polarized parallel and perpendicular to the cylinder axis) reaches a maximum value of 0.366 near the first photonic band edge at \(\lambda \approx 6.52\ \mu m\). By contrast, at wavelength 589.3 nm the birefringence of quartz (crystalline \(SiO_2\)) is 0.0091 whereas that of calcite (\(CaCo_3\)) is 0.172\[130\]. Photonic crystal birefringence could be used in a wide variety of photonic devices, including wave-plates, polarization rotators, optical isolators and beam splitters.

2-D PBG materials, in contrast to 3-D microstructures, are much more amenable to controlled fabrication owing to mature nanofabrication technology. As a consequence, 2-D photonic crystals have been more thoroughly investigated than their 3-D counterparts\[131, 132, 133, 134\]. Although they do not provide 3-D light guiding or confinement, 2-D structures could already bring a sizeable part of the advantages expected from 3-D structures. The enhancement and suppression of spontaneous emission in thin-film 2-D photonic crystals at room temperature have been investigated\[110\]. The optical and confinement properties of 2-D photonic crystals have been studied\[135, 136, 137\]. Triangular and hexagonal 2-D defect mode PBG microcavities showing a \(Q\)-factor of at least 900 have already been

\(^9\)Birefringence or double refraction is defined as the difference in the effective refractive indices seen by the electric fields associated with two orthogonal polarizations.
demonstrated[138, 139]. One can think of such cavity modes as filters that select only the resonant frequency[140]. The use of two-dimensional PBG materials to localize light to a single defect and thereby form a high-Q nanocavity laser with a modal volume of less than 0.03 cubic microns, constituting the smallest laser ever made, is reported[80, 141]. The performance and guiding properties of waveguides fabricated in 2-D PBG materials are investigated[126, 142].

The potential applications of PBG materials for passive devices such as filters, waveguides, antenna substrates and reflectors have already been discussed. The possibility of dynamically controlling the spectral and spatial properties of PBG materials by active elements is expected to open new application prospects[143]. The latest development in this direction involves the use of liquid crystals as active elements to tune PBG materials made out of inverted opals. This novel concept of tunability was first proposed by Busch and John[144, 145] who studied the effects of partially infiltrating (coating) the spherical voids of an inverted opal made out of silicon with a low index nematic liquid crystal[146]. They showed that the width of a band gap can be adjusted or the band gap can be eliminated altogether by an applied electric field which changes the orientation of the nematic director. In this manner, the PBG can be either globally altered or locally addressed by applied voltages. The time scale of electro-optic modulation of the PBG may be on the scale of microseconds to milliseconds depending on the response time of the nematic.

The recent spectacular technological advances in the synthesis of 3-D PBG materials at infra-red and optical frequencies have made it possible to envision a PBG material as a platform for integrating an all-optical circuitry. An array of densely packed photonic crystal microcavities[138], waveguides[126], prisms[128], switches[147] and light sources[80, 141] can be integrated on a very small area of a photonic crystal by engineering the appropriate defects inside the crystal, paving the way for all optical computing.
Chapter 3

Coherent Control of Spontaneous Emission Near a Photonic Band Edge: The Leading Approximation

Recently quantum interference and coherence in a multilevel atomic system has attracted a lot of attention, because it leads to such interesting effects as the enhancement of the index of refraction with greatly reduced absorption, electromagnetically induced transparency, and optical amplification without population inversion[14]. The coherent control of molecular chemical reactions[148] is an emerging frontier in chemical physics[149]. Using the coherence properties of an external laser field driven interaction, radiatively controlled chemical pathways can be enhanced or retarded by quantum mechanical interference effects. Selective photo-dissociation of molecules mediated by the interference between two two-photon excitation processes has been reported[150]. Coherent control of current in a semiconductor has also been demonstrated[151]. Here, the direction of the electrical current, formed by inter band transitions in a bulk semiconductor via coherent one- and two-photon absorption, is controlled by simply adjusting the relative phase of the two beams that are optically
generating carriers across the gap. In view of these achievements, it is of great interest to consider the combined effects of coherent control by means of external laser fields and the coherent localization effects facilitated by a photonic band gap.

In this and the next chapter we investigate the combined effects of coherent control and photon localization on spontaneous emission for a three-level atom located within a perfect photonic band-gap structure, with one resonant frequency near the edge of the photonic band gap. The initial state of the three-level atom is prepared by an ultra-short pump laser field, and the two upper levels of the atom are driven by a continuous wave control laser. The model is considered in what we call the leading approximation whereby a number of spontaneous emission effects and non-radiative interactions are neglected. This renders the problem amenable to analytical solution, the results of which are used to investigate the populations and coherences of the upper levels of the three-level atom. The corrections to this picture are considered in chap(4). Section (3.1) deals with a description of the model system which includes writing the appropriate Hamiltonian, and deriving the equations of motion for the relevant variables. In section (3.2) we consider the case when our model system is in free space. This case, besides being an interesting case in its own right, will be useful to compare and interpret the results of the PBG case which is discussed in section(3.3). Considerations beyond the leading approximation are deferred to chapter (4).

### 3.1 Model system

#### 3.1.1 Description of the Model System

The physical system we consider consists of a single three-level atom placed inside a PBG material which is then driven by a laser field, see Fig. (3.1). We let \(|0\rangle\) denote the ground level of the atom; and \(|1\rangle\) and \(|2\rangle\) the two excited levels with orthonormality conditions \(\langle i|j\rangle = \delta_{ij}\), where \(\delta_{ij}\) is the Kronecker delta function. We designate the energy of an atomic level \(|i\rangle\) by \(\hbar \omega_i\) and the frequency separation between levels \(|i\rangle\) and \(|j\rangle\) by \(\omega_{ij} = \omega_i - \omega_j\). The transition between levels can be described using the atomic operators \(\sigma_{ij} = |i\rangle\langle j|\), with
the property $\sigma_{ij}|k\rangle = \delta_{jk}|i\rangle$, from which the commutation relation

$$[\sigma_{ij}, \sigma_{ik}] = \delta_{jl}\sigma_{ik} - \delta_{ik}\sigma_{lj}$$

(3.1)

can readily be obtained.

The upper atomic level $|2\rangle$ is dipole coupled to the ground level $|0\rangle$ by radiation modes (photon reservoir) in a three dimensional periodic dielectric structure. The transition frequency $\omega_{20}$ is assumed to be near the edge of the gap in the density of the reservoir photon modes. Each mode of the photon reservoir is characterized by a wave vector $k$ and a polarization index $\lambda(= 1, 2)$ and can be treated as a quantum oscillator with frequency $\omega_k$. Transitions between photon occupation number states $|n_{k\lambda}\rangle$ are described by the radiation field annihilation ($a_{k\lambda}$) and creation ($a_{k\lambda}^\dagger$) operators satisfying the standard Bose algebra

$$[a_{k\lambda}, a_{k'\lambda'}^\dagger] = \delta_{kk'}\delta_{\lambda\lambda'}.$$  

(3.2)

We assume that atomic operators $\sigma_{ij}$ commute with the field operators $a_{k\lambda}$ and $a_{k\lambda}^\dagger$. The transition $|2\rangle \rightarrow |1\rangle$ between the two upper levels is driven by a resonant control laser field.

In this chapter we assume that spontaneous emission on the transitions $|2\rangle \rightarrow |1\rangle$ and $|1\rangle \rightarrow |0\rangle$ is inhibited either by symmetry considerations or by the presence of the photonic band gap. This assumption constitutes the leading approximation to our model system. This approximation describes the essential physics contained in the model system. In chapter (4), we consider corrections to this leading approximation brought about by the inclusion of the spontaneous emission channels $|2\rangle \rightarrow |1\rangle$ and $|1\rangle \rightarrow |0\rangle$, and other non-radiative interactions.

First we consider a three-level atom in the so called V configuration[Fig. (3.1a)]. In such a configuration, the upper levels $|2\rangle$ and $|1\rangle$ are of the same symmetry so that single photon spontaneous emission $|2\rangle \rightarrow |1\rangle$ is not dipole allowed. Now if we assume that the transition frequency $\omega_{10}$ is deep inside the gap, then single photon spontaneous emission for the transition $|1\rangle \rightarrow |0\rangle$ will lead to a photon-atom bound state[65, 152]. Thus, for
Figure 3.1: Schematic representations of a driven three level system (a) in the V configuration and (b) in the \( \Lambda \) configuration. The transition frequency \( \omega_{20} \) is near the band edge frequency \( \omega_c \) of a PBG. Lines with arrows at both ends denote the control laser field of Rabi frequency \( \Omega \) driving the transition \( |2\rangle \leftrightarrow |1\rangle \). Double arrowed lines denote two-photon transitions. Dashed lines denote dipole allowed transitions. In the V configuration levels \( |2\rangle \) and \( |1\rangle \) are of the same symmetry and \( \omega_{10} \) is deep inside the PBG so that there are no single photon spontaneous emissions on the transitions \( |2\rangle \rightarrow |1\rangle \) and \( |1\rangle \rightarrow |0\rangle \). Similarly in the \( \Lambda \) configuration levels \( |1\rangle \) and \( |0\rangle \) are of the same symmetry and \( \omega_{21} \) is deep inside the PBG so that there are no single photon spontaneous emissions on the transitions \( |2\rangle \rightarrow |1\rangle \) and \( |1\rangle \rightarrow |0\rangle \). The control laser field drives a two-photon transition \( (2\omega_L = \omega_{21}) \) in the V configuration and a single-photon transition \( (\omega_L = \omega_{21}) \) in the \( \Lambda \) configuration. Fig.(1c) shows indirect coupling of levels \( |1\rangle \) and \( |2\rangle \) in a \( V \) system via another level \( |3\rangle \). Such a scheme will allow us to strongly couple levels \( |1\rangle \) and \( |2\rangle \) even when the transition \( \omega_{21} \) lies in the infra-red or far infra-red.
a three-level atom in the V configuration, the leading approximation is satisfied if \( \omega_{10} \) lies deep inside the gap. In a V system, the external control laser field of frequency \( \omega_L \) which couples levels \(|2\rangle\) and \(|1\rangle\) drives a two-photon transition \((2\omega_L = \omega_{21})\), since the levels are of the same symmetry. From a practical point of view, we want the transition frequency \( \omega_{21} \) to be as large as possible, as it may be difficult to generate microwave (MW) fields of sufficient amplitude to drive the required two-photon transition. However, the magnitude of \( \omega_{21} \) is restricted by the width of the photonic band gap. For a gap centered at frequency \( \omega_o \) and with a gap to mid-gap ratio of \( r \equiv \Delta \omega/\omega_o \), conditions that \( \omega_{20} \) be near the edge of the gap and that \( \omega_{10} \) be deep inside the gap require that \( \omega_{21} = \omega_{20} - \omega_{10} < r\omega_o \). Thus, to make \( \omega_{21} \) large we need a gap with as high a central frequency as possible and as large a width as possible. For a gap centered at an optical frequency \( \omega_o \sim 10^{16} Hz \) and with a gap to mid-gap ratio of 10\%, the frequency separation \( \omega_{21} \) between levels \(|2\rangle\) and \(|1\rangle\) must be approximately \( 5 \times 10^{14} Hz \).

Another means of overcoming the above practical limitation associated with the V system is to couple levels \(|1\rangle\) and \(|2\rangle\) indirectly by way of a transition to a higher level \(|3\rangle\) which lies far above level \(|2\rangle\). This will allow us to both strongly couple levels \(|1\rangle\) and \(|2\rangle\) and use a narrow band gap, even when the transition frequency \( \omega_{21} \) lies in the near or far infra-red. Level \(|3\rangle\) is dipole coupled to level \(|1\rangle\) (and hence to level \(|2\rangle\), since they are of the same symmetry) and the transitions \( \omega_{31} \) and \( \omega_{32} \) are both in the visible and both lie outside the gap, as shown in Fig. (3.1c). The transition \( \omega_{31} \) is then pumped by a resonant laser \( \omega_p = \omega_{31} \) followed by a stimulated emission into level \(|2\rangle\) using a laser which couples levels \(|3\rangle\) and \(|2\rangle\)[153].

Next we consider a three-level system in the \( \Lambda \) configuration [Fig. (3.1b)]. In such a configuration, levels \(|1\rangle\) and \(|0\rangle\) have the same symmetry and there is no dipole-allowed single photon spontaneous emission between these levels. Now, if we further assume that the transition frequency \( \omega_{21} \) is far inside the gap, the dipole allowed transition \(|2\rangle \rightarrow |1\rangle\) will create a photon-atom bound state whose radiative lifetime is given by the two photon spontaneous emission time for the \(|1\rangle \rightarrow |0\rangle\) transition. Thus, for a three-level atom in the
A configuration, the leading approximation is satisfied if $\omega_{21}$ lies deep inside the gap. To reconcile the conditions that $\omega_{20}$ is near the band-edge and that $\omega_{21}$ is deep in the gap, we require that $\omega_{10} = \omega_{20} - \omega_{21} \leq r\omega_0$. Given the practical fact that $r \leq 0.1$, it follows that levels $|1\rangle$ and $|0\rangle$ should be close to each other but both far from level $|2\rangle$, as shown in Fig. (3.1b). This, in turn, will reduce the decay rate of the photon-atom bound state due to two-photon spontaneous emission from $|1\rangle \rightarrow |0\rangle$. However, since $\omega_{21}$ is within the gap, the control laser driving the single-photon transition $|2\rangle \rightarrow |1\rangle$ must be injected by means of engineered or naturally occurring defect or waveguide modes within the band gap material.

Our model system may be realized by trapping cold atoms in the void regions of a photonic crystal, using the properties of the electromagnetic eigenmodes of a 3-D PBG material. If the PBG material is illuminated by an intense laser field with frequency near the bottom of the "air" band, a nearly standing wave electromagnetic field will arise with strong electric field gradients and peak intensities that lie in the void fraction of the material. This field distribution will act as an optical trapping potential for a cold atom vapor[107]. This will trap atoms in the void regions of the photonic crystal where the field is most intense and prevent the atoms from colliding with the dielectric backbone of the PBG material. In a typical 3-D PBG material, the void fraction forms a connected network that accounts for nearly 75% of the volume of the material. Atoms which are optically trapped in this extensive void network will be immune to collisional dephasing and decoherence phenomena arising from direct interaction with atoms in the solid dielectric backbone. Another way of realizing our model system is by doping the solid fraction of the PBG material with an impurity three level atom such as a rare-earth atom or by means of a quantum dot (artificial atom) incorporated inside the semiconducting back-bone of the photonic crystal with the required electronic transitions near the photonic band edge. However, in either of these latter cases, decoherence effects arising from the interaction of the three-level system with phonons in the dielectric fraction of the PBG material need to be carefully considered.
3.1.2 Model Hamiltonian and Equations of Motion

The Hamiltonian describing the leading approximation to our model system can be written as [see Appendix (A)]

\[ H = H_A + H_R + H_{AR} + H_{AL} \]

(3.3)

where

\[ H_A = \sum_{i=0}^{2} \hbar \omega_i \sigma_{ii} \]

(3.4)

\[ H_R = \sum_{\lambda=1}^{2} \sum_{k} \hbar \omega_k a_{k\lambda}^\dagger a_{k\lambda} \]

(3.5)

\[ H_{AR} = i\hbar \sum_{\lambda=1}^{2} \sum_{k} g_{k\lambda} (a_{k\lambda}^\dagger \sigma_{02} - \sigma_{20} a_{k\lambda}) \]

(3.6)

Here \( H_A \) represents the Hamiltonian of the bare atom whereas \( H_R \) stands for the Hamiltonian of the photon reservoir (neglecting the zero-point energy). The Hamiltonian \( H_{AR} \) describes interaction between the atomic transition \( |2\rangle \rightarrow |0\rangle \) and the photon reservoir. Here \( g_{k\lambda} \) is the frequency dependent coupling constant (assumed to be real) between the atomic transition \( |2\rangle \rightarrow |0\rangle \) and the mode \( \{k\lambda\} \) of the radiation field:

\[ g_{k\lambda} = \frac{\omega_{20} d_{20}}{\hbar} \left( \frac{\hbar}{2\epsilon_o \omega_k V} \right)^{1/2} \hat{e}_{k\lambda} \cdot \hat{d}_{20} \]

(3.7)

In this expression \( d_{20} \) and \( \hat{d}_{20} \) are the magnitude and unit vector of the atomic dipole moment \( d_{20} \) for the transition \( |2\rangle \rightarrow |0\rangle \), \( V \) is the sample volume, \( \hat{e}_{k\lambda} \) are the two transverse (polarization) unit vectors and \( \epsilon_o \) is the Coulomb constant. The coupling constant \( g_{k\lambda} \) fully characterizes the density of modes in the photon reservoir.

The interaction Hamiltonian \( H_{AR} \) is written in the electric dipole approximation[70]. It is also written in the rotating wave approximation[70] in which virtual processes of excitation (de-excitation) of the atom with simultaneous creation (annihilation) of a photon [that is, terms of the form \( a_{k\lambda}^\dagger \sigma_{20} \) and \( a_{k\lambda} \sigma_{02} \)] are neglected.

In Eq. (3.3), the Hamiltonian \( H_{AL} \) represents the interaction between the atom and the
coherent monochromatic laser field driving the transition $|2\rangle \leftrightarrow |1\rangle$. We assume that the driving field is sufficiently strong that it can be treated classically. In the $\Lambda$ configuration, levels $|2\rangle$ and $|1\rangle$ are of opposite symmetry, and, therefore, the transition $|2\rangle \rightarrow |1\rangle$ is a dipole allowed transition. Thus, the external control laser field which couples levels $|2\rangle$ and $|1\rangle$ drives a one-photon transition ($\omega_L = \omega_{21}$). In this case $H_{AL}$ can be written as[154]

$$H_{AL}^\Lambda = i\hbar \Omega_\Lambda [e^{i(\omega_L t + \phi)} \sigma_{12} - e^{-i(\omega_L t + \phi)} \sigma_{21}],$$

(3.8)

where $\omega_L$, $\Omega_\Lambda$, and $\phi$ represent, the angular frequency, the Rabi frequency, and the phase of the driving laser.

The Rabi frequency[70] is a measure of the strength of the interaction between the driving field and the atomic transition $|2\rangle \rightarrow |1\rangle$. For a dipole-allowed transition, such as $|2\rangle \rightarrow |1\rangle$ in the $\Lambda$ system, the Rabi frequency of the driving field is proportional to the product of the atomic dipole moment $d_{21}$ and the amplitude $E_0 = |E_0|$ of the sinusoidal optical field:

$$\hbar \Omega_\Lambda = d_{21} E_0.$$  

(3.9)

We can relate $\Omega_\Lambda$ to the intensity $I$ of the driving field as follows. The rate of flow of electromagnetic energy per unit area is given by the Poynting vector $S = E \times H$, where $E$ and $H$ are the electric and magnetic fields, respectively. The intensity $I$ of an electromagnetic wave is the time average of $S$ over the detector response time, $I = \langle |S| \rangle$. For a plane wave ($E = E_0 e^{i(kx-\omega t + \phi)}$), the amplitude $E_0 = |E_0|$ is related to the intensity $I$ by

$$I[W/m^2] \approx 1.33 \times 10^{-3} |E_0[V/m]|^2,$$

(3.10)

From Eqs. (3.9) and (3.10) we obtain

$$\Omega_\Lambda [Hz] \approx 2.2 \times 10^6 \frac{d_{21}}{ea_0} \sqrt{I[W/m^2]},$$

(3.11)

where $e = 1.6 \times 10^{-19} C$ is the electronic charge and $a_0 = 5.29 \times 10^{-11} m$ the Bohr radius, the
radius of the smallest Bohr orbit in hydrogen. For an optical transition \( \omega_{21} \approx 10^{15} \text{Hz} \), the dipole moment \( d_{21} \approx e a_0 \) which reduces Eq. (3.11) to \( \Omega_\Lambda \approx 2.2 \times 10^6 \sqrt{I} \). Thus, when \( \omega_{21} \) is in the optical regime, a laser power of \( I \approx 10^{17} \text{W/m}^2 \) (with a corresponding field amplitude of \( E_0 \approx 10^{10} \text{V/m} \)) is required to make \( \Omega_\Lambda \) comparable to \( \omega_{21} \). This would cause dielectric breakdown and is many orders of magnitude larger than that used in any resonance experiment[70]. Thus, when \( \omega_{j0} \) is in the optical regime, we can safely assume that

\[
\Omega_\Lambda \ll \omega_{j0}. \tag{3.12}
\]

The optical field will complete many cycles during one Rabi cycle. For example, for a 100 mW continuous wave laser focused on a 100 \( \mu \text{m} \) by 100 \( \mu \text{m} \) sample so that \( I \approx 10^7 \text{W/m}^2 \), we have \( E_0 \approx 10^5 \text{V/m} \) which corresponds to \( \Omega_\Lambda \approx 10^{10} \text{Hz} \), at optical transitions.

On the other hand, in the \( V \) configuration, the upper levels \( |2\rangle \) and \( |1\rangle \) are of the same symmetry and, therefore, the transition \( |2\rangle \rightarrow |1\rangle \) is not dipole allowed. Thus, the external control laser field of frequency \( \omega_L \) which couples levels \( |2\rangle \) and \( |1\rangle \) drives a two-photon transition \( (2\omega_L = \omega_{21}) \). In this case, the interaction Hamiltonian \( H_{AL} \) will be of the form

\[
H_{AL}^V = \hbar \Omega_V [e^{i(2\omega_L t + \phi')} \sigma_{12} - e^{-i(2\omega_L t + \phi')} \sigma_{21}], \tag{3.13}
\]

where the Rabi frequency \( \Omega_V \) is now obtained from second-order perturbation theory[155]

\[
\hbar \Omega_V = \sum_I \frac{(d_{2I}.E_0)(d_{I1}.E_0)}{\hbar(\omega_L - \omega_{I1})}. \tag{3.14}
\]

Here the summation is over all intermediate states \( |I\rangle \) of the atom.

For the sake of notational simplicity (not to write almost identical equations separately for the \( \Lambda \) and \( V \) systems) we write the interaction Hamiltonian \( H_{AL} \) in both the \( \Lambda \) and \( V \) cases as

\[
H_{AL} = \hbar \Omega [e^{i(\omega_{c} t + \phi_c)} \sigma_{12} - e^{-i(\omega_{c} t + \phi_c)} \sigma_{21}], \tag{3.15}
\]

assuming that what we mean by \( \Omega, \omega_c \) and \( \phi_c \) can be understood from the context.
In the framework of perturbation theory, which is usually employed in quantum electrodynamics, the Hamiltonian

$$H_o = H_A + H_R$$

is regarded as the Hamiltonian of the unperturbed system whereas

$$H_I = H_{AR} + H_{AL}$$

describes the perturbation. In this thesis we employ the Schrödinger picture of quantum mechanics[39] where observables are described by time-independent operators. In this picture the basis vectors of the appropriate Hilbert space are stationary (like the fixed co-ordinate system in ordinary geometry), and state vectors move continuously in this space according to the time-dependent Schrödinger equation

$$i \frac{\partial}{\partial t} |\Psi(t)\rangle = H |\Psi(t)\rangle,$$

where $H$ is the Hamiltonian of the system. At any instant of time, a state vector is given by a linear combination of the stationary basis vectors.

The stationary states (eigenstates) of the unperturbed Hamiltonian $H_0$ of Eq. (3.5) are listed below together with their corresponding eigenvalues:

$$|2, \{0\}, \hbar \omega_2; \; |1, \{0\}, \hbar \omega_1; \; |0, \{1_{\kappa \lambda}\}, \hbar (\omega_0 + \omega_k).$$

Here the state vector $|j, \{0\} \equiv |j\rangle|\{0\}\rangle$ represents the atom in the upper states $|j\rangle$ and the vacuum electromagnetic field (that is, no photons present in the system). On the other hand, the state vector $|0, \{1_{\kappa \lambda}\}$ represents the atom in the ground state $|0\rangle$ and a single photon in a mode $\{k\lambda\}$. The state $|j, \{0\}\rangle$ is a direct product of the atomic state $|j\rangle$ and the radiation state $|\{0\}\rangle$, since the atomic operators are assumed to commute with the radiation field operators. Similarly, $|0, \{1_{\kappa \lambda}\}\rangle$ is a direct product of $|0\rangle$ and $|\{1_{\kappa \lambda}\}\rangle$.

\(^1\)On the other hand, in the Heisenberg picture of quantum mechanics, operators are time dependent, the basis vectors are continuously moving (like a rotating system in ordinary geometry), but state vectors are stationary.
The vectors $|1, \{1_{k\lambda}\}\rangle$ are assumed to be inaccessible in the $V$ system, since there is no single photon spontaneous emission on the transition $|2\rangle \rightarrow |1\rangle$. Two-photon spontaneous emission is considered to be negligible compared to the two-photon stimulated emission from $|2\rangle$ to $|1\rangle$, induced by the classical control laser field. This latter effect is described by the classical Rabi field (2-photon transition) amplitude $\hbar \Omega_ e^{i(2\omega_L t + \phi_e)}$. Similarly, in the $\Lambda$ system, single-photon spontaneous emission from $|2\rangle$ to $|1\rangle$, although allowed in the control laser mode, is assumed to be negligible compared to stimulated emission driven by the control laser field\(^3\). For the $\Lambda$-system, the effects of stimulated emission are described by the classical Rabi field (1-photon transition) amplitude $\hbar \Omega_ e^{i(\omega_L t + \phi_e)}$.

We assume the following initial configuration for the model system. At $t = 0$, the radiation-field reservoir is initially in the vacuum state (no photon in the system), and the atom is prepared in a coherent superposition of its two upper levels $|2\rangle$ and $|1\rangle$ in the form

$$|\Psi(0)\rangle = \cos \theta |2, \{0\}\rangle + e^{i\phi_r} \sin \theta |1, \{0\}\rangle.$$  \hspace{1cm} (3.20)

The parameter $\theta$ measures the degree of superposition of levels $|2\rangle$ and $|1\rangle$. A value of $\theta = 0$ means that atom is initially prepared on the upper level $|2\rangle$, whereas $\theta = \pi/4$ means that atom is initially prepared as an equal superposition of the upper levels $|1\rangle$ and $|2\rangle$. The factor $e^{i\phi_r}$ gives the relative phase between the expansion coefficients of $|2\rangle$ and $|1\rangle$, and plays a significant role in physical predictions[156]. The coherent superposition state (3.20) can be prepared by an ultra-short pumping laser pulse of appropriate pulse area[70].

As a result of the perturbation $H_l$ applied at time $t = 0$, the initial state $|\Psi(0)\rangle$ evolves in time according to Schrödinger equation (3.18). At any time $t$, the state vector of the system can be written as a linear combination of the eigenstates of $H_0$. Accordingly, we write

$$|\Psi(t)\rangle = c_2(t)e^{-i\omega_2 t}|2, \{0\}\rangle + c_1(t)e^{-i\omega_1 t}|1, \{0\}\rangle + \sum_{k\lambda} c_{k\lambda}(t)e^{-i(\omega_k + \omega_0) t}|0, \{k\lambda\}\rangle,$$  \hspace{1cm} (3.21)

\(^3\)Single-photon spontaneous emission is NOT allowed into the other modes having frequency $\omega_{21}$ because $\omega_{21}$ is deep within the gap
where the time dependences of the amplitudes due to the unperturbed Hamiltonian $H_0$ are explicitly factored out in the form of exponentials. Comparing Eqs. (3.21) and (3.20), we obtain

$$c_2(0) = \cos \theta, \quad c_1(0) = e^{i\phi} \sin \theta, \quad c_{k\lambda}(0) = 0,$$

as the initial values for the amplitudes $c_{1,2}(t)$ and $c_{k\lambda}(t)$ corresponding to the initial state (3.20).

The function $c_j(t)$ gives the probability amplitude to find the atom in the excited state $|j\rangle$ and the photon reservoir in the vacuum state. On the other hand, $c_{k\lambda}(t)$ gives the probability amplitude to find the atom on the ground state $|0\rangle$ and a single photon of wave vector $k$ and polarization $\lambda$ in the photon reservoir. Thus, the probability of finding the atom in the excited level $|j\rangle$, more commonly known as the population of level $|j\rangle$, is given by

$$n_j(t) = |c_j(t)|^2, \quad (j = 1, 2).$$

The steady-state population of the atomic level $|j\rangle$ is then

$$n_{js} = \lim_{t \to \infty} n_j(t), \quad (j = 1, 2).$$

Using Eq. (3.22) in (3.23), we obtain

$$n_1(0) = \sin^2 \theta, \quad n_2(0) = \cos^2 \theta$$

for the initial populations of the upper levels $|1\rangle$ and $|2\rangle$.

An important quantity in applications such as quantum computing is the cross-term

$$n_c(t) \equiv c_2(t)c_1^*(t).$$

This term measures the interference between the atomic amplitudes $c_1(t)$ and $c_2(t)$ and is
known as the *coherence*. Its steady-state value is

\[ n_{cs} = \lim_{{t \to \infty}} n_c(t). \]  

(3.27)

Using Eqs. (3.3) and (3.21) in Eq. (3.18), and projecting the result onto the eigenstates \(|0, \{k\lambda\} \rangle, |1, \{0\} \rangle\) and \(|2, \{0\} \rangle\) of \(H_o\), respectively, we obtain the following (infinite) set of coupled equations for the amplitudes \(c_j(t)\) and \(c_{k\lambda}(t)\):

\[
\begin{align*}
\dot{c}_{k\lambda}(t) &= g_{k\lambda} c_2(t) e^{i\mu k t}, \\
\dot{c}_1(t) &= \Omega e^{i\phi} c_2(t), \\
\dot{c}_2(t) &= -\Omega e^{-i\phi} c_1(t) - \sum_{k\lambda} g_{k\lambda} c_{k\lambda}(t) e^{-i\mu k t}.
\end{align*}
\]

(3.28) \hspace{1cm} (3.29) \hspace{1cm} (3.30)

Here the dot over an amplitude signifies the total time derivative and

\[ \mu_k = \omega_k - \omega_0 \]

represents the detuning of the radiation mode frequency \(\omega_k\) from the atomic transition frequency \(\omega_0\). Eq. (3.28) can be integrated (in time), using the initial condition (3.22), to give

\[ c_{k\lambda}(t) = g_{k\lambda} \int_0^t c_2(t') e^{i\mu k t'} dt'. \]

(3.32)

Substituting this expression for \(c_{k\lambda}(t)\) in Eq.(3.30) yields the following two coupled integro-differential equations

\[
\begin{align*}
\dot{c}_1(t) &= \Omega e^{i\phi} c_2(t), \\
\dot{c}_2(t) &= -\Omega e^{-i\phi} c_1(t) - \int_0^t G(t - t') c_2(t') dt',
\end{align*}
\]

(3.33) \hspace{1cm} (3.34)

where

\[ G(t - t') = \sum_{k\lambda} g_{k\lambda}^2 e^{-i\mu k\lambda (t-t')} \]

(3.35)

is the delay *Green’s function* of the problem. In writing down Eqs.(3.33)and (3.34) we
have exchanged the order of summation over $k\lambda$ and integration over time. The resulting Green’s function depends very strongly on the photon density of states of the relevant photon reservoir. In essence, $G(t-t')$ is a measure of the photon reservoir’s memory of its previous state on the time scale for the evolution of the atomic system, hence the alternative name memory kernel.

The main objective of this chapter is to solve the coupled equations (3.33) and (3.34) for the amplitudes $c_j(t)$ in a given photon reservoir. These amplitudes can then be used to evaluate the populations and coherences of the atomic levels. The integral on the right hand side of Eq. (3.34) is a convolution integral which suggests solution by Laplace transformation[157]. The Laplace transform $\tilde{f}(s)$ of a function $f(t)$ is defined by

$$\mathcal{L}\{f(t)\} \equiv \tilde{f}(s) = \int_0^\infty e^{-st} f(t)dt.$$  \hspace{1cm} (3.36)

The Laplace transform of the derivative $\dot{f}(t)$ is related to that of $f(t)$ by

$$\mathcal{L}\{\dot{f}(t)\} = s\tilde{f}(s) - f(0).$$ \hspace{1cm} (3.37)

The Laplace transform of the convolution of two functions is the product of the Laplace transforms of the two functions:

$$\mathcal{L}\left\{\int_0^t f(t-t')g(t')dt'\right\} = \tilde{f}(s)\tilde{g}(s).$$ \hspace{1cm} (3.38)

Upon taking the Laplace transforms of Eqs.(3.33) and (3.34) and using the initial condition (3.22), we find that

$$\tilde{c}_2(s) = \frac{s \cos \theta - \Omega e^{i\phi} \sin \theta}{s^2 + s\tilde{G}(s) + \Omega^2},$$ \hspace{1cm} (3.39)

$$\tilde{c}_1(s) = \frac{e^{i\phi} \sin \theta[s + \tilde{G}(s)] + \Omega e^{i\phi c} \cos \theta}{s^2 + s\tilde{G}(s) + \Omega^2},$$ \hspace{1cm} (3.40)
where \( \tilde{c}_j(s) \) and \( \tilde{G}(s) \) are the Laplace transforms of \( c_j(t) \) and \( G(t) \), respectively, and

\[
\phi = \phi_p - \phi_c \tag{3.41}
\]
is determined by the relative phase between the control and pump lasers.

For a given dispersion relation \( \omega_k \), we can calculate \( G(t - t') \) from Eq. (3.35) which, in turn, can be used to calculate \( \tilde{G}(s) \). This \( \tilde{G}(s) \) can then be used in Eqs.(3.39) and (3.40) and the resulting expressions inverted to find analytical expressions for the amplitudes \( c_j(t) \).

### 3.2 Model system in vacuum

In this section, we consider the case when our model system is in free space. Besides being an interesting case in its own right[14], the free space case will be useful to compare and interpret the results of the PBG case which will be discussed in later sections. The leading approximation described by the Hamiltonian (3.3) can be valid in free space if \( \gamma_{10} \ll \gamma_{20} \) in a \( V \) system (so that spontaneous emission on the transition \( |1\rangle \rightarrow |0\rangle \) can be ignored), or if \( \gamma_{21} \ll \gamma_{20} \) in a \( \Lambda \) system (so that spontaneous emission on the transition \( |2\rangle \rightarrow |1\rangle \) can be ignored).

The electromagnetic vacuum is characterized by the dispersion relation

\[
\omega(k) = ck. \tag{3.42}
\]

For such a dispersion relation the Green’s function (3.35) takes the form

\[
G(t - t') = \gamma_{20} \delta(t - t'), \tag{3.43}
\]

where

\[
\gamma_{ij} = \frac{1}{4\pi \varepsilon_0} \frac{4\omega_{ij}^2 d_{ij}^2}{6\hbar c^3} \tag{3.44}
\]
is half the spontaneous emission rate \( \Gamma_{ij} \) for the transition \( |i\rangle \rightarrow |j\rangle \), and \( \delta(t - t') \) is the Dirac delta function[see Appendix(B)]. Thus, in free space, the memory kernel is proportional to
the delta function. This is because free space is an infinitely broad photon reservoir (flat spectrum), and, therefore, its response should be instantaneous. Interactions governed by such a delta function dependent memory kernel are said to be Markovian[14].

From Eq. (3.43) we obtain

$$\tilde{G}(s) = \gamma_0,$$  \hspace{1cm} (3.45)

and using this in Eqs. (3.39) and (3.40) we obtain

$$\tilde{c}_2(s) = \frac{s \cos \theta - \Omega e^{i\phi} \sin \theta}{D(s)},$$

$$\tilde{c}_1(s) = \frac{e^{i\phi_p} \sin \theta (s + \gamma_0) + \Omega e^{i\phi_c} \cos \theta}{D(s)},$$  \hspace{1cm} (3.47)

where

$$D(s) = s^2 + \gamma_0 s + \Omega^2 = \prod_{j=1}^{2} (s - q_j),$$  \hspace{1cm} (3.48)

and $q_j (j = 1, 2)$ are the roots of the quadratic equation

$$x^2 + \gamma_0 x + \Omega^2 = 0,$$  \hspace{1cm} (3.49)

found by substituting $x = s$ in the equation $D(s) = 0$. They are given by

$$q_{1,2} = -\frac{\gamma_0}{2} \pm \sqrt{\left(\frac{\gamma_0}{2}\right)^2 - \Omega^2}.$$  \hspace{1cm} (3.50)

Eqs. (3.46) and (3.47) are easily inverted to give

$$c_2(t) = \sum_{j=1}^{2} D_j e^{\eta_j t}, \quad c_1(t) = \sum_{j=1}^{2} E_j e^{\eta_j t},$$  \hspace{1cm} (3.51)

where

$$D_j = \frac{q_j \cos \theta - \Omega e^{i\phi} \sin \theta}{q_j - q_k}, \quad (j \neq k),$$

$$E_j = \frac{(q_j + \gamma_0)e^{i\phi_p} \sin \theta + \Omega e^{i\phi_c} \cos \theta}{q_j - q_k}, \quad (j \neq k).$$  \hspace{1cm} (3.53)
From Eq. (3.50) we see that both roots \( q_j \) are (a) negative when \( \Omega \leq \gamma_{20}/2 \), and (b) complex (with a negative real part equal to \(-\gamma_{20}/2\)) when \( \Omega > \gamma_{20}/2 \). Thus the time evolution of amplitudes \( c_j(t) \) [and hence of the upper level populations \( n_j(t) \)] can be divided into two regimes of different behavior. For \( \Omega > \gamma_{20}/2 \), the populations display pronounced oscillations before decaying to zero. On the other hand, when \( \Omega < \gamma_{20}/2 \) the populations barely complete an oscillation before decaying to zero. Thus, the driving field induces oscillations on the populations of the upper levels. The stronger the driving field (i.e., the larger the \( \Omega \)), the faster the oscillations. This behavior of the populations is depicted in Fig. (3.2) where \( n_2(t) \) is plotted as a function of the scaled time \( \gamma_{20}t \) for various of \( \Omega \). Even if \( \Omega \leq \gamma_{20}/2 \) (so that both roots \( q_{1,2} \) are real), the coefficients \( D_{1,2} \) of Eq. (3.52) may have opposite signs leading to a minimum in \( n_2(t) \) as is the case for \( \Omega = 0.35\gamma_{20} \) in Fig. (3.2). When \( \Omega = \gamma_{20}/2 \), Eq. (3.49) has a double root \( q_1 = q_2 = -\gamma_{20}/2 \) and inversion of Eqs. (3.46) and (3.47) gives

\[
\begin{align*}
c_2(t) &= \left\{ \cos \theta - \left( \gamma_{20}/2 \right) \cos \theta + \Omega e^{i\phi} \sin \theta \right\} e^{-\left(\gamma_{20}/2\right)t}, \\
c_1(t) &= e^{i\phi} \left\{ \sin \theta + \left( \gamma_{20}/2 \right) \sin \theta + \Omega e^{-i\phi} \sin \theta \right\} e^{-\left(\gamma_{20}/2\right)t}.
\end{align*}
\]

Eq. (3.50) shows that both roots \( q_{1,2} \) have a negative real part, irrespective of the value of \( \Omega \). This means that the amplitudes \( c_{1,2}(t) \) decay in time and tend to zero as \( t \to \infty \) so that the steady state populations \( n_{1s} \) and \( n_{2s} \) are both zero:

\[
n_{js} \equiv \lim_{t \to \infty} |b_j(t)|^2 = 0, \quad (j = 1, 2).
\]

In other words, in free space, the populations of the excited states \(|2\rangle \) and \(|1\rangle \) eventually decay to the ground level \(|0\rangle \) (there is no population trapped on the upper levels), independent of the strength \( \Omega \) of the driving field. The only effect of the driving field is to cause transfer of populations from \(|2\rangle \) to \(|1\rangle \) and vice versa until all the upper level population decays to the ground level. This is a general result valid for almost any broad band smoothly varying electromagnetic density of states. On the other hand, when the density
of electromagnetic modes vanishes in the vicinity of an atomic transition (such as near a photonic band edge) photon localization leads to non-zero steady state atomic populations on the excited levels [65]. The extent of localization depends sensitively on \( \Omega \), and on the initial atomic state as will be seen in section (3.3).

### 3.3 Model System in a PBG material

In this section we investigate the dynamics of our model system in the leading approximation when the atom is located within a photonic-band structure. First, we introduce a model dispersion relation for the PBG material. This dispersion relation is then used to evaluate the corresponding Green’s functions according to Eq. (3.35). The Laplace transform of this Green’s function is then used in Eqs. (3.39) and (3.40) to evaluate \( \hat{c}_j(s) \) which, in turn, is inverted to find analytic expressions for the amplitudes \( c_j(t) \) from which the relevant populations and coherences can be evaluated.
In a PBG one finds a modified dispersion relation for the photons in the radiation reservoir, with a gap(s) in the photon density of states. We begin by considering an isotropic effective mass approximation\cite{5, 6} for the photon dispersion relation in a PBG material:

$$\omega_k \approx \omega_c + A(k - k_0)^2.$$  \hspace{1cm} (3.57)

Here $\omega_c$ is the upper band edge frequency, $k$ is the modulus of the wave vector $k$, $k_0$ is a constant characteristic of the periodic structure, and the constant $A = (1/2)(\partial^2 \omega / \partial k^2)_{k=k_0}$ measures the curvature of the dispersion curve $\omega(k)$ at $k = k_0$\cite{65}. For an isotropic dispersion relation $A \approx \omega_c/k_0^2 \approx c^2/\omega_c$. The dispersion relation (3.57) is valid for frequencies close to the upper photonic band edge. If the photonic band gap is large, and if the relevant atomic transitions are near the upper band edge, it is a very good approximation to completely neglect the effects of the lower band.

The dispersion relation (3.57) is isotropic since it depends only on the magnitude $k$ of the wave vector $k$. While there is no physical PBG material with an isotropic gap, this provides an instructive toy model for studying quantum optical effects. Such a dispersion relation associates the band edge wave vector with a sphere in $k$ space, $|k| = k_0$ (spherical Brillouin zone). By associating the band edge with the entire sphere $|k| = k_0$, the isotropic model (3.57) artificially increases the true phase space available for photon propagation near the band edge. This results in a photonic density of states $\rho(\omega)$ which, near the band edge $\omega_c$, behaves as $(\omega - \omega_c)^{-1/2}$ for $\omega > \omega_c$, the square-root singularity being characteristic of a one-dimensional phase space\cite{5, 6}.

In a real three-dimensional dielectric crystal with an allowed point-group symmetry, the gap is highly anisotropic and the band edge is associated with a point $k = k_0$ (or a finite collection of symmetry related points) in $k$ space\cite{67}, rather than with the entire sphere $|k| = |k_0|$. In other words, the magnitude of the band edge wave vector varies as $k$ is rotated throughout the Brillouin zone. Thus, a more realistic picture of the band edge
behavior requires the incorporation of the Brillouin-zone anisotropy. In the effective mass approximation, the photon dispersion relation takes the vector form

\[ \omega_k \approx \omega_c + A(k - k_0)^2. \]  

(3.58)

In this case, however, we cannot use the approximation \( A \approx c^2/\omega_c \) because the dispersion curve, in general, exhibits different slopes in different directions. Instead, we use \( A \approx f c^2/\omega_c \) where \( f \) is a dimensionless scaling factor, whose value depends on the nature of the dispersion relation near the band edge \( \omega_c \). The anisotropic effective mass dispersion (3.58) relation leads to a photonic density of states at a band edge \( \omega_c \) which behaves as \( \rho(\omega) \sim (\omega - \omega_c)^{1/2} \) for \( \omega > \omega_c \), characteristic of a three-dimensional phase space\[5, 6\].

The isotropic dispersion relation (3.57) leads to qualitatively correct physics. However, the anisotropic model (3.58) introduces important quantitative corrections\[5\]. The most significant difference between the anisotropic and isotropic models comes out more explicitly when considering an undriven two level atom with frequency near the edge of a photonic band-gap. In this case the isotropic model leads to a non-zero steady-state population on the upper level, even when the transition frequency is slightly outside the gap\[65\]. On the other hand, the anisotropic model leads to fractionalized steady state population on the upper level only when the transition frequency is inside the gap\[67\] [see Appendix (E)].

Using the anisotropic effective mass dispersion relation (3.58) in Eq. (3.35) we can evaluate the corresponding Green's function. For \( (t - t') \) large enough to satisfy \( \omega_c(t - t') \gg 1 \), we obtain[see Appendix (B)]

\[ G(t - t') = -\alpha e^{i[\delta(t - t') + \pi/4]} / \sqrt{4\pi(t - t')^3}, \quad \omega_c(t - t') \gg 1, \]  

(3.59)

where

\[ \delta = \omega_0 - \omega_c \]  

(3.60)

represents the detuning of the atomic transition frequency \( \omega_0 \) from the upper band edge frequency \( \omega_c \). The full expression for \( G(t - t') \), including its short time behavior, is rather
complicated[158] but differs from the approximate expression (3.59) only in the region 
\((t - t') \to 0_+\), which is not of much interest to us[158], as we are mainly interested in long

time memory effects.

The constant \(\alpha\) in Eq. (3.59) is given by[see Appendix(B)]

\[
\alpha^2 \approx \frac{1}{16f^3} \left(\frac{\gamma_{20}}{\omega_{20}}\right)^2 \omega_c ,
\]

(3.61)

where \(\gamma_{20}\) [given by Eq. (3.44)] is half the vacuum spontaneous emission rate \(\Gamma_{j0}\) for the

transition \(|2\rangle \to |0\rangle\), and \(f\) is the dimensionless scaling factor mentioned above. At optical

frequencies \(\gamma_{20} \sim 10^8 \text{ Hz}\) and \(\omega_{20} \sim 2\pi \times 10^{15} \text{ Hz}\) so that \(\alpha^2 \sim 10^{-17} f^{-3} \omega_c\). For \(f = 10^3\),

we have \(\alpha^2 \sim 10^{-8} \omega_c\) which translates to \(\alpha^2 \sim 10^7 \text{ Hz}\) when \(\omega_c\) is in the optical regime.

Eq. (3.59) shows that, for an anisotropic PBG in the effective mass approximation, the

memory kernel \(G(t - t')\) decays with time as \((t - t')^{-3/2}\). This is unlike the free space case (3.43), where \(G(t - t')\) exhibits a delta function time dependence. Thus, Eq. (3.59) describes long time memory effects in atom-photon interaction due to the presence of the PBG material, indicating that atom-photon interaction within a PBG material is highly non-Markovian[158]. For the isotropic dispersion relation (3.57) the memory kernel \(G(t - t')\) decays in time more slowly as \((t - t')^{-1/2}\) [see Appendix (B)]. This enhanced memory for

the isotropic model is an artifact of the singular phase space occupied by the band edge

photons of vanishing group velocity.

3.3.2 Populations and Coherences

For the anisotropic effective mass dispersion relation (3.58), the Green's function is given

by (3.59), and its Laplace transform is

\[
\tilde{G}(s) = \alpha e^{i\pi/4} \sqrt{s - i\delta}.
\]

(3.62)
Using this in Eqs. (3.39) and (3.40) we obtain

\[
\tilde{c}_2(s + i\delta) = \frac{(s + i\delta)\cos \theta - \Omega e^{i\phi}\sin \theta}{D(s)},
\]

\[
\tilde{c}_1(s + i\delta) = \frac{(s + \alpha e^{i\pi/4} \sqrt{s} + i\delta)e^{i\phi}\sin \theta + \Omega e^{i\phi}\cos \theta}{D(s)},
\]

where

\[
D(s) = (s + i\delta)^2 + \alpha e^{i\pi/4}(s + i\delta)\sqrt{s} + \Omega^2 = \prod_{j=1}^{4}(\sqrt{s} - e^{i\pi/4} u_j).
\]

Here \(u_j(j = 1, \ldots, 4)\) are the roots of the quartic equation

\[
x^4 + \alpha x^3 + 2\delta x^2 + \alpha \delta x - (\Omega^2 - \delta^2) = 0
\]

found by substituting \(x = e^{-i\pi/4} \sqrt{s}\) in the equation \(D(s) = 0\). These roots are given by\[159\]

\[
u_{1,3} = -\sigma_1 \pm \left[ A - r/2 + \sigma_1^2 \right]^{1/2},
\]

\[
u_2 = \nu_i^* = -\sigma_2 - i \left[ A + r/2 - \sigma_2^2 \right]^{1/2},
\]

where

\[
A = \left( r^2/4 + \Omega^2 - \delta^2 \right)^{1/2},
\]

\[
\sigma_{1,2} = \frac{1}{4} \left( \alpha \pm \sqrt{\alpha^2 - 8\delta + 4r} \right),
\]

\[
r = (B - q/2)^{1/3} - (B + q/2)^{1/3} + \eta_1/3,
\]

\[
B = \left[ \left( \frac{p}{3} \right)^3 + \left( \frac{q}{2} \right)^2 \right]^{1/2},
\]

\[
p = -\frac{\eta_1^2}{3} + \eta_2 , \quad q = -2 \left( \frac{\eta_1}{3} \right)^3 + \frac{\eta_1 \eta_2}{3} + \eta_3,
\]

\[
\eta_1 = 2\delta , \quad \eta_2 = \alpha^2 \delta + 4(\Omega^2 - \delta^2),
\]

\[
\eta_3 = (\alpha^2 - 8\delta)(\Omega^2 - \delta^2) - \alpha^2 \delta^2.
\]

Numerical analysis shows that the roots \(u_{1,3}\) are real (\(u_1\) is positive but \(u_3\) is negative) whereas the roots \(u_{2,4}\) are complex conjugates of each other with a negative real part (\(u_2\).
and $u_4$ lie in the third and second quadrants, respectively).

The amplitude $c_j(t)$ is found by inverting $\tilde{c}_j(s+i\delta)$ using the complex inversion formula\[157\] which involves a contour integration in the complex $s$ plane [see Appendix (C)]. We obtain

$$
c_2(t) = \sum_{j=1}^{2} P_jQ_je^{i(u_j^2+\delta)t} + \frac{\alpha e^{i\pi/4}}{\pi} \int_0^\infty \frac{g_2(x)e^{-(x+i\delta)t}}{Z(x)} dx, \quad (3.76)
$$
$$
c_1(t) = \sum_{j=1}^{2} P_jR_je^{i(u_j^2+\delta)t} + \frac{\alpha\Omega e^{i(\phi_0+\pi/4)}}{\pi} \int_0^\infty \frac{g_1(x)e^{-xt}}{Z(x)} dx, \quad (3.77)
$$

where

$$
P_j = \frac{2u_j}{(u_j - u_l)(u_j - u_m)(u_j - u_n)}, \quad (l, m, n = 1, \ldots , 4, j \neq l \neq m \neq n), \quad (3.78)
$$
$$
Q_j = (u_j^2 + \delta)\cos \theta + i\Omega e^{i\phi}\sin \theta, \quad (3.79)
$$
$$
R_j = (u_j^2 + \alpha u_j + \delta)e^{i\phi}\sin \theta - i\Omega e^{i\phi}\cos \theta, \quad (3.80)
$$
$$
g_2(x) = [(-x + i\delta)\cos \theta - \Omega e^{i\phi}\sin \theta](-x + i\delta)\sqrt{x}, \quad (3.81)
$$
$$
g_1(x) = [(-x + i\delta)\cos \theta - \Omega e^{i\phi}\sin \theta]\sqrt{x}, \quad (3.82)
$$
$$
Z(x) = [(-x + i\delta)^2 + \Omega^2]^2 + i\alpha^2(-x + i\delta)^2. \quad (3.83)
$$

Since $u_1$ is real, and $u_2$ is complex (with negative real and imaginary parts), the first term on the right hand side of Eq. (3.76) is a non-decaying oscillatory term, whereas the second term is also oscillatory but decays exponentially to zero as $t \to \infty$. The last term containing the integral represents the branch cut contribution (arising from the deformation of the contour of integration around a branch point in the complex inversion formula). This also decays to zero as $t \to \infty$, albeit faster than the second term.

Eq. (3.76) shows that level $|2\rangle$ is split into two dressed states. This dressed state splitting is the combined effect of vacuum-field Rabi splitting by the gap[43] and the Autler-Townes splitting[71] by the external field. The dressed states occur at frequencies (noting that root $u_1$ is real, whereas $\text{Re}\{u_2^2\} < 0$)

$$
\omega_{20} - (\delta + \text{Im}\{iu_1^2\}) = \omega_c - \text{Im}\{iu_1^2\} = \omega_c - u_1^2,
$$
\[ \omega_{20} - (\delta + \text{Im}\{iu_2^2\}) = \omega_c - \text{Im}\{iu_2^2\} = \omega_c - \text{Re}\{u_2^2\} = \omega_c + |\text{Re}\{u_2^2\}|. \]

The dressed state at frequency \( \omega_c - u_1^2 \) lies inside the gap and corresponds to the photon-atom bound dressed state with no decay in time. A photon emitted by an atom in such a dressed state will exhibit tunneling on a length scale given by the localization length \( \xi_{\text{loc}} \) before being Bragg reflected back to the emitting atom to re-excite it. For a band gap to center frequency ratio of \( \Delta\omega/\omega_0 \sim 5\% \), the photon localization length \( \xi_{\text{loc}} \geq L \), where \( L \) is the lattice constant of the dielectric\([5, 6]\). The photon-atom bound state is the optical analog of an electron-impurity level bound state in the gap of a semiconductor.

The dressed state at the frequency \( \omega_c + |\text{Re}\{u_2^2\}| \) lies outside the gap and decays at a rate of \( \text{Im}\{u_2^2\} \). It results in highly non-Markovian decay of the atomic population \( n_2(t) \). As \( \omega_{20} \) is detuned further into the gap (i.e. as \( \delta \) becomes more negative), a greater fraction of the light is localized in the gap dressed state. Conversely as \( \omega_{20} \) is moved out of the gap, total emission intensity from the decaying dressed state is increased\([65, 158]\). As a result of interference between the three terms in Eq. (3.76), the spontaneous emission dynamics displays oscillatory behavior\([65]\). As can be seen from Eqs. (3.76) and (3.77), the dynamics of spontaneous emission strongly depends on the detuning \( \delta = \omega_{20} - \omega_c \) of level \(|2\) from the upper band edge, the initial coherent superposition state as defined by the parameter \( \theta \), the intensity \( \Omega \) of the control laser driving the transition between the upper levels, and the relative phase \( \phi = \phi_p - \phi_c \) between the cw control laser field and the pumping laser pulse.

In Fig. (3.3) we plot the atomic population \( n_2(t) \) as a function of the scaled time \( \alpha^2 t \) for various values of the relative phase \( \phi \). This figure shows that, all other conditions being equal, the fractionalized steady state population on the excited states is maximum or minimum when the relative phase is \( \phi = -\pi/2 \) or \( \phi = \pi/2 \), respectively. Fig. (3.4) depicts the population \( n_2(t) \) for various values of \( \Omega \). From this figure we note that, as \( \Omega \) is increased, \( n_2(t) \) oscillates faster and reaches its steady-state value more quickly. Moreover, the steady state value \( n_{2s} \) increases with \( \Omega \).
Figure 3.3: Atomic population $n_2(t)$ in a PBG material as a function of the scaled time $\alpha^2 t$ for $\Omega/\alpha^2 = 2$, $\delta/\alpha^2 = 1$, $\theta = \pi/4$, and for various values of the relative phase $\phi$. The photon dispersion is described by the anisotropic effective mass approximation $(3.58)$. The steady state population of level $|2\rangle$ is largest for the relative phase $\phi = -\pi/2$. The results are obtained in the leading approximation making them applicable to both the $\Lambda$ and $V$ configurations.

Figure 3.4: Atomic population $n_2(t)$ as a function of the scaled time $\alpha^2 t$ for $\delta = 0$ (i.e. when the transition $|2\rangle \rightarrow |0\rangle$ coincides with the anisotropic band edge), for the relative phase $\phi = -\pi/2$, for $\theta = \pi/4$, and for different values of $\Omega$. Note that as $\Omega$ is increased, $n_2(t)$ oscillates faster and reaches its steady-state value more quickly. Moreover, the steady state value $n_{2s}$ increases with $\Omega$. The results are obtained in the leading approximation making them applicable to both the $\Lambda$ and $V$ configurations.
In the long time limit, only the first terms in Eqs.(3.76) and (3.77) remain dominant, since $u_1$ is real whereas $u_2$ is complex with a negative real part. The steady-state populations $n_{js}$ on the upper levels $|2\rangle$ and $|1\rangle$ are thus given by

$$ n_{2s} = |P_1 Q_1|^2 , \quad n_{1s} = |P_1 R_1|^2 \quad (3.84) $$

This phenomenon of population trapping is due to the presence of a PBG material and is absent in free space. It is apparent from Eqs.(3.69)-(3.75), and Eqs. (3.78)-(3.83) that the steady state populations $n_{js}$ depend strongly on the parameters $\theta$, $\phi = \phi_p - \phi_c$, $\delta = \omega_{20} - \omega_c$, and $\Omega$.

Fig. (3.5) shows the variation of the steady-state population $n_{2s}$ of level $|2\rangle$ with respect to the detuning $\delta$. We see that as $\delta$ increases from zero (that is, as level $|2\rangle$ is pushed farther away from the band edge into the continuum) the steady-state population $n_{2s}$ initially increases and attains its maximum value of about 0.295 at about $\delta \approx 0.5\alpha^2$ before it begins to decrease vary rapidly. In other words, there is a fractionalized steady-state atomic population on the excited state $|2\rangle$ even when the bare excitation frequency of this level lies outside of the photonic bandgap, but not far from the band edge. Remarkably, spontaneous emission is partially inhibited even within the allowed electromagnetic continuum as a consequence of quantum interference with the driving field which couples level $|2\rangle$ to the photon-atom bound state associated with level $|1\rangle$. When there is no driving field, our model system can be viewed as a two level system consisting of levels $|2\rangle$ and $|0\rangle$, with the transition frequency $\omega_{20}$ near the edge of a PBG. As shown in Appendix (E), for such a two level atom and the anisotropic dispersion relation (3.58), the steady-state population on the excited level $|2\rangle$ vanishes when the level is at the band edge or outside the gap. However, population trapping in a V system, on level $|2\rangle$ outside the PBG, in the absence of a control laser field, may be recaptured by going beyond the leading approximation and including the spontaneous emission channel $|1\rangle \rightarrow |0\rangle$.

Fig. (3.6) depicts the variation of $n_{2s}$ with respect to the strength $\Omega$ of the driving field for various values of the relative phase $\phi$. This figure shows that $n_{2s}$ can be an increasing
Figure 3.5: Steady-state population $n_2$ of level $|2\rangle$ as a function of the detuning $\delta$ from the anisotropic 3D band edge for $\Omega/\alpha^2 = 3$, $\theta = \pi/4$, and for the relative phase $\phi = -\pi/2$ (which, as seen in Fig. (3.3), leads to a large steady-state population). Note that $n_2$ is non-zero even for $\delta > 0$ (that is, even when $\omega_{20}$ lies outside the gap). In fact $n_2$ attains its maximum value outside the gap at $\delta \approx 0.5\alpha^2$.

Figure 3.6: Steady-state population $n_2$ of level $|2\rangle$ as a function of $\Omega$ for $\delta = 0$, $\theta = \pi/4$, and for different values of $\phi$. $n_2$ can be an increasing or decreasing function of $\Omega$ depending on the relative phase $\phi$. Results for the $\Lambda$ and $V$ configurations are the same in the leading approximation.
or decreasing function of $\Omega$ depending on the value of the relative phase $\phi$.

For a driving laser field so strong that $\Omega \gg \alpha^2, \delta$, the steady state populations $n_{js}$ are given approximately by [see Appendix (C)]

$$n_{2s} \approx n_{1s} \approx \frac{1}{4}(1 - \sin 2\theta \sin \phi), \quad \Omega \gg \alpha^2, \delta. \quad (3.85)$$

Thus, when $\theta = 0$ (when the atom is initially on level $|2\rangle$) or when $\theta = \pi/2$ (when the atom is initially on level $|1\rangle$), we have $n_{2s} = n_{1s} = 1/4$. In other words, for the case of a strong laser field, the steady-state atomic populations $n_{2s}$ and $n_{1s}$ are independent of the initial relative phase $\phi$ (if the system is not initially prepared as a coherent superposition of the upper states). However, if the atom is initially prepared in a coherent superposition of the two upper states $|2\rangle$ and $|1\rangle$, so that $\sin(2\theta) \neq 0$ in Eq. (3.85), the steady state atomic populations will also depend on $\phi$. For instance when $\theta = \pi/4$, spontaneous emission is strongly enhanced ($n_{2s} + n_{1s} \approx 0$) for $\phi = \pi/2$, whereas it is totally suppressed ($n_{2s} + n_{1s} \approx 1$) for $\phi = -\pi/2$. Clearly, the steady state atomic population keeps memory of the initial relative phase $\phi$. It can be controlled by changing the optical paths of the pumping and controlling lasers. Moreover, due to the effects of photon localization, the atom keeps memory of the intensity and phase of the pump (input) laser pulse. This suggests that our model system can serve as an optical memory device on the atomic scale.

Next we evaluate the coherences between the upper levels $|2\rangle$ and $|1\rangle$ as defined by Eqs. (3.26) and (3.27). From Eqs. (3.76) and (3.77) we obtain

$$n_{cs} = \lim_{t \to \infty} c_2(t)c_1^*(t) = |P_1|^2 Q_1 R_1^*, \quad (3.86)$$

and for a very strong control laser field ($\Omega \gg \alpha^2, \delta$) this reduces to [see Appendix (C)]

$$n_{cs} \approx \frac{i e^{-i \phi_c}}{4} (1 - \sin 2\theta \sin \phi), \quad \Omega \gg \alpha^2, \delta. \quad (3.87)$$

Thus, not only do the upper levels $|2\rangle$ and $|1\rangle$ have non-zero populations ($n_{2s}$ and $n_{1s}$) in the steady-state limit, as required for a classical memory device, but the coherences are non-zero
in the steady-state limit, as required for quantum memory. In essence, coherence is forced on the atomic system by means of the external laser field. Like the populations \( n_{1,2}(t) \), the coherence \( n_c(t) = c_2(t)c_1^*(t) \) depends strongly on the parameters \( \theta, \phi = \phi_p - \phi_c, \delta = \omega_0 - \omega_c \) and \( \Omega \). Eq. (3.87) shows that for large \( \Omega \) and when the system is initially prepared in a coherent superposition of the upper states (that is, when \( \theta \neq 0, \pi/2 \)), the coherence \( n_c(t) \) between levels \( |2 \rangle \) and \( |1 \rangle \) can be controlled by the relative phase \( \phi \) and attains its maximum value when \( \phi = -\pi/2 \). In Fig. (3.7) we plot the magnitude \( |n_c(t)| = |c_2(t)c_1^*(t)| \) of the coherence as a function of the scaled time \( \alpha^2 t \) for different values of \( \Omega \). We see that, for the chosen conditions, \( n_c(t) \) increases with increasing \( \Omega \). In chapter (4) we discuss how the coherence \( n_c(t) \) is influenced by other spontaneous emission and non-radiative effects that are not considered within the leading approximation.

The above considerations suggest that quantum information can be "written" onto a single three-level atom by choosing the "area" of the incident laser pulse, the intensity of the cw-laser, and the relative optical path lengths of the cw- and pulse-laser beams. In other words, the precise nature of the information written onto the quantum bit or "qubit" can be controllably altered by varying these external parameters. Furthermore, the phase
and intensity of the control laser field can be adjusted so that spontaneous emission can be totally suppressed in our model system. That is to say, at steady state, the system can be in a coherent superposition of the upper states $|2\rangle$ and $|1\rangle$ as $|\phi \rangle = a_2|2\rangle + a_1|1\rangle$ with $|a_2|^2 + |a_1|^2 = 1$, the amplitudes $a_2$ and $a_1$ being dependent on the phase and intensity of the pump laser pulse. Since this superposition state is immune to single photon radiative decay it is a promising candidate for a two-level quantum bit to encode information in quantum computations. In chapter (4) we discuss some possible decoherence mechanisms. This is the greatest obstacle to quantum computation since it causes a pure quantum state to evolve into a mixture of states and to thereby lose two of its key properties: interference and entanglement[162].
Chapter 4

Coherent Control of Spontaneous Emission Near a Photonic Band Edge: Effects of Other Spontaneous Emission Terms

4.1 Model Hamiltonian and Equation of Motion

In the leading approximation for our model system of Fig. 3.1 we have assumed that spontaneous emission on the transitions $|2\rangle \rightarrow |1\rangle$ and $|1\rangle \rightarrow |0\rangle$ is inhibited, either by symmetry consideration or by the presence of the PBG. We next relax this assumption to see its effects on the system dynamics. To this end we consider the V configuration of Fig. 3.1(b), where the upper levels $|1\rangle$ and $|2\rangle$ are of the same symmetry and are both coupled by dipole transitions to the ground level $|0\rangle$. In this case the unperturbed Hamiltonian $H_o$ is still the same as that of Eq.(3.3), whereas $H_I$ has an additional term due to the now allowed $|1\rangle \rightarrow |0\rangle$ transition. It is given by [see Appendix (A)]

$$H_I = i\hbar\Omega \left[ e^{i(\omega t + \phi_e)} \sigma_{12} - e^{-i(\omega t + \phi_e)} \sigma_{21} \right]$$
where $\Omega$ is the Rabi frequency of the driving field given by Eq. (3.14), $\omega_c = 2\omega_L$ and

$$g_{ij}^{kl} = \frac{\omega_{ij}d_{ij}}{\hbar} \left( \frac{\hbar}{2\epsilon_0\omega_k V} \right)^{1/2} \hat{e}_{k\lambda} \cdot \hat{d}_{ij},$$

is the coupling constant between the atomic transition $|i\rangle \rightarrow |j\rangle$ and the mode $\{k\lambda\}$ of the radiation field. The various factors in Eq. (4.2) are defined in section (3.1.2) in connection with Eq. (3.7).

With interaction Hamiltonian (4.1), Eqs. (3.28)-(3.30) are now replaced by

$$\dot{c}_{k\lambda}(t) = g_{k\lambda}^{20} c_2(t)e^{i\Omega_2 t} + g_{k\lambda}^{10} c_1(t)e^{i\Omega_1 t},$$
$$\dot{c}_1(t) = \Omega e^{i\phi_c} c_2(t) - \sum_{k\lambda} g_{k\lambda}^{10} c_{k\lambda}(t)e^{-i\Omega_{10} t},$$
$$\dot{c}_2(t) = -\Omega e^{-i\phi_c} c_1(t) - \sum_{k\lambda} g_{k\lambda}^{20} c_{k\lambda}(t)e^{-i\Omega_{20} t},$$

where

$$\mu_{ij}^{kl} = \omega_k - \omega_{ij},$$

is the detuning of the radiation mode frequency $\omega_k$ from the atomic transition frequency $\omega_{ij}$. Formal integration (in time) of Eq. (4.3), with the initial condition $c_{k\lambda}(0) = 0$ of Eq. (3.22), yields

$$c_{k\lambda}(t) = g_{k\lambda}^{20} \int_0^t c_2(t')e^{i\Omega_2 t'} dt' + g_{k\lambda}^{10} \int_0^t c_1(t')e^{i\Omega_{10} t'} dt'.$$

Substituting this expression for $c_{k\lambda}(t)$ in Eqs. (4.4) and (4.5) we obtain

$$\dot{c}_1(t) = \Omega e^{i\phi_c} c_2(t) - \int_0^t G_{11}(t - t')c_1(t')dt' - e^{-i\omega_{21} t} \int_0^t G_{12}(t - t')c_2(t')dt',$$
$$\dot{c}_2(t) = -\Omega e^{-i\phi_c} c_1(t) - \int_0^t G_{21}(t - t')c_2(t')dt' - e^{i\omega_{21} t} \int_0^t G_{22}(t - t')c_1(t')dt',$$

where

$$G_{ij}(t - t') = \sum_{k\lambda} g_{k\lambda}^{i0} g_{k\lambda}^{j0} e^{-i\mu_{ij}^{kl}(t-t')}$$

(4.10)
are the delay Green’s functions. Eqs. (4.8) and (4.9) are the generalized versions of Eqs. (3.33) and (3.34) for a V system including spontaneous emission on the transition \(|1\rangle \rightarrow |0\rangle\). As in Chapter (3), we are interested in solving these generalized equations for the amplitudes \(c_{1,2}(t)\) which can then be used to evaluate the populations and coherences of the atomic levels.

In order to explicitly see the effects of the laser field driving the transition \(|2\rangle \rightarrow |1\rangle\) on the system dynamics, we consider the case without a driving field separately from the case with a driving field. We refer to the case without a driving field \((\Omega = 0)\) as the quantum beats case. The case when the driving field is in the form of continuous wave laser so that \(\Omega\) is a non-zero constant independent of time is referred to as the coherent control case. The quantum beats case, besides being an interesting case in its own right, will be a valuable reference case for interpreting the results of the coherent control cases.

4.2 Model System in Vacuum

In this section, we consider the case when the three-level atom in the V configuration is in free space. Using the free space dispersion relation \((3.42)\) in Eq. (4.10), we obtain [see Appendix(B)]

\[
G_{ij}(t - t') = \eta_{ij} \sqrt{\gamma_{m0} \gamma_{0}} \delta(t - t'),
\]

where \(\gamma_{m0}\) is half the spontaneous emission rate for the transition \(|m\rangle \rightarrow |0\rangle\) given by Eq. (3.44), \(\delta(t - t')\) is the Dirac delta function, and

\[
\eta_{ij} = \delta_{ij} + \eta(1 - \delta_{ij}).
\]

Here \(\delta_{ij}\) is the Kronecker delta function and \(\eta\) is a constant [defined in Appendix (B)] which satisfies \(|\eta| \leq 1\), the equality sign holding when the dipoles associated with the transitions \(|i\rangle \rightarrow |0\rangle\) and \(|j\rangle \rightarrow |0\rangle\) are parallel or antiparallel so that \(d_{i0} = \pm d_{j0}\). The factor \(\eta_{ij}\) measures the “strength” of the scattering processes in which a quantum is first emitted in the transition \(|0\rangle \rightarrow |i\rangle\) and then reabsorbed in the transition \(|0\rangle \rightarrow |j\rangle\), or vice versa. We
refer to this factor as the “scattering coefficient”.

Using Eq. (4.11) in the general equations (4.8) and (4.9), we obtain

\[
\begin{align*}
\dot{c}_1(t) &= -\gamma_{10}c_1(t) + \left[\Omega e^{i\phi_c} - \eta \bar{\gamma} e^{-i\omega_{21}t}\right] c_2(t), \\
\dot{c}_2(t) &= -\gamma_{20}c_2(t) - \left[\Omega e^{-i\phi_c} + \eta \bar{\gamma} e^{i\omega_{21}t}\right] c_1(t),
\end{align*}
\]

where

\[
\bar{\gamma} = \sqrt{\gamma_{10}\gamma_{20}}.
\]

If we neglect spontaneous emission on the transition \(|1\rangle \to |0\rangle\) so that both \(\gamma_{10}\) and \(\eta\) are zero, Eqs. (4.13) and (4.14) reduce to Eqs. (3.33) and (3.34) when we use Eq. (4.11). In other words, the leading approximation of chapter (3) is a special case of the general problem considered in this chapter when both \(\gamma_{10}\) and \(\eta\) are set to zero.

### 4.2.1 Quantum Beats in Vacuum

In the quantum beats problem, there is no field driving the transition \(|2\rangle \leftrightarrow |1\rangle\) so that \(\Omega = 0\). In this case, Eqs. (4.13) and (4.14), have closed analytical solutions given by [see Appendix (D.1)]

\[
\begin{align*}
c_2(t) &= e^{-\gamma_{20}t} \sum_{j=1}^{2} A_j e^{q_j t}, \\
c_1(t) &= e^{-(\gamma_{20}+i\omega_{21})t} \sum_{j=1}^{2} B_j e^{q_j t},
\end{align*}
\]

where

\[
\begin{align*}
q_{1,2} &= \frac{\lambda}{2} \pm \sqrt{\left(\frac{\lambda}{2}\right)^2 + (\eta \bar{\gamma})^2}, \\
\lambda &= \gamma_{20} - \gamma_{10} + i\omega_{21}, \\
A_j &= \frac{q_k c_k(0) + \eta \bar{\gamma} c_1(0)}{q_k - q_j}, \quad (k \neq j), \\
B_j &= -q_j A_j / \eta \bar{\gamma}.
\end{align*}
\]

A special case of the quantum beat problem (\(\Omega = 0\)) is when multiple scattering events are ignored so that \(\eta = 0\), such as when the dipole moments associated with the two
allowed transitions are perpendicular. In this special case, Eqs. (4.13) and (4.14) have simple exponentially decaying solutions

\[ c_j(t) = c_j(0)e^{-\gamma_j t}, \quad \text{for } \eta = 0, \]  

(4.21)

where \( c_j(0) \) represents the initial value of \( c_j(t) \) as given by Eq. (3.22). Comparing these solutions with the general solutions Eqs. (4.16) and (4.16) we see that

\[ A_1 = 0, \quad A_2 = c_2(0), \quad B_1 = c_1(0), \quad B_2 = 0, \quad \text{for } \eta = 0. \]  

(4.22)

The phenomenon of quantum beats in the absence of multiple scattering events [that is, when both \( \Omega \) and \( \eta \) are set to zero in Eqs. (4.13) and (4.14)] is now a standard textbook problem[14]. The case when \( \eta = 1 \), that is when the dipoles associated with the two allowed transitions are parallel or antiparallel has been discussed in detail in Ref.[163]. The general model considered here, which is valid for \( 0 \leq \eta \leq 1 \), recaptures these specialized results[164].

Since the roots \( q_{1,2} \) are in general complex, Eq. (4.16) shows that the decay of the amplitudes \( c_{1,2}(t) \) [and hence of the populations \( n_{1,2}(t) \)] are not purely exponential and may display oscillatory behavior depending on the initial coherent superposition state defined by \( c_1(0) \) and \( c_2(0) \), on the decay rates \( \gamma_{10} \) and \( \gamma_{20} \), and on the frequency separation \( \omega_{21} \) between the two upper levels[163]. If, for instance, the system is initially prepared in the state \( |\Psi(0)\rangle = |2\rangle \), then, in the course of time, the population of level \( |1\rangle \) increases from zero to a maximum and then decreases to zero, while that of level \( |2\rangle \) monotonically decreases to zero (see Fig. 4.1).

The detected signal resulting from spontaneous emission from the three-level system is proportional to

\[ J(t) = |\sum_{k\lambda} c_{k\lambda}(t) \exp\{i(kr - \omega_k t)\}|^2 \]  

(4.23)

where \( r \) is the position of the detector relative to the emitting atom[14]. According to Eq. (4.7), the amplitude \( c_{k\lambda}(t) \) contains contributions from \( c_1(t') \) and \( c_2(t') \) (\( 0 \leq t' \leq t \))
which will, in general, interfere with each other. The temporal interference of the two possible transitions $|2\rangle \to |0\rangle$ and $|1\rangle \to |0\rangle$ gives rise to a fluorescence signal that has a component modulated at the difference frequency $\omega_{21}$. This is the phenomenon of quantum beats and is the basis of a spectroscopic technique used to determine the difference in frequency between two atomic levels[14]. When $\eta = 0$, no quantum beats are observed[14] if either $c_1(0)$ or $c_2(0)$ vanishes (i.e. if the system is not initially prepared in a coherent superposition of the upper states). However quantum beats do indeed occur when $\eta \neq 0$, even if either $c_1(0)$ or $c_2(0)$ is zero. In other words, when multiple scattering events are taken into account, quantum beats do indeed occur even if the system is not initially prepared in a coherent superposition of the upper states.

Thus, in the free space quantum beats problem, the "scattering coefficient" $\eta$ plays a prominent role in the system dynamics and it is important to include multiple scattering events. This is especially true when the allowed transitions $|2\rangle \to |0\rangle$ and $|1\rangle \to |0\rangle$ are very close to each other so that $\omega_{21} \ll \omega_{20}, \omega_{10}$. The interference between the two possible transitions accounts for the dark line in the spontaneous emission spectrum of a three level

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Figure 4.1: Atomic populations $n_1(t)$, $n_2(t)$, and total excited-state population $n_e(t) = n_1(t) + n_2(t)$ functions of the scaled time $\gamma_{20}t$ in the vacuum quantum beat problem ($\Omega = 0$) for $\gamma_{10} = 0.5\gamma_{20}$ and $\omega_{21} = \gamma_{20}$. We have assumed that dipoles associated with the two allowed transitions are parallel so that $\eta = 1$ and have used the initial condition $\theta = 0$ (that is, the atom is initially on level $|2\rangle$). The dot-dashed curve is the simple exponential curve $e^{-\gamma_{20}t}$ drawn for reference.
atom in the V configuration observed in Ref.[163]. In the absence of interference between the two spontaneous emission decay processes, one expects the spectrum of the three level atom to consist of two Lorentzian distributions peaked at the two transition frequencies. Instead, what is obtained is a single distribution with a dark band, whose width depends on the decay rates $\gamma_{10}$ and $\gamma_{20}$.

A coherently excited three level atom in the V configuration can decay via the emission of a photon of frequency $\omega_{20}$ or $\omega_{10}$. However, since both transitions lead to the same final atomic state, one cannot determine along which radiative path ($|2\rangle \rightarrow |0\rangle$ or $|1\rangle \rightarrow |0\rangle$) the atom decays. This uncertainty in the radiative trajectory leads to interference of the transition amplitudes, which can be observed as quantum beats. This process is analogous to Young's double-slit experiment where interference takes place because we are unable to distinguish between the different photon paths that lead to the detector. On the other hand, a coherently excited atom in the $\Lambda$ configuration (where $|2\rangle \rightarrow |1\rangle$ and $|2\rangle \rightarrow |0\rangle$ are the only dipole allowed transitions) will also decay along the radiative path $\omega_{20}$ or $\omega_{21}$. However, since the two emission pathways lead to different final states, a measurement of the final state of the atom would tell us which decay channel was taken. Consequently, no beats are expected in this case[14]. The situation would, of course, be different in the presence of a driving field coupling levels $|2\rangle$ and $|1\rangle$.

4.2.2 Coherent Control in Vacuum

In the coherent control problem, the transition $|2\rangle \leftrightarrow |1\rangle$ is driven by a steady-state continuous wave laser characterized by a constant (non-zero) Rabi frequency, $\Omega \neq 0$. In this case the coupled equations (4.13) and (4.14) must, in general, be solved numerically. However, in the special case when $\eta = 0$, the equations can be solved using the method shown in Appendix (D) to solve Eqs. (4.13) and (4.14) in the quantum beats case. We obtain

$$c_j(t) = e^{-\gamma_{j0} t} f_j(t) , \quad (j = 1, 2), \quad (4.24)$$
where
\[ f_2(t) = \sum_{j=1}^{2} D_j e^{r_j^* t}, \quad f_1(t) = e^{-\lambda t} \sum_{j=1}^{2} E_j e^{r_j^* t}, \] (4.25)

with
\[
\lambda = \gamma_{20} - \gamma_{10}, \quad (4.26)
\]
\[
r_{1,2} = \frac{\lambda}{2} \pm \frac{i}{2} \sqrt{\Omega^2 - \left(\frac{\lambda}{2}\right)^2}, \quad (4.27)
\]
\[
D_j = \frac{r_k c_2(0) + e^{i\phi_c} \Omega c_1(0)}{r_k - r_j} \quad (k \neq j), \quad (4.28)
\]
\[
E_j = -e^{-i\phi_c r_j} \frac{D_j}{\Omega}. \quad (4.29)
\]

In both the coherent control case and the quantum beat case, the population dynamics depends on the initial coherent superposition state [as defined by \( \theta \) and \( \phi_p \)] as well as on the parameters \( \gamma_{10}, \gamma_{20}, \omega_{21}, \) and \( \eta. \) In the coherent control case, the atomic population has an additional dependence on the intensity \( \Omega \) and phase \( \phi_c \) of the control laser field. The driving field causes transfer of populations from \( |2\rangle \) to \( |1\rangle \), as shown in Fig. 4.2 by the oscillations in \( n_1(t) \) for various values of \( \Omega \). The stronger the driving field, the higher the frequency of oscillation of the populations \( n_{1,2}(t) \). For free space, the steady state atomic populations on the upper levels are zero, irrespective of the strength \( \Omega \) of the control laser field. In other words there is no population trapping in free space.

### 4.3 Model System in a PBG Material

For the anisotropic effective mass dispersion relation (3.58), the Green’s functions (4.10) take the form [see Appendix (B)]
\[
G_{ij}(t - t') = -\eta_{ij} \alpha \frac{e^{i[\delta_{j1}(t-t') + \pi/4]}}{\sqrt{\pi(t-t')^3}} \quad \omega_c(t - t') \gg 1 \quad (4.30)
\]
where \( \eta_{ij} \) and \( \alpha^2 \) are given, respectively, by Eqs. (4.12) and (3.61), and

\[
\delta_{ij} = \omega_{ij} - \omega_c
\]  

(4.31)

is the detuning of the atomic transition frequency \( \omega_{ij} \) from the upper band-edge frequency \( \omega_c \). Substituting Eq. (4.30) into Eqs. (4.8) and (4.9) we can find the coupled equations for the amplitudes \( c_{1,2}(t) \) appropriate for a PBG analogous to Eqs. (4.13) and (4.14) for vacuum. However, in the PBG case, it is convenient to introduce the new amplitudes \( h_{1,2}(t) \):

\[
c_j(t) = h_j(t)e^{i\delta_{j0}t}, \quad (j = 1, 2).
\]  

(4.32)

In terms of these new amplitudes and the Green’s functions (4.30), Eqs. (4.3) and (4.4) can be rewritten as

\[
\dot{h}_1(t) = -i\delta_{10}h_1(t) + \Omega e^{i(\omega_{21}t + \phi_e)}h_2(t) - \int_0^t G(t - t')[h_1(t') + \eta h_2(t')]dt', \\
\dot{h}_2(t) = -i\delta_{20}h_2(t) - \Omega e^{-i(\omega_{21}t + \phi_e)}h_1(t) - \int_0^t G(t - t')[h_2(t') + \eta h_1(t')]dt',
\]  

(4.33)  

(4.34)
where
\[ G(t - t') = -\alpha e^{i\pi/4} \sqrt{4\pi(t - t')^3}. \] (4.35)

In the next sections we discuss the coupled equations (4.33) and (4.34) for the quantum beats and coherent control cases, separately.

### 4.3.1 Quantum Beats Near the Edge of a PBG

The problem of quantum beats near the edge of a photonic band gap corresponds to the case when \( \Omega = 0 \) in Eqs. (4.33) and (4.34). This case has also been investigated in Ref.[160], using the "effective mass" isotropic dispersion model (3.57). In this chapter we discuss the problem using the more realistic anisotropic dispersion model (3.58).

For \( \Omega = 0 \), Eqs. (4.33) and (4.34) can solved to give closed analytic expressions for the amplitudes \( h_{1,2}(t) \). These expressions take particularly simple forms when the band-edge \( \omega_c \) is midway between the two upper of levels of the V system so that \( \delta_{20} = -\delta_{10} = \delta \) (thus \( \delta \geq 0 \)), and when the atomic dipoles associated with the transitions \( |2\rangle \rightarrow |0\rangle \) and \( |1\rangle \rightarrow |0\rangle \) are parallel (or antiparallel) so that \( \eta = 1 \). In this special case the solutions to Eqs. (4.33) and (4.34) are given by [see Appendix (D.2)]

\[
\begin{align*}
c_2(t) &= \sum_{j=1}^{2} S_j T_j e^{i(v_j^2 + \delta)t} + \frac{e^{i\pi/4}}{\pi} \int_0^\infty \frac{f_2(x) e^{-(x+i\delta)\mu} dx}{W(x)}, \\
c_1(t) &= \sum_{j=1}^{2} S_j U_j e^{i(v_j^2 + \delta)t} + \frac{e^{i\pi/4}}{\pi} \int_0^\infty \frac{f_1(x) e^{-(x-i\delta)\mu} dx}{W(x)},
\end{align*}
\] (4.36)

where

\[
\begin{align*}
S_j &= \frac{2v_j}{(v_j - v_l)(v_j - v_m)(v_j - v_n)}, \quad (l, m, n = 1, ..., 4, j \neq l \neq m \neq n), \quad (4.38) \\
\rho &= \alpha[c_2(0) - c_1(0)], \quad (4.39) \\
T_j &= (v_j^2 - \delta)c_2(0) - v_j \rho = (v_j^2 + \alpha v_j - \delta)c_2(0) - \alpha v_j c_1(0), \quad (4.40) \\
U_j &= (v_j^2 + \delta)c_1(0) - v_j \rho = (v_j^2 + \alpha v_j + \delta)c_1(0) - \alpha v_j c_2(0), \quad (4.41) \\
f_2(x) &= [\rho(x^2 + \delta^2) + 2\alpha c_2(0)(x + i\delta)x] \sqrt{x},
\end{align*}
\] (4.42)
\[ f_1(x) = [\rho(x^2 + \delta^2) + 2\alpha c_1(0)(x - i\delta)x]\sqrt{x}, \quad (4.43) \]
\[ W(x) = (x^2 + \delta^2)^2 + i4\alpha^2 x^3. \quad (4.44) \]

Here \( v_j (j = 1, ..., 4) \) are the roots of the quartic \( x^4 + 2\alpha x^3 - \delta^2 = 0 \) given by [159]

\[
\begin{align*}
    v_{1,3} &= -\sigma_1/2 \pm \sqrt{(\sigma_1/2)^2 - \xi_2}, \\
    v_2 &= v_4^* = -\sigma_2/2 - i\sqrt{\xi_1 - (\sigma_2/2)^2}, \\
    \sigma_{1,2} &= \alpha \pm \sqrt{\alpha^2 + u}, \\
    \xi_{1,2} &= u/2 \pm \sqrt{(u/2)^2 + \delta^2}, \\
    u &= -(2\alpha^2 \delta^2)^{1/3} [(A + 1)^{1/3} - (A - 1)^{1/3}], \\
    A &= [1 + (4/27)(2\delta/\alpha^2)^2]^{1/2}. \quad (4.49)
\end{align*}
\]

Roots \( v_1 \) and \( v_3 \) are both real, whereas roots \( v_2 \) and \( v_4 \) are complex conjugates of each other. We have dropped the global phase factor \( e^{i\delta t} \) from the right hand sides of Eqs. (4.36) and (4.37), since such a phase factor does not play a role in physical predictions.

The solutions (4.36) and (4.37) for the amplitudes \( c_{1,2}(t) \) show that (a) the spontaneous emission is oscillatory and (b) each of the upper levels splits into two dressed states analogous to vacuum-field Rabi splitting in a high-Q cavity[43]. The splitting is solely due to the interaction of the atom with the photon-reservoir, since there is no driving field. Furthermore, (c) there is a fractionalized steady-state population on each of the upper levels as a result of the localization of light in the vicinity of the emitting atom, and (d) quantum interference leads to non-zero steady state population on level \( |2\rangle \) even when it lies outside the PBG (but not far form the band-edge). This reveals an important distinction between the realistic anisotropic PBG model and the isotropic dispersion model[160]. In the anisotropic model, spontaneous emission from level \( |2\rangle \) (outside of the PBG) can be inhibited by quantum interference with level \( |1\rangle \) (inside the PBG). This inhibition does not occur in the absence of the coupling to level \( |1\rangle \). In the isotropic model, inhibition of spontaneous emission from level \( |2\rangle \) occurs even in the absence of coupling to level \( |1\rangle \).
In Fig. 4.3 we plot, using the expression (4.36) and (4.37), the atomic populations $n_{1,2}(t)$ as functions of the scaled time $\alpha^2 t$ assuming that, initially, the atom was on level $|2\rangle$. As a result of quantum interference between the two allowed transitions, the population of level $|1\rangle$ (which was initially zero) increases from zero to a maximum before it settles down to a steady-state value. Similar oscillations occur in free space quantum beats [Eqs. (4.16) and (4.16)]. The major difference is the non-zero steady state populations in the PBG case. These steady state populations are given by

$$n_{2s} = |S_1 T_1|^2, \quad n_{1s} = |S_1 U_1|^2,$$

(4.51)

and depend on the parameters $\theta$, $\phi$ and $\delta$. Fig. (4.4) shows $n_{2s}$ as a function of the detuning $\delta$, for different values of $\theta$. We notice that, even in the absence of a driving field which couples level $|2\rangle$ to the photon atom bound state associated with level $|2\rangle$, there is a steady-state population on level $|2\rangle$, provided that it is not far from the upper band edge $\omega_c$. In other words, quantum interference allows the partial inhibition of spontaneous emission even in the normally allowed continuum.

In order to see the detailed differences between the isotropic and anisotropic model dispersion relations [Eqs. (3.57) and (3.58), respectively], we plot in Fig. 4.5 the populations $n_{1,2}(t)$ in the PBG quantum beats problem for the isotropic dispersion model, assuming that the atom was initially on level $|2\rangle$. Apart from the difference in time scales, the main distinction between the two models is that interference of spontaneous emission between the two allowed transitions and the localization effects of the photonic band gap are considerably enhanced for the isotropic model relative to the anisotropic model. In the isotropic model, the populations oscillate for hundreds of cycles before decaying to their final larger steady-state values. The amplitudes of these oscillations depend on the initial values $c_2(0)$ and $c_1(0)$. As mentioned in section (3.3.1), this enhancement is an artifact of the singular photon density of states at the isotropic band edge.
Figure 4.3: V system atomic populations $n_2(t)$, $n_1(t)$, and total excited-state population $n_e(t) = n_2(t) + n_1(t)$ as functions of the scaled time $\alpha^2 t$ in the PBG quantum beats problem ($\Omega = 0$) for the initial condition $\theta = 0$ (i.e. atom initially on level $|2\rangle$) and for $\phi = -\pi/2$ and $\eta = 1$. The anisotropic band edge is midway between the two upper levels with detuning $\delta_{01} = -\delta_1 0 = 0.5\alpha^2$. Note that, as a result of quantum interference between the two allowed transitions, the population of level $|1\rangle$ (which was initially zero) increases from zero to a maximum before it settles down to a steady-state value of about 0.05.

Figure 4.4: Steady-state population $n_{2s}$ of level $|2\rangle$ as a function of the detuning $\delta$ from the anisotropic 3D band-edge in the quantum beats problem ($\Omega = 0$), for $\phi = 0$, $\eta = 1$ and for different values of $\theta$. That $n_{2s} \neq 0$ for $\delta > 0$ (that is, when level $|2\rangle$ lies outside the gap) shows that quantum interference with the transition $|1\rangle \rightarrow |0\rangle$ (which lies inside the gap) leads to partial inhibition of spontaneous emission even in the normally allowed continuum.
Figure 4.5: V system atomic populations $n_2(t)$, $n_1(t)$, and total excited-state population $n_e(t) = n_2(t) + n_1(t)$ as functions of the scaled time $\beta t$ in the PBG quantum beats problem ($\Omega = 0$) for the isotropic dispersion model (3.57). The quantities $\theta$, $\phi$, $\delta_{20}$, $\delta_{10}$, and $\phi$ are the same as those in Fig. 4.3, which is the corresponding figure for the anisotropic dispersion model (3.58). Clearly visible are the vacuum Rabi oscillations and the fractional localization near the photonic band edge. These oscillations and localization are considerably enhanced for the isotropic model relative to the anisotropic model.

4.3.2 Coherent Control Near the Edge of a PBG

When $\Omega \neq 0$, Eqs. (4.33) and (4.34) correspond to the problem of coherent control near the edge of a PBG. In this case the equations do not have simple analytic solutions. They must be solved numerically. For illustration purpose it is simpler to use the isotropic rather than the anisotropic model. The Green's function $G_{im}(t - t')$ of the isotropic model [Eq. (B.19)] exhibits an integrable square root singularity [165] at $t = t'$, whereas the complete Green's function in the anisotropic model, which also has an integrable square root singularity at $t = t'$, is rather cumbersome [158].

Fig. 4.6 depicts the PBG coherent control problem for the isotropic dispersion model (3.57), with the additional spontaneous emission effects included. Note that, for $\Omega \neq 0$, $n_2(t)$ displays rapid oscillations within a relatively slowly varying envelope. When $\Omega \neq 0$, there are two causes for the oscillations of $n_2(t)$. The first one (slow oscillations) is the quantum interference between the two allowed transitions ($|2\rangle \rightarrow |0\rangle$ and $|1\rangle \rightarrow |0\rangle$), as in the quantum beats problem. Superimposed on this is the exchange of populations between levels $|2\rangle$ and $|1\rangle$ caused by the driving field (rapid oscillations). As $\Omega$ increases the amplitude
Figure 4.6: V system atomic population $n_2(t)$ as a function of the scaled time $\beta t$ in the PBG (including spontaneous emission channels which go beyond the leading approximation) for $\theta = \pi/4$, $\phi = -\pi/2$, and for various values $\Omega$. The isotropic band edge is midway between the two upper levels with detuning $\delta_{20} = -\delta_{10} = 0.5\beta$, and $\eta = 1$. For $\Omega \neq 0$, $n_2(t)$ displays rapid oscillations within a slowly varying envelope. The frequency of the oscillations within the envelope increases with $\Omega$.

of the envelope oscillations decreases but the frequency of the oscillations within the envelope increases. Moreover, the steady state value $n_{2s}$ increases with $\Omega$. In fact for large $\Omega$, $n_2(t)$ changes little from its initial value $n_2(0)$ even though $\omega_{20}$ lies slightly outside the gap ($\delta_{20} = 0.5\beta$). This is because, when $\Omega$ is large, level $|2\rangle$ will be strongly coupled to level $|1\rangle$ which lies inside the gap ($\delta_{10} = -0.5\beta$).

In Fig. 4.7 we plot the magnitude $|n_c(t)|$ of the coherence $n_c(t)$ as a function of the scaled time $\beta t$ for different values of $\Omega$. The scale factor $\beta$, given by Eq. (B.22), is of the order $\beta \sim 10^{10} \text{s}^{-1}$ at optical frequencies, and, therefore, is larger than the normal vacuum Lamb shift of ($\sim 10^9 \text{Hz}$) of the $2p_{1/2}$ level of hydrogen relative to the $2s_{1/2}$ level. Comparing Fig. 4.7 with Fig. 3.7, we see that, just as in the case of the populations $n_{1,2}(t)$, the spontaneous emission channel $|1\rangle \rightarrow |0\rangle$ introduces further oscillations to the coherence $n_c(t)$ over and above those induced by the driving field. Nevertheless, we obtain non-zero steady-state coherences (and populations) as long as level $|2\rangle$ is not detuned far outside the gap.

The results in this section are qualitatively similar to those in section (3.3.2) where
spontaneous emission on the transition $|1\rangle \rightarrow |0\rangle$ is neglected. The incorporation of the decay channel $|1\rangle \rightarrow |0\rangle$, together with the use of the isotropic dispersion relation, leads to additional oscillations in the transient dynamics. However, it does not alter the presence of non-zero steady-state populations and coherences on the upper levels nor does it alter the ability to control these steady-state populations and coherences by the intensity and phase of the driving field.

4.4 Higher order radiative and non-radiative interactions

As discussed in section (3.3.2), an excited atom in a PBG interacts strongly with its own radiation field, leading to the formation of the photon-atom bound state, in which the photon emitted by the excited atom can tunnel through the dielectric host on a length scale given by the localization length $\xi_{loc}$ before being Bragg reflected back to the emitting atom. The result is a stationary state superposition of a localized photon and a partially excited atom as manifested by the nonzero fractionalized steady state population given by Eqs. (3.84) and (4.51).

Inside a PBG, single photon spontaneous emission is inhibited. Thus the photon-atom
bound state can decay only by other relaxation mechanisms[166, 167]. One such mechanism is spontaneous two photon emission. This may be relevant for the case of a cold atom which has been optically trapped in the void regions of the PBG material and, therefore, is not in mechanical contact with the vibrational degrees of freedom of the dielectric host. For a dipole allowed transition such as the $|1\rangle \rightarrow |0\rangle$ transition in a V system [Fig. 3.1(a)], two-photon decay yields a lifetime for the photon atom bound state on the scale of days[6], if the transition lies in the visible spectrum. On the other hand, for a dipole forbidden transition such as the $|1\rangle \rightarrow |0\rangle$ in the $\Lambda$ configuration [Fig. 3.1(b)], two-photon emission may occur by means of a pair of dipole transitions which occurs considerably faster. For instance the $2s \rightarrow 1s$ transition in Hydrogen occurs in $1/7$ second.

For an impurity atom embedded in a solid dielectric host, the vibrational modes of the host can provide an alternative relaxation mechanism for the photon-atom bound state by altering the electronic spectrum of the impurity[6]. Next we give a simple semi-quantitative discussion of phonon relaxation for the $V$ system depicted Fig. 3.1(a). In a semi-classical picture, phonon interactions cause the energy levels of an atom to experience small, random, time-varying, Stark shifts. In our simplified picture, we assume that this phenomenon can be modeled by adding random shifts $\delta \omega_{j0}(t)$ to the transition frequencies $\omega_{j0}$. The random functions $\delta \omega_{j0}(t)$ are as often positive as negative and hence the ensemble averages $\langle \delta \omega_{j0}(t) \rangle$ are zero. Thus we can simulate phonon interaction by Gaussian random variables $\delta \omega_{j0}$ of zero mean and variance $\gamma$, whose value depends on the strength of these interactions. We assume that the phonon reservoir is Markovian[14] so that the averages of the products $\langle \delta \omega_{j0}(t) \delta \omega_{j0}(t') \rangle$ are zero unless $t \approx t'$. We also assume that variations in $\delta \omega_{j0}(t)$ are very rapid compared to other changes in the system which occur on the time-scale $1/\gamma_{j0}$ (where $\gamma_{j0}$ is the free space spontaneous emission rate for the transition $|j\rangle \rightarrow |0\rangle$), and take

$$\langle \delta \omega_{j0}(t) \delta \omega_{j0}(t') \rangle = \gamma_{j0d} \delta(t - t'), \quad (j = 1, 2)$$

(4.52)

where $\gamma_{j0d}$ are the dephasing rates which are of the order of the Debye frequency $\nu_D$ of
the crystal\textsuperscript{1}. Thus, when phonon interactions are taken into account, Eqs. (4.33) and (4.34) have to be rewritten with \( \delta j_0 \) and \( \omega_{21} \) replaced by, respectively, \( \delta j_0 + \delta \omega j_0(t) \) and \( \omega_{21} + \delta \omega_{20}(t) - \delta \omega_{10}(t) \), where \( \delta j_0 \), and \( \omega_{21} \) are the corresponding quantities in the absence of random Stark shifts. For example if \( \delta_{20} \) is set to zero, it means that, in the absence of Stark shifts, the transition frequency \( \omega_{20} \) coincides with the photonic band edge \( \omega_c \) and, therefore, the shifts \( \delta \omega_{20}(t) \) slightly detune level \( |2\rangle \) in and out of the band gap in a random fashion.

Fig. 4.8 depicts the excited state population \( n_2(t) \) on level \( |2\rangle \) as a function of the scaled time \( \beta t \) for \( \delta_{20} = -\delta_{10} = 0.5\beta \), and for different values of \( \Omega \), when \( \delta \omega_{20}(t) \) and \( \delta \omega_{10}(t) \) are taken as Gaussian random variables of zero mean and 0.5\( \beta \) variance. These and other numerical simulations show that, even when the dephasing rate \( \gamma_{20d} \) is comparable to \( \beta \), the phase sensitive memory effects which we obtained without including dephasing effects, can be recaptured provided that the external Rabi frequency \( \Omega \) is large compared to the dephasing rate. In other words, \textit{dephasing effects simply determine the minimum required intensity of the external laser field for achieving coherent control of radiative dynamics.}\textsuperscript{2} The effect of the random shifts of the atomic levels \( |1\rangle \) and \( |2\rangle \) on the coherence \( n_c(t) \) between the levels is shown in Fig. 4.9. We see that, just in the case of the populations, these effects can be offset by intense driving fields.

### 4.5 Discussion and Conclusions

Atom-photon interaction in free space is characteristically Markovian, meaning that the future of an atomic system coupled to a free space photon reservoir is entirely determined by the present and not by the past - \textit{the atom loses all memory of its past}. Mathematically, this is expressed by the delta function time dependence of the memory kernel (3.43) associated with free space.

By contrast, atom-photon interaction in a PBG photon reservoir is highly non-Markovian.

\textsuperscript{1}The Debye frequency of a crystal is a measure of the maximum photon frequency in the crystal\cite{17}. For Silicon \( \omega_D \approx 1.25 \times 10^{13} \text{Hz} \) whereas for Germanium \( \omega_D \approx 7.2 \times 10^{12} \text{Hz} \)

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Figure 4.8: V system excited state population $n_2(t)$ on level $|2\rangle$ near the isotropic photonic band edge as a function of the scaled time $\beta t$ for $\theta = \pi/4$, $\phi = -\pi/2$, and for different values of $\Omega$, in the presence of dipolar dephasing Gaussian random Stark shifts $\delta \omega_{20}(t)$ and $\delta \omega_{10}(t)$ (each of zero mean and $0.5\beta$ variance) of the transition frequencies $\omega_{20}$ and $\omega_{10}$. In the absence of the random Stark shifts, the band edge is assumed to be midway between the two upper levels with detuning $\delta_{20} = -\delta_{10} = 0.5\beta$. Compare this figure with the corresponding figure (Fig. 4.6) in the absence of phonon mediated dephasing.

Figure 4.9: V system coherence $n_c(t) = |c_2(t)c_1^*(t)|$ between levels $|2\rangle$ and $|1\rangle$ near the isotropic photonic band edge as a function of the scaled time $\beta t$ for $\theta = \pi/4$, $\phi = -\pi/2$, and for different values of $\Omega$, in the presence of dipolar dephasing Gaussian random Stark shifts $\delta \omega_{20}(t)$ and $\delta \omega_{10}(t)$ (each of zero mean and $0.5\beta$ variance) of the transition frequencies $\omega_{20}$ and $\omega_{10}$. In the absence of the random Stark shifts, the band edge is assumed to be midway between the two upper levels with detuning $\delta_{20} = -\delta_{10} = 0.5\beta$. Compare this figure with the corresponding figure (Fig. 4.7) in the absence of phonon mediated dephasing.
The atomic level splitting, oscillatory behavior, and fractionalized steady-state atomic population on an excited state are all direct consequences of this highly non-Markovian interaction. It is distinct from the well-known Jaynes-Cummings oscillations which arise from the interaction of the atom with an isolated cavity or dielectric mode[122]. In our model no defect mode is present.

The dynamics of the excited state populations, their steady-state values, and their coherences strongly depend on the detuning $\delta = \omega_{20} - \omega_c$ of level $|2\rangle$ from the upper band edge, the initial coherent superposition state as defined by the parameter $\theta$, the intensity $\Omega$ of the control laser driving the transition $|2\rangle \leftrightarrow |1\rangle$, and the relative phase $\phi = \phi_p - \phi_c$ between the control laser field and the pumping laser pulse used to prepare the initial state. In particular, the steady-state populations and coherences keep memory of the relative phase $\phi$ as well as the intensity and phase of the pumping laser pulse. This suggests a possible application as an atomic-scale optical memory device. Moreover, since both populations and coherences can be maintained between the two upper atomic levels $|1\rangle$ and $|2\rangle$, these levels can be used as a qubit (two-state quantum system) to encode information for quantum computation. Two or more such systems can be used to construct quantum logic gates[171, 172]. An experimental realization of a quantum computer requires both (a) isolated quantum systems that act as qubits, and (b) the presence of controlled interaction between the qubits that allows for construction of quantum logic gates.

Chapter(4) deals with the influences of other spontaneous emission terms and non-radiative effects on the results found in chapter (3), using the V configuration as an example. The inclusion of the spontaneous emission channel $|1\rangle \rightarrow |0\rangle$ leads to quantum interference between the two allowed transitions $|2\rangle \rightarrow |0\rangle$ and $|1\rangle \rightarrow |0\rangle$ which induces further oscillations to the populations and coherences over and above those induced by the driving field. Moreover, in the absence of a driving field coupling levels $|2\rangle$ and $|1\rangle$, quantum interference between the two allowed transitions $|2\rangle \rightarrow |0\rangle$ and $|1\rangle \rightarrow |0\rangle$ leads to the partial inhibition of spontaneous emission even in the normally allowed continuum. However, the inclusion of the spontaneous emission channel $|1\rangle \rightarrow |0\rangle$ does not alter the presence of non-zero
steady-state populations and coherences on the upper levels, nor does it alter the ability to control these steady-state populations and coherences by the intensity and phase of the driving field.

Phonon dephasing is a relevant decay mechanism for the photon atom bound state in the case of impurity atoms embedded in the solid fraction of the PBG material. Such dephasing effects can be offset by making the Rabi frequency of the control laser field large compared to the dephasing rates.

In both chapters (3) and (4) we employed the effective mass dispersion relation (3.58) or (3.57). While the effective mass approximation gives qualitatively correct physics[6], an investigation of our model system using a full anisotropic dispersion relation of a realistic band structure is a worthy undertaking[12, 170]. This would involve a realistic evaluation of the Green’s function[67] using the full dispersion relation ωk appropriate to a real photonic crystal [see for instance Eq. (B.5)]. The resulting equations of motion for radiative dynamics would then need to be solved numerically.

Finally there is the question of how to place the active elements (the three level atoms in our case) inside the photonic crystal. From a material standpoint, it is possible to dope an existing PBG material using ion beam implantation methods. For instance, it has recently been shown that Er3+ ions implanted into bulk silicon exhibit sharp free-atom-like spectra[173, 174, 175, 176]. Intense photoluminescence (PL) at 1.54 μm is observed in the system at low temperatures2. This wavelength is particularly significant because it corresponds to the minimum absorption of silica fiber-based optical communication system. Because the photoluminescence at 1.54 μm is due to the spin-orbit split 4I_{13/2} \rightarrow 4I_{15/2} of 4f electrons in the Er3+ ions which are shielded by outer 5s^25p^6 shells, the influence of the host lattice on the luminescence wavelength is weak3. It would be of considerable interest to study the radiative properties of Er3+ ions implanted into a 3D silicon PBG material in which a photonic band gap is engineered to occur at 1.54 μm. In spite of the screening of

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2When the host material is crystalline, Er-related photoluminescence is quenched at temperatures above 80 K so that it cannot be detected at room temperatures.

3The key to the success of erbium is that the upper level of the amplifying transition 4I_{13/2} is separated by a large energy gap from the next-lowest level 4I_{15/2} so that its lifetime is very long and mostly radiative. The value of the lifetime is around 10 ms and varies depending on the host and erbium concentration.
the atomic transition by the outer shells, it is likely that thermal phonons in the silicon host would cause significant dephasing of the quantum degrees of freedom within the Erbium 4f shell. Consequently such a system must be cooled to liquid Helium temperatures.

Alternatively, active elements can be placed inside a photonic crystal by pumping a dilute atomic vapor into the void region of the crystal. If such atoms are optically trapped in the void regions, they are not in mechanical contact with the vibrational degrees of freedom of the dielectric host and therefore do not experience phonon dephasing effects as in the case of Erbium doped silicon. Doppler broadening due to the random motion of the gas molecules may be partially alleviated by laser cooling techniques[177].

A third approach to realize our model system is by means of an “artificial atom” or quantum dot structure embedded in the solid fraction of the PBG material. Semiconductor quantum dots (QDs) are nanoscale quantum structures that allow electronic properties to be tailored through quantum confinement. They exhibit distinctive features similar to atoms such as atomic-like excitation spectra with discrete and extremely sharp spectral lines[178]. With their well-defined localized states, QDs offer the possibility of coherent manipulation of a single localized quantum system in a way similar to that achieved in atoms but with the technological advantages of a solid-state system.
Chapter 5

Discrete Bound States and Resonance Raman Scattering in Photonic Band Gap Materials

In this chapter we study the resonance Raman scattering of light from a three-level atom embedded in a PBG material or in a frequency-dispersive medium whose photon spectrum exhibits a gap due to photon coupling to medium excitations such as excitons and optical phonons. We demonstrate that the one-particle spectrum of the system consists of either a continuous part with energy lying outside the gap or a single discrete mode with energy lying inside the gap. In the case of the continuous spectrums, the Rayleigh and Stokes lines are shifted as well as narrowed (or broadened) as the corresponding transition frequencies are shifted relative to the upper band edge, providing a distinctive experimental signature of atom-photon interactions near a photonic band edge. Sec. (5.1) deals with the description of the modes system and the derivation of the equations of motion for the relevant variables. These equations are then investigated in Sec. (5.2) for discrete bound-state solutions and for continuous spectra. Sec. (5.3) deals with the case when the model system is in a PBG material; Sec. (5.4) deals with the case when the model system is in a frequency dispersive medium.
5.1 Description of the Model system

5.1.1 Model Hamiltonian

We consider the resonance Raman scattering of a laser pulse of frequency $\omega_L = \omega_{20}$ from the three-level atom embedded in a PBG material or in a Frequency Dispersive Medium (FDM). The three level atom is assumed to be in the $\Lambda$ configuration [see Fig. 5.1] where the uppermost atomic level $|2\rangle$ is dipole coupled to the lower levels $|1\rangle$ and $|0\rangle$ by radiation modes (photon reservoir) in a three dimensional PBG or FDM. The transition $|1\rangle \rightarrow |0\rangle$ is dipole forbidden. The radiation scattered by the atom consists of Stokes and Rayleigh components with frequencies $\omega_S = \omega_L - \omega_{10}$ and $\omega_R = \omega_L$, respectively. Accordingly, we divide the photon reservoir into two parts, one consisting of the Rayleigh modes (identified by the subscript R) and the other consisting of the Stokes modes (identified by the subscript S).

Our consideration in this chapter is restricted to the case of isotropic photonic band gaps where the band edge is associated with a sphere $|k| = k_0$ in $k$-space (spherical Brillouin zone). Due to the spherical symmetry of the problem, it is convenient to use the spherical harmonic representation for dynamical variables of the system. In this representation, the operators $E(\mathbf{r})$ and $H(\mathbf{r})$ of the electric and magnetic components of the field are expanded in terms of the spherical harmonic vectors\[179, 180]\n
$$E(\mathbf{r}) = \sum_{j=1}^{\infty} \sum_{m=-j}^{m=j} \int_0^\infty \frac{d\omega}{2\pi} \left[ E_{\omega_j}(\mathbf{r}) c_{\omega_j}^m + E_{\omega_j}^*(\mathbf{r}) c_{\omega_j}^{*m} \right]$$ (5.1)
\[
H(r) = \sum_{j=1}^{\infty} \sum_{m=-j}^{m=j} \int_0^{\infty} \frac{d\omega}{2\pi} \left[ H_{\omega j}^m(r) c_{\omega j}^m + H_{\omega j}^{m*}(r) c_{\omega j}^{m*} \right]
\]

(5.2)

where the operators \( c_{\omega j}^m (c_{\omega j}^{m*}) \), obeying the commutation rules,

\[
[c_{\omega j}^m, c_{\omega j'}^{m*}] = 2\pi \delta(\omega - \omega') \delta_{jj'} \delta_{mm'}
\]

(5.3)

annihilate (create) a photon of frequency \( \omega \), angular momentum \( j \), and projection thereof \( m \). The Hamiltonian of the free field is then given by

\[
H_F = \frac{1}{8\pi} \int d^3r \left[ E^2(r) + H^2(r) \right],
\]

(5.4)

and, choosing a coordinate system with the origin at the position of the atom, the operator of atom field coupling has the form (in the dipole approximation)

\[
H_{AF} = -(d_{20} + d_{21}) E(0).
\]

(5.5)

The restriction that photons interact with the transitions under consideration only in the electric dipole approximation reduces the model Hamiltonian of our atom-field system to the following one dimensional form \[179\]

\[
H = \sum_{j=1,2} \omega_{j0} \sigma_{jj} + \sum_{\alpha=R,S} \int_C \frac{d\omega}{2\pi} \omega a_{\alpha}^{\dagger}(\omega) a_{\alpha}(\omega) - \int_C \frac{d\omega}{2\pi} \sqrt{z_{20}(\omega)} [a_R^{\dagger}(\omega) \sigma_{02} + \sigma_{20} a_R(\omega)]
\]

\[
- \int_C \frac{d\omega}{2\pi} \sqrt{z_{21}(\omega)} [a_S^{\dagger}(\omega) \sigma_{12} + \sigma_{21} a_S(\omega)].
\]

(5.6)

The first term on the right-hand side of Eq. (5.6) represents the Hamiltonian of the bare atom\(^1\), with the zero of energy at level \( |0\rangle \) so that \( \omega_0 = 0 \). Here \( \sigma_{ij} = |i\rangle \langle j| \) represent atomic operators satisfying the commutation relation (3.1).

The second term on the right-hand side of Eq. (5.6) represents the Hamiltonian of the photon reservoir which is decomposed into Rayleigh and Stokes modes. Associated with

\[\text{Throughout this chapter we use units in which} \quad \hbar = c = 1.\]

(5.7)
the Rayleigh and Stokes modes are the radiation field creation and annihilation operators, \( a_j^\dagger(\omega) \) and \( a_j(\omega) \), obeying the commutation rules

\[
[a_\alpha(\omega), a_{\alpha'}^\dagger(\omega')] = 2\pi \delta_{\alpha\alpha'} \delta(\omega - \omega').
\]

(5.8)

We assume that the Rayleigh and Stokes components are well separated in frequency so that their corresponding operators commute. We also assume that the atomic operators \( \sigma_{ij} \) commute with the field operators \( a_\alpha(\omega) \) and \( a_{\alpha'}^\dagger(\omega) \).

The last two terms in Eq. (5.6) represent interaction Hamiltonians. The third term describes the interaction between the atom and the Rayleigh part of the photon reservoir, whereas the fourth term describes the interaction between the atom and the Stokes part of the photon reservoir. These interaction Hamiltonians are written in the electric dipole approximation. They are also written in the rotating wave approximation[70] in which virtual processes of excitation (de-excitation) of the atom with simultaneous creation (annihilation) of a photon (i.e., terms of the form \( a_R^\dagger(\omega)\sigma_{20} \) and \( a_R(\omega)\sigma_{02} \) are neglected[see Appendix (A)].

A typical dispersion relation for an isotropic PBG material is shown in the inset of Fig. (5.2). The photon frequency varies from zero to \( \omega_v \) within the lower branch, and from \( \omega_c \) to \(+\infty\) within the upper branch, where \( \omega_v \) and \( \omega_c \) are the lower and upper band edge frequencies, respectively. The gap width is \( \Delta \equiv \omega_c - \omega_v \) and the gap to mid-gap ratio is \( r \equiv \Delta/\omega_o \), where \( \omega_o \) is the mid-gap frequency. We denote by \( G \) the frequency range spanning the gap:

\[
G \equiv (\omega_v, \omega_c).
\]

(5.9)

Within the band gap, there are no propagating photon modes in any direction in space so that the density of photon modes

\[
\rho(\omega) = 0 \quad , \quad \omega \in G.
\]

(5.10)
The integration contours $C_\alpha$ in Eq. (5.6) are subsets of the full contour

$$C = [0, \omega_e] \cup [\omega_c, \infty) = [0, \infty) - G. \quad (5.11)$$

In the Hamiltonian (5.6), the factor $\sqrt{z_{2j}(\omega)}$ represents the frequency dependent coupling constant between the atomic transition $|2\rangle \rightarrow |j\rangle$ and the corresponding photon reservoir. It is given by $z_{2j}(\omega) = \gamma_{2j}(\omega) z(\omega)$, where $\gamma_{2j}(\omega)$ represents the coupling constant of the atomic transition $|2\rangle \rightarrow |j\rangle$ with the free space photon reservoir, and

$$z(\omega) = \left(\frac{d\omega}{dk}\right)^{-1} = \frac{2\pi^2 \rho(\omega)}{k^2(\omega)} \quad (5.12)$$

is the "atomic form factor"[181]. Thus, $z(\omega)$ is proportional to the photon density of states $\rho(\omega)$ and vanishes wherever $\rho(\omega)$ vanishes, such as within a photonic band gap:

$$z(\omega) = 0, \quad \omega \in G. \quad (5.13)$$

In the resonance approximation, we assume that the main contribution to $\gamma_{2j}(\omega)$ comes from the neighborhood of the resonance frequency $\omega_{2j}$, and replace $\gamma_{2j}(\omega)$ by the constant value

$$\gamma_{2j}(\omega) \rightarrow \gamma_{2j} = \frac{4}{3} \omega_{2j}^3 d_{2j}^2, \quad (5.14)$$

where $d_{2j}$ represents the magnitude of the dipole moment for the transition $|2\rangle \rightarrow |j\rangle$. This approximation allows us to extend the limit of integration in Eq. (5.6) to $-\infty$. Thus, in the resonance approximation, we use

$$z_{ij}(\omega) = \gamma_{ij} z(\omega), \quad (5.15)$$

for the coupling constant and

$$C_\infty = (-\infty, \omega_e] \cup [\omega_c, \infty) = (-\infty, \infty) - G, \quad (5.16)$$

for the contour $C$ in Eq. (5.11).
For our model system, the operator for the number of excitations (atom + radiation field), the number operator, is given by

$$N = \sigma_{22} + \sum_{\alpha=R,S} \int \frac{d\omega}{2\pi} a_\alpha^\dagger(\omega)a_\alpha(\omega).$$

(5.17)

This operator commutes with the model Hamiltonian $H$, $[N, H] = 0$. It follows that all eigenstates of $H$ can be classified with respect to the number of excitations or eigenvalues of the operator $N$[156]. Thus, we can separately study the sectors of Hilbert space containing different numbers of excitations. In this chapter we consider the one-excitation sector of the Hilbert space. We look for one-particle eigenstates of the number operator $N$ in the form

$$|\Psi_1\rangle = \left[\xi \sigma_{20} + \int \frac{d\omega}{2\pi} \left[\psi_S(\omega)a_S^\dagger(\omega)\sigma_{10} + \psi_R(\omega)a_R^\dagger(\omega)\right]\right]|\text{vac}\rangle.$$

(5.18)

where $|\text{vac}\rangle$ represents the vacuum state in which the atom is in the ground state $|0\rangle$ and no photon in both reservoirs so that $a_\alpha(\omega)|\text{vac}\rangle = 0$. The amplitude $\xi$ gives the probability of finding the atom in the excited state $|2\rangle$ and both photon reservoirs in the vacuum state. The operator product $a_S^\dagger(\omega)\sigma_{10}$ acting on the vacuum state $|\text{vac}\rangle$ excites the atom to the upper level $|1\rangle$ while creating a Stokes photon of frequency $\omega$, and $\psi_S(\omega)$ gives the amplitude for this process. On the other hand $a_R^\dagger(\omega)$ acting on $|\text{vac}\rangle$ creates a Rayleigh photon of frequency $\omega$ but leaves the atom in the ground state $|0\rangle$, and $\psi_R(\omega)$ gives the amplitude for this process.

### 5.1.2 Equations of Motion

Applying the number operator $N$ to the state $|\Psi_1\rangle$ we obtain $N|\Psi_1\rangle = |\Psi_1\rangle$, which shows that $|\Psi_1\rangle$ is, indeed, a one-particle eigenstate $N$. Now let $|\Psi_1\rangle$ also be an eigenstate of the Hamiltonian (5.6) with the eigenvalue $\epsilon$:

$$H|\Psi_1\rangle = \epsilon|\Psi_1\rangle.$$
Projecting this eigenvalue equation onto \( a_R^\dagger(\omega)|\text{vac}\rangle \), \( a_S^\dagger(\omega)\sigma_{10}|\text{vac}\rangle \) and \( \sigma_{20}|\text{vac}\rangle \), respectively, we obtain the following coupled equations for the amplitudes \( \xi \) and \( \psi_\alpha(\omega) \):

\[
(\omega - \epsilon)\psi_R(\omega|\epsilon) = \sqrt{z_{20}(\omega)}\xi(\epsilon), \tag{5.20}
\]

\[
(\omega - \epsilon + \omega_0)\psi_S(\omega|\epsilon) = \sqrt{z_{21}(\omega)}\xi(\epsilon), \tag{5.21}
\]

\[
(\omega_0 - \epsilon)\xi(\epsilon) = \int_{C_\infty} \frac{d\omega}{2\pi} \left[ \sqrt{z_{20}(\omega)} \psi_R(\omega|\epsilon) + \sqrt{z_{21}(\omega)} \psi_S(\omega|\epsilon) \right]. \tag{5.22}
\]

The general solutions of Eqs. (5.20) and (5.21) are

\[
\psi_R(\omega|\epsilon) = 2\pi \chi_R(\epsilon) \delta(\omega - \epsilon) + \frac{\sqrt{z_{20}(\omega)}}{\omega - \epsilon - i0} \xi(\epsilon), \tag{5.23}
\]

\[
\psi_S(\omega|\epsilon) = 2\pi \chi_S(\epsilon) \delta(\omega - \epsilon + \omega_0) + \frac{\sqrt{z_{21}(\omega)}}{\omega - \epsilon + \omega_0 - i0} \xi(\epsilon), \tag{5.24}
\]

where \( \chi_R(\epsilon) \) and \( \chi_S(\epsilon) \) are as yet undetermined arbitrary functions which can be fixed by specifying asymptotic (boundary) values for \( \psi_R \) and \( \psi_S \). In Eqs. (5.23) and (5.24), the eigenvalue \( \epsilon \) is made slightly complex by adding a small positive imaginary part \( (i0) \) to it. This is a standard procedure in scattering theory\([182]\) to get around the singularities associated with both 1/(\( \omega - \epsilon \)) and 1/(\( \omega - \epsilon + \omega_0 \)).

### 5.2 Discrete and Continuous Solutions

Eqs. (5.23) and (5.24), together with a choice of values for the arbitrary functions \( \chi_R(\epsilon) \) and \( \chi_S(\epsilon) \) [depending on the boundary conditions for \( \psi_R(\omega|\epsilon) \) and \( \psi_S(\omega|\epsilon) \)] completely determine the one-particle spectrum of the system. In this section we investigate the solutions of these equations for discrete bound states and for continuous spectra. We do this first by introducing the so called self-energy function and re-writing Eqs. (5.23) and (5.24) in an auxiliary space which is related to the \( \epsilon \)-space by Fourier transformation.
5.2.1 Self Energy

Substituting Eqs. (5.23) and (5.24) into Eq. (5.22) to eliminate the wave functions $\psi_\alpha(\omega|\epsilon)$, we obtain

\[ [\omega_0 - \epsilon - \Sigma(\epsilon)]\xi(\epsilon) = \sqrt{z_{20}(\epsilon)} \chi_R(\epsilon) + \sqrt{z_{21}(\epsilon - \omega_{10})} \chi_S(\epsilon), \]  

(5.25)

where

\[ \Sigma(\epsilon) = \int_{C_\infty} \frac{d\omega}{2\pi} \left[ \frac{z_{20}(\omega)}{\omega - \epsilon + i0} + \frac{z_{21}(\omega)}{\omega - \epsilon + \omega_{10} - i0} \right] \]  

(5.26)

is referred to as the self energy of the system. Using the identity

\[ \lim_{\eta \to 0} \left( \frac{1}{x \pm i\eta} \right) = P \left( \frac{1}{x} \right) \mp i\pi \delta(x), \]  

(5.27)

where $P$ denotes the Cauchy principal value\[183\], we can decompose the self energy $\Sigma(\epsilon)$ into real and imaginary parts as

\[ \Sigma(\epsilon) = \Sigma'(\epsilon) + i\Sigma''(\epsilon), \]  

(5.28)

where

\[ \Sigma'(\epsilon) = \gamma_{20} P \int_{C_\infty} \frac{d\omega}{2\pi} \frac{z(\omega)}{\omega - \epsilon} + \gamma_{21} P \int_{C_\infty} \frac{d\omega}{2\pi} \frac{z(\omega)}{\omega - \epsilon + \omega_{10}}, \]  

(5.29)

\[ \Sigma''(\epsilon) = \frac{1}{2} [\gamma_{20} z(\epsilon) + \gamma_{21} z(\epsilon - \omega_{10})], \]  

(5.30)

and we have used Eq. (5.15). From Eq. (5.29) we obtain

\[ \frac{d\Sigma'(\epsilon)}{d\epsilon} = \int_C \frac{d\omega}{2\pi} \left[ \frac{\gamma_{20} z(\omega)}{(\omega - \epsilon)^2} + \frac{\gamma_{21} z(\omega)}{(\omega - \epsilon + \omega_{10})^2} \right] > 0, \]  

(5.31)

which indicates that $\Sigma'(\epsilon)$ is a monotonically increasing function of $\epsilon$.

The real part $\Sigma'(\epsilon)$ gives the energy shift of level $|2\rangle$ due to the atom-field interaction\[39\]. Assuming that the main contribution to $\Sigma'(\epsilon)$ comes from the region near the transition
frequency $\omega_{20}$, we can approximate $\Sigma'(\epsilon)$ by its value at $\epsilon = \omega_{20}$,

$$\Sigma'(\epsilon) \approx \Sigma'(\omega_{20}).$$  \hspace{1cm} (5.32)

This value can then be incorporated into the definition of the energy of level $|2\rangle$ to introduce

$$\nu_{20} = \omega_{20} - \Sigma'(\omega_{20}) $$  \hspace{1cm} (5.33)

as the new transition frequency when the atom is in the medium. The first integral on the RHS of Eq. (5.29) is of the order $\gamma_{20}$ except in the neighborhood of $\omega \sim \epsilon$ or in regions where $z(\omega)$ [or, equivalently, the photon density of states $\rho(\omega)$] is very large. On the other hand, the decay rate $\gamma_{20}$ is smaller than the optical transition frequency $\omega_{20}$ by at least six orders of magnitudes ($\gamma_{20} \sim 10^8 \text{s}^{-1}$ whereas $\omega_{20} \sim 10^{15} \text{s}^{-1}$). Thus, whatever the value of the first integral, it cannot offset the smallness of $\gamma_{20}$ and we can safely assume that the first term on the RHS of Eq. (5.29) is very small compared to an optical transition frequency. The same applies for the second term. It follows that $\Sigma'(\omega_{20}) \ll \omega_{20}$ which means that the transition frequency $\nu_{20}$ differs little from the bare value $\omega_{20}$.

The imaginary part $\Sigma''(\epsilon)$ of the self energy gives the decay rate of the upper level $|2\rangle$. In other words, the atom decays from the excited state $|2\rangle$ with a lifetime of $1/\Sigma''(\epsilon)$. From Eqs. (5.12) and (5.30) we see that

$$\Sigma''(\epsilon) \sim \frac{\gamma_{20}}{2} \rho(\epsilon) + \frac{\gamma_{21}}{2} \rho(\epsilon - \omega_{10}).$$  \hspace{1cm} (5.34)

Thus, like the real part $\Sigma'(\epsilon)$, the imaginary part $\Sigma''(\epsilon)$ is proportional to the decay rates $\gamma_{2j}$ and, therefore, is very small except in regions where the local photon density of states is very large. However, unlike $\Sigma'(\epsilon)$, $\Sigma''(\epsilon)$ cannot possibly be neglected since this would lead to infinite lifetime for the excited atomic state $|2\rangle$.

For the electromagnetic vacuum, the dispersion relation is given by $\omega_k = k$ (recall that
we choose units in which \( c = 1 \). For such a dispersion relation, Eqs. (5.12) and (5.15) give

\[
\imath_0^\prime (\omega) = 1, \quad \imath_0^\prime (\omega) = \gamma_{j0}, \quad \rho(\omega) = \omega^2 / 2\pi^2. \tag{5.35}
\]

Moreover, since there is no gap in the free space photon density of states the contour \( C_\infty \) of Eq. (5.16) is given by

\[
C_\infty = (-\infty, \infty). \tag{5.36}
\]

Thus, in the case of vacuum, Eqs. (5.29) and (5.30) give

\[
\Sigma'(\epsilon) = P \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left( \frac{\gamma_{20}}{\omega - \epsilon} + \frac{\gamma_{21}}{\omega - \epsilon + \omega_0} \right) = 0, \tag{5.37}
\]

\[
\Sigma''(\epsilon) = (\gamma_{20} + \gamma_{21})/2. \tag{5.38}
\]

The real part of the free space self energy is identically zero, while the imaginary part is half the sum of the decay rates for the two allowed decay channels \(|2\rangle \rightarrow |0\rangle\) and \(|2\rangle \rightarrow |1\rangle\). The principal part in Eq. (5.37) vanishes because we extended the lower limit of integration to \(-\infty\) by applying the resonance approximation. Had we not done so, the principal part would have given rise to a non-zero contribution (in fact, divergent contribution which has to be corrected by introducing a cut-off\([39, 184]\) on the energy of the photons) which is associated with the Lamb shift of the atomic levels. Even in this case, however, we can set the principal value term to zero by assuming that the Lamb shifts are incorporated into the definition of our state energies.

### 5.2.2 Amplitude Functions in Auxiliary Space

The amplitude functions \( \psi_\alpha(\omega|\epsilon) \) can be cast into a more familiar form by introducing a \( \tau \)-space, which is auxiliary to the \( \omega \)-space. This auxiliary space is defined\([189]\) by the Fourier transforms

\[
a_\alpha(\tau) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega a_\alpha(\omega)e^{i\omega\tau}, \tag{5.39}
\]

\[
\psi_\alpha(\tau|\epsilon) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \psi_\alpha(\omega|\epsilon)e^{i\omega\tau}. \tag{5.40}
\]
The inverse Fourier transforms are given by

\[ a_\alpha(\omega) = \int_{-\infty}^{\infty} d\tau a_\alpha(\tau)e^{-i\omega \tau}, \]

\[ \psi_\alpha(\omega|\epsilon) = \int_{-\infty}^{\infty} d\tau \psi_\alpha(\tau|\epsilon)e^{-i\omega \tau}. \]

Taking the Fourier transforms of the general solutions (5.23) and (5.24) we obtain

\[ \psi_R(\tau|\epsilon) = [\chi_R(\epsilon) + i\sqrt{z_{20}(\epsilon)} \xi(\epsilon)\Theta(\tau)]e^{i\epsilon \tau}, \]

\[ \psi_S(\tau|\epsilon) = [\chi_S(\epsilon) + i\sqrt{z_{21}(\epsilon - \omega_{10})} \xi(\epsilon)\Theta(\tau)]e^{i(\epsilon - \omega_{10}) \tau}, \]

where

\[ \Theta(t) = \begin{cases} 1 & \text{for } t > 0 \\ 0 & \text{for } t < 0. \end{cases} \]

represents the unit step function, and \( \xi(\epsilon) \) is given by Eq. (5.25):

\[ \xi(\epsilon) = \frac{\sqrt{z_{20}(\epsilon)} \chi_R(\epsilon) + \sqrt{z_{21}(\epsilon - \omega_{10})} \chi_S(\epsilon)}{\omega_{20} - \epsilon - \Sigma(\epsilon)}. \]

From Eqs.(5.43) and (5.44) we identify \( i\sqrt{z_{20}(\epsilon)} \xi(\epsilon) \) and \( i\sqrt{z_{21}(\epsilon - \omega_{10})} \xi(\epsilon) \) as the scattering amplitudes for the Rayleigh and Stokes modes, respectively. Thus, the scattering cross-sections - the probability for an incident photon of frequency \( \omega \) to be scattered in the Rayleigh or the Stokes channel - are given by

\[ \sigma_R(\omega) \propto \gamma_{20} z(\omega)|\xi(\omega)|^2, \]

\[ \sigma_S(\omega) \propto \gamma_{21} z(\omega - \omega_{10})|\xi(\omega)|^2, \]

where we have used Eq. (5.15). Noting, according to Eq. (5.12), that \( z(\omega) \) is proportional to \( \rho(\omega) \), we obtain

\[ \sigma_R(\omega) \propto \rho(\omega), \quad \sigma_S(\omega) \propto \rho(\omega - \omega_{10}). \]
5.2.3 Discrete Modes

In this section we look for a discrete mode of the Hamiltonian (5.6) with frequency $\epsilon_d$ such that both $\epsilon_d$ and $\epsilon_d - \omega_{10}$ lie within the frequency gap $G$. This requires that

$$\epsilon_d \in G', \quad (5.50)$$

where

$$G' \equiv (\omega_v + \omega_{10}, \omega_c), \quad (5.51)$$

is a sub-interval of $G = (\omega_v, \omega_c)$. Thus, for both $\epsilon_d$ and $\epsilon_d - \omega_{10}$ to lie within the gap $G$, we must have $\omega_v + \omega_{10} < \omega_c$ or

$$\omega_{10} < \omega_c - \omega_v = \Delta. \quad (5.52)$$

In other words, the transition frequency $\omega_{10} = \omega_{20} - \omega_{21}$ must be less than the width of the gap. When both $\epsilon_d$ and $\epsilon_d - \omega_{10}$ lie within the gap, Eq. (5.13) shows that both $z(\epsilon_d)$ and $z(\epsilon_d - \omega_{10})$ are zero so that, according to Eq. (5.30), the imaginary part $\Sigma''(\epsilon)$ of the self energy is also zero. Thus, for $\epsilon_d \in G'$ the self energy $\Sigma(\epsilon_d)$ is purely real:

$$\Sigma(\epsilon_d) = \Sigma'(\epsilon_d) \quad , \quad \epsilon_d \in G'. \quad (5.53)$$

When both $\epsilon_d$ and $\epsilon_d - \omega_{10}$ lie within the gap, we have $\omega \neq \epsilon_d$ and $\omega \neq \epsilon_d - \omega_{10}$, since single-photon spontaneous emission is completely inhibited within a photonic band gap. Thus, the first terms on the RHS of both Eq. (5.23) and (5.24) do not contribute to $\psi_R(\omega|\epsilon_d)$ and $\psi_S(\omega|\epsilon_d)$, respectively. Therefore, when condition (5.50) is satisfied, we can choose

$$\chi_R(\epsilon_d) = \chi_S(\epsilon_d) = 0 \quad , \quad \epsilon_d \in G'. \quad (5.54)$$

This choice can then be used in Eqs. (5.23), (5.24), and (5.25) to obtain

$$\psi_R(\omega|\epsilon_d) = \frac{\sqrt{z_{20}(\omega)}}{\omega - \epsilon_d} \xi(\epsilon_d) \quad , \quad \psi_S(\omega|\epsilon_d) = \frac{\sqrt{z_{21}(\omega)}}{\omega - \epsilon_d + \omega_{10}} \xi(\epsilon_d), \quad (5.55)$$
and

$$[\omega_2 - \epsilon_d - \Sigma'(\epsilon_d)]\xi(\epsilon_d) = 0. \quad (5.56)$$

In Eq. (5.56) we have used Eq. (5.53) to replace \(\Sigma(\epsilon_d)\) by the real part \(\Sigma'(\epsilon_d)\).

Apart from the trivial solution \(\xi(\epsilon_d) = 0\), Eq. (5.56) has a nontrivial solution \(\xi(\epsilon_d) \neq 0\) provided that an eigenenergy \(\epsilon_d\) is found such that

$$\omega_2 - \epsilon_d - \Sigma'(\epsilon_d) = 0, \quad \epsilon_d \in G'. \quad (5.57)$$

Since \(\Sigma'(\epsilon)\) is a monotonically increasing function of \(\epsilon\) [see Eq. (5.31)], Eq. (5.57) can have only one root \(\epsilon = \epsilon_d\) given by the intersection of the curve \(\Sigma'(\epsilon)\) with the straight line \(\omega_2 - \epsilon\). Clearly, a discrete bound state occurs only if the straight line \(y(\epsilon) = \omega_2 - \epsilon\) intersects the curve \(\Sigma''(\epsilon)\) within the frequency range \(G'\) where, according to Eq. (5.53), \(\Sigma''(\epsilon) = 0\). Substituting Eq. (5.55) into Eq. (5.18), we find this discrete eigenstate of the system to be

$$|\Psi_d\rangle = \xi(\epsilon_d) \left[ \sigma_20 + \int_{C_{\infty}} \frac{d\omega}{2\pi} \left( \frac{\sqrt{z_{21}(\omega)}}{\omega - \epsilon_d + \omega_{10}} a_S^\dagger(\omega) \sigma_{10} + \frac{\sqrt{z_{20}(\omega)}}{\omega - \epsilon_d} a_R^\dagger(\omega) \right) \right] |vac\rangle. \quad (5.58)$$

The function \(\xi(\epsilon_d)\) is determined from the normalization condition \(\langle \Psi_d | \Psi_d \rangle = 1\). We obtain

$$|\xi(\epsilon_d)|^2 = \left[ 1 + \gamma_20 \int_{C_{\infty}} \frac{d\omega}{2\pi} \frac{z(\omega)}{(\omega - \epsilon_d)^2} + \gamma_21 \int_{C_{\infty}} \frac{d\omega}{2\pi} \frac{z(\omega)}{(\omega - \epsilon_d + \omega_{10})^2} \right]^{-1}. \quad (5.59)$$

Since both \(\epsilon_d\) and \(\epsilon_d - \omega_{10}\) lie within the band gap \(G\) they do not lie on the contour of integration \(C_{\infty}\) given by Eq. (5.16). Thus the integrals in Eq. (5.59) are not principal value integrals, and therefore can be approximated by the values \(\gamma_20\) and \(\gamma_21\). It follows that, when the system (atom + field) is in the discrete bound state \(|\Psi_d\rangle\), the probability to find the atom in the excited state \(|2\rangle\) is given approximately by

$$P_2 \equiv |\xi(\epsilon_d)|^2 \approx (1 + \gamma_20 + \gamma_21)^{-1} \approx 1, \quad (5.60)$$

since \(\gamma_{2j} \ll 1\). That the probability \(P_2\) is close to unity means that, when the system is in
the photon-atom bound state $|\psi_d\rangle$, the atom spends most of its time on the excited state $|2\rangle$. Once in a while, the atom drops to the lower state $|1\rangle$ by emitting a Stokes photon or to the ground state $|0\rangle$ by emitting a Rayleigh photon. However, the emitted photon is quickly re-absorbed by the atom which is then re-excited to level $|2\rangle$.

The discrete mode (5.58) describes the three-level atom analog of the photon-atom bound state predicted by John and Wang[5] for a hydrogenic atom. In the present case there are two scattering channels, the elastic (Rayleigh) and the inelastic (Stokes or anti-Stokes) channels. A photon of frequency $\omega$ incident on an unexcited atom (atom on level $|0\rangle$) can be scattered in the Rayleigh channel preserving its frequency or in the Stokes channel, in which the atom is excited to level $|1\rangle$ and a Stokes photon of frequency $\omega_S = \omega - \omega_{10}$ is created. Alternately, the photon can be scattered from an excited atom on level $|1\rangle$ accompanied by the creation of an anti-Stokes photon of frequency $\omega_{AS} = \omega + \omega_{10}$ and the de-excitation of the atom to the ground level $|0\rangle$. Thus, in the three-level atom case, the radiation field is bound to the atom as a result of two-channel scattering, while the atom switches between levels $|0\rangle$ and $|1\rangle$ via the intermediate state $|2\rangle$.

According to Eq. (5.33) the solution of Eq. (5.57) can be written in terms of the shifted atomic transition frequency $\nu_{20}$ as $\epsilon_d = \nu_{20}$. There will be a discrete bound state only if $\nu_{20}$ (or roughly $\omega_{20}$) lies in the frequency range between $\omega_v + \omega_{10}$ and $\omega_c$. Moreover, condition (5.50) for the occurrence of a discrete bound state requires that the transition frequency $\omega_{10} = \omega_{20} - \omega_{21}$ be less than the width $\Delta = \omega_c - \omega_v$ of the gap. Thus for a discrete bound state to occur we must have $\omega_{21} \in G$ and $\omega_{20} \in G'$, where the frequency intervals $G$ and $G'$ are given by Eqs. (5.9) and (5.51), respectively. If, for instance, $\omega_{21} \not\in G$, the bound state becomes quasi-bound. Its lifetime is determined by the possible irradiation of a photon of frequency $\omega_{21}$ in resonance with the escape channel $|2\rangle \rightarrow |1\rangle$.

### 5.2.4 Continuous Spectrum

In this section we consider the case when the eigenvalue $\epsilon$ of Eq. (5.19) does not satisfy the discrete bound state condition (5.50). This requires that $\epsilon \not\in G'$. We investigate the
scattering of a single incident laser photon of frequency $\omega$ from the three-level atom initially in the ground state. This initial condition (atom on level $|0\rangle$, a laser photon and no Stokes photon) determines the asymptotic (large distance) values of the amplitudes $\psi_R$ and $\psi_S$ uniquely and therefore fixes the functions $\chi_R(\epsilon)$ and $\chi_S(\epsilon)$ of Eqs. (5.23) and (5.24) to

$$\chi_R(\epsilon) = 1 \quad , \quad \chi_S(\epsilon) = 0.$$  (5.61)

Using this choice in Eq. (5.46), and substituting the result in Eqs. (5.47) and (5.48) we obtain

$$\sigma_R(\omega) \propto \frac{\gamma_{20}^2 z^2(\omega)}{[\omega_{20} - \omega - \Sigma'(\omega)]^2 + [\Sigma''(\omega)]^2},$$  (5.62)

$$\sigma_S(\omega) \propto \frac{\gamma_{20}\gamma_{21} z(\omega - \omega_{10})}{[\omega_{20} - \omega - \Sigma'(\omega)]^2 + [\Sigma''(\omega)]^2},$$  (5.63)

where $\Sigma'(\omega)$ and $\Sigma''(\omega)$ are the real and imaginary parts of the self-energy given, respectively, by Eqs. (5.29) and (5.30). Thus, the scattering cross sections $\sigma_R(\omega)$ and $\sigma_S(\omega)$ are strongly dependent on the atomic form factor $z(\omega)$ and on the self energy $\Sigma(\omega)$. Eqs. (5.62) and (5.63) are written as functions of the incident laser photon frequency $\omega$. The frequency of the scattered Rayleigh photons is $\omega_R = \omega$, whereas that of the Stokes photons is $\omega_S = \omega - \omega_{10}$.

In free space, the self energies $\Sigma'(\omega)$ and $\Sigma''(\omega)$ are given by Eqs. (5.37) and (5.38), respectively. Using them in Eqs. (5.62) and (5.63) we obtain

$$\sigma_R(\omega) \propto \frac{\gamma_{20}^2}{(\omega_{20} - \omega)^2 + (\gamma_{20} + \gamma_{21})^2/4},$$  (5.64)

$$\sigma_S(\omega_S) \propto \frac{\gamma_{20}\gamma_{21}}{(\omega_{21} - \omega_S)^2 + (\gamma_{20} + \gamma_{21})^2/4},$$  (5.65)

where we used the relation $\omega_{21} = \omega_{20} - \omega_{10}$ to write $\sigma_S(\omega_S)$ as a function of the Stokes frequency $\omega_S = \omega - \omega_{10}$. Eqs. (5.64) and (5.65) represent two Lorentzian distributions of full width at half maximum $\Gamma = \gamma_{20} + \gamma_{21}$ peaked at the two transition frequencies $\omega_{20}$ and $\omega_{21}$. The line width $\Gamma$ is determined by the total life-time of level $|2\rangle$ which can decay via
either \( |2\rangle \rightarrow |0\rangle \) (with decay rate \( \gamma_{20} \)) or \( |2\rangle \rightarrow |1\rangle \) (with decay rate \( \gamma_{21} \)).

In a reservoir with a modified density of photon modes, the spectra of both the Rayleigh and the Stokes components of the scattered light will depart from a Lorentzian distribution to the extent that the form factors \( z(\omega) \) and the self energy \( \Sigma(\omega) \) in the medium deviate from their corresponding free space forms. In analyzing the continuous spectrum in such modified reservoirs, we assume that the Rayleigh transition frequency \( \omega_{20} \) lies far above the upper band-edge \( \omega_c \), and we consider cases when the Stokes transition frequency \( \omega_{21} \) lies in the neighborhood of \( \omega_c \). Thus the Rayleigh transition \( |2\rangle \rightarrow |0\rangle \) can be considered to be occurring in a normal free space (Markovian reservoir) whereas the Stokes transition \( |2\rangle \rightarrow |1\rangle \) occurs in a modified non-Markovian reservoir. Our main interest is to investigate effects on the Rayleigh spectrum due to the coupling of the Stokes transition \( |2\rangle \rightarrow |1\rangle \) to the modified reservoir.

We decompose the self energy (5.26) into Rayleigh and Stokes components as

\[
\Sigma(\epsilon) = \Sigma_R(\epsilon) + \Sigma_S(\epsilon),
\]

where

\[
\Sigma_R(\epsilon) = \gamma_{20} \int_{C_{\infty}} \frac{dw}{2\pi} \frac{z(\omega)}{\omega - \epsilon - i0};
\]

\[
\Sigma_S(\epsilon) = \gamma_{21} \int_{C_{\infty}} \frac{dw}{2\pi} \frac{z(\omega)}{\omega - \epsilon + \omega_{21} - i0}.
\]

When \( \omega_{20} \) lies far above the gap, the real and imaginary parts of \( \Sigma_R(\epsilon) \) can be approximated by the free space values \( \Sigma'_R(\epsilon) = 0 \) and \( \Sigma''_R(\epsilon) = i\gamma_{20}/2 \). On the other hand, the real and imaginary parts of \( \Sigma_S(\epsilon) \) are given by

\[
\Sigma'_S(\epsilon) = \gamma_{21} P \int_{C_{\infty}} \frac{dw}{2\pi} \frac{z(\omega)}{\omega - \epsilon + \omega_{21}},
\]

\[
\Sigma''_S(\epsilon) = \frac{\gamma_{21}}{2} z(\epsilon - \omega_{10}).
\]

Thus, when \( \omega_{20} \) is far above \( \omega_c \), and when \( \omega_{21} \) is in the neighborhood of \( \omega_c \), the real and
imaginary parts of the the total self energy (5.26) are given approximately by

\begin{align}
\Sigma'(\epsilon) &= \Sigma'_S(\epsilon), \\
\Sigma''(\epsilon) &= \gamma_{20}/2 + \Sigma''_S(\epsilon).
\end{align}

Using these in Eq. (5.63) we finally obtain

\[ \sigma_R(\omega) \propto \frac{\gamma_{20}^2 \omega^2(\omega)}{[\omega_{20} - \omega - \Sigma'_S(\omega)]^2 + [\gamma_{20} + 2\Sigma''_S(\omega)]^2/4}, \]

for the scattering cross-section of the Rayleigh component of the scattered light. We investigate specific features of this cross-section for the case of PBG and FDM separately in the following sections.

### 5.3 System in a PBG material

In a PBG one finds a modified dispersion relation for the photons in the radiation reservoir with a gap(s) in the photon density of states. A simple model dispersion relation which exhibits a gap in the photon density of states is given by [185]

\[ \omega(k) = \sqrt{k_0^2 + \sigma^2} \pm \sqrt{(k - k_0)^2 + \sigma^2}, \quad (5.74) \]

where \( k = |k| \) is the modulus of the wave vector, and \( k_0 \) and \( \sigma \) are parameters related to the periodic dielectric structure. The + (−) applies for the upper (lower) branch of the photon spectrum. Physically model (5.74) corresponds to placing an isotropic band gap of the form shown in the inset in Fig. 5.2 whose center and band edge frequencies are given by

\[ \omega_o = \sqrt{k_0^2 + \sigma^2}, \quad \omega_u = \omega_o - \sigma, \quad \omega_c = \omega_o + \sigma. \quad (5.75) \]

The width \( \Delta \) of the photonic band gap, and its gap to midgap ratio \( r \) are then

\[ \Delta \equiv \omega_c - \omega_u = 2\sigma, \quad r \equiv \Delta/\omega_o = 2\sigma/\sqrt{k_0^2 + \sigma^2}. \quad (5.76) \]
For the isotropic dispersion relation (5.74), the atomic form factor $z(\omega)$ takes the form [see Appendix (F)]

$$z(\omega) = \begin{cases} \frac{|\omega-\omega_p|}{\sqrt{(\omega-\omega_v)(\omega-\omega_c)}}, & \omega \notin G, \\ 0, & \omega \in G, \end{cases}$$  \hspace{1cm} (5.77)

which, when used in Eq. (5.12), gives

$$\rho(\omega) = \begin{cases} \frac{|\omega-\omega_p|}{2\pi^2} \left[ \frac{\omega^2-2\omega_v\omega+2\omega_v\omega_c}{\sqrt{(\omega-\omega_v)(\omega-\omega_c)}} \pm 2\sqrt{\omega_v\omega_c} \right], & \omega \notin G, \\ 0, & \omega \in G, \end{cases}$$  \hspace{1cm} (5.78)

for the density of photon modes. Here the $+(-)$ sign applies for $\omega > \omega_c$ ($\omega < \omega_v$).

The density of states (5.78) is plotted in Fig. 5.2. For regions far removed from the band edges, $\rho(\omega)$ differs little from its free-space value of $\omega^2/2\pi^2$, as expected. Near the band edges $\omega_v$ and $\omega_c$, Eq. (5.78) shows that $\rho(\omega)$ behaves as $\rho(\omega) \sim (\omega_v - \omega)^{-1/2}$ for $\omega < \omega_v$ and as $\rho(\omega) \sim (\omega - \omega_c)^{-1/2}$ for $\omega > \omega_c$, the square-root singularities being characteristic of a one-dimensional phase space[5]. These singularities are artifacts of the isotropic dispersion relation (5.74) which associates the band edge wave vector with a sphere in $k$ space, $|k| = k_0$ (spherical Brillouin-zone) and thereby artificially increases the true phase space available for photon propagation near the band edge.

Using Eqs. (5.77) and Eq. (5.15) in Eq. (5.30), we obtain the imaginary part $\Sigma''(\epsilon)$ of the self energy. Likewise the real part $\Sigma'(\epsilon)$ is determined by using Eqs. (5.15) and (5.77) in Eq. (5.29) and evaluating (numerically) the integral. For illustration, we consider an isotropic PBG with a gap to mid-gap ratio of $r = 15\%$, a ratio which can be achieved in practical PBG materials[12]. Thus, in units of the mid-gap frequency $\omega_o$, the band width is $\Delta = 0.15$, and the band edge frequencies are at $\omega_v = 1-r/2 = 0.925$ and $\omega_c = 1+r/2 = 1.075$. In what follows, we fix the transition frequency $\omega_{10}$ to $\omega_{10} = \Delta/10 = 0.015\omega_o$. The frequency intervals $G$ and $G'$ are thus given by $G = (0.925, 1.075)$ and $G' = (0.94, 1.075)$. Also we choose $\gamma_{20} = \gamma_{21} = \omega_{10}/10$ for visualization purposes. In reality, for $\omega_{10}$ in the optical regime, $\gamma_{20} \ll \omega_{10}$. However, as seen in Eqs. (5.30) and (5.29), the constants $\gamma_{2j}$ appear only as pre-factors in the expressions for $\Sigma'(\epsilon)$ and $\Sigma''(\epsilon)$, and, therefore, their only effect
Figure 5.2: The photon density of states $\rho(\omega)$ in a PBG material described by the isotropic dispersion relation (5.74). The dashed curve represents the free space density of photon states, $\rho(\omega) = \omega^2/2\pi^2$. For isotropic PBG, $\rho(\omega)$ exhibits singularities at both band edges behaving like $\rho(\omega) \sim (\omega - \omega_{\text{v}})^{-1/2}$ for $\omega < \omega_{\text{v}}$, and like $\rho(\omega) \sim (\omega - \omega_{\text{c}})^{-1/2}$ for $\omega > \omega_{\text{c}}$. Far from the band-edges, $\rho(\omega)$ is essentially the same as that of free space, as expected. A typical isotropic dispersion relation for a PBG material is shown in the inset.

is to change the scale of plots.

In Fig. 5.3 we plot the real part $\Sigma'(\epsilon)$ (solid curve) and the imaginary part $\Sigma''(\epsilon)$ (dashed curve) of the self energy $\Sigma(\epsilon)$ as functions of the scaled frequency $\epsilon/\omega_0$ for the case when both $\omega_{z0}$ and $\omega_{z1}$ lie inside the gap. The dotted curve represents the imaginary part of the free space self energy given by Eq. (5.38). The real part of the free space self energy is identically zero [see Eq. (5.37)]. According to Eq. (5.53), $\Sigma''(\epsilon) = 0$ for $\epsilon \in G' = (0.94, 1.075)$. Near the band edges $\omega_{\text{v}}$ and $\omega_{\text{c}}$, $\Sigma''(\epsilon)$ grows steadily from its free space value of unity, exhibiting singularities at $\omega_{\text{v}} = 0.925$, $\omega_{\text{v}} + \omega_{10} = 0.94$, $\omega_{\text{c}} = 1.075$, and $\omega_{\text{c}} + \omega_{10} = 1.09$. The singularities at the band edge frequencies $\omega_{\text{v}}$ and $\omega_{\text{c}}$ are due to the term containing $z(\epsilon)$ [see Eqs. (5.30) and (5.77)], whereas the singularities at the shifted frequencies $\omega_{\text{v}} + \omega_{10}$ and $\omega_{\text{c}} + \omega_{10}$ are due to the term containing $z(\epsilon - \omega_{10})$. Likewise, the real part $\Sigma'(\epsilon)$ of the self energy is singular at the frequencies $\omega_{\text{v}}$, $\omega_{\text{v}} + \omega_{10}$, $\omega_{\text{c}}$ and $\omega_{\text{c}} + \omega_{10}$. In the frequency range $\epsilon \in G'$, $\Sigma'(\epsilon)$ is a monotonically increasing function of $\epsilon$. It is negative for $\epsilon < \epsilon_0$ and positive for $\epsilon > \epsilon_0$, where $\epsilon_0$ denotes the zero of $\Sigma'(\epsilon)$. Far from the band gap, $\Sigma'(\epsilon)$ and $\Sigma''(\epsilon)$ are practically indistinguishable from their corresponding free space values, as
Figure 5.3: The real part $\Sigma'(\epsilon)$ (solid curve) and the imaginary part $\Sigma''(\epsilon)$ (long-dashed curve) of the self energy [Eqs. (5.29) and (5.30)] for a PBG material as functions of the scaled frequency $\epsilon/\omega_0$. Here, $r = 15\%$, $\omega_v = 0.925\omega_0$, $\omega_c = 1.075\omega_0$, and the band gap width is $\Delta = 0.15\omega_0$. Also $\omega_{10} = \Delta/10 = 0.015\omega_0$ and $\omega_{20} = 0.95\omega_0 = 1.012\omega_0$ so that $\omega_{20} \in G'$ and $\omega_{21} = 1.006\omega_0 \in G$. We also take $\gamma_{20} = \gamma_{21} = \Delta/100$. In the frequency range $G'$ the imaginary part of the self energy is identically zero whereas the real part is a monotonically increasing function of frequency. Both the real and imaginary parts of the self energy exhibit singularities at the frequencies $\omega_v$, $\omega_v + \omega_{10}$, $\omega_c$ and $\omega_c + \omega_{10}$. The dotted curve represents the imaginary part of the free space self energy given by $(\gamma_{20} + \gamma_{10}/2)$. The real part of the free space self energy is identically zero [see Eq. (5.38)]. The dot-dashed curve represents the straight line $y(\epsilon) = \omega_{20} - \epsilon$. This line intersects $\Sigma'(\epsilon)$ at $\epsilon \approx 1.02\omega_0 \in G'$.

5.3.1 Discrete Modes in a PBG

In this section we investigate the photon-atom bound state in a PBG described by the isotropic dispersion relation (5.74). As discussed in Sec.5.2.3, a discrete bound state can occur only when $\omega_{20} \in G'$ and $\omega_{21} \in G$, which, in turn, require $\omega_{10}$ to satisfy condition (5.52). The discrete bound state is given by the intersection of the straight line $y(\epsilon) = \omega_{20} - \epsilon$ with the curve $\Sigma'(\epsilon)$, provided that this intersection lies within the frequency range $G'$ where $\Sigma''(\epsilon) = 0$.

Using the same $\omega_{10}$ and PBG parameters as in section (5.3), we investigate the occurrence of a discrete bound state when $\omega_{20}$ (and hence $\omega_{21}$) is varied with respect to the upper band edge $\omega_c$.

In Fig. 5.3 we plot the curves $\Sigma'(\epsilon)$ and $y(\epsilon) = \omega_{20} - \epsilon$, as functions of the scaled frequency.
for the case when \( \omega_0 \) lies just below the upper band edge \((\omega_0 = 0.95 \omega_c = 1.021)\) so that \( \omega_0 \in G' \) and \( \omega_2 = 1.006 \in G \). In this case we see that the straight line \( y(\epsilon) = \omega_0 - \epsilon \) intersects the curves \( \Sigma'(\epsilon) \) at \( \epsilon \approx 1.02 \omega_0 \) which lies within the frequency interval \( G' \). This intersection point then represents the eigenvalue of a non-decaying photon-atom bound state \(|\Psi_d\rangle\) given by Eq. (5.58) with \( \epsilon_d \approx 1.02 \). As \( \omega_0 \) (and hence \( \omega_2 \)) is pushed further towards the upper band edge \( \omega_c \), the bound state is also pushed towards \( \omega_c \). When \( \omega_0 \) lies outside the gap, the straight line \( y(\epsilon) \) no longer intersects the real part \( \Sigma'(\epsilon) \) of the self energy within the frequency range \( G' \) where \( \Sigma''(\epsilon) = 0 \). This means that the solution of \( \omega_0 - \epsilon - \Sigma(\epsilon) = 0 \) has a non-zero imaginary part which serves to damp the system. Thus any initial population on the excited levels \(|2\rangle\) and \(|1\rangle\) will eventually decay to the ground level \(|0\rangle\), just as in free-space. However, unlike the free-space case, the decay may be oscillatory instead of simple exponential[65].

5.3.2 Continuous Spectrum in PBG

We now analyze the spectrum equation (5.73) in the case of the PBG. In what follows we fix \( \omega_0 \) to a value far above \( \omega_c \) and plot the Rayleigh spectrum \( \sigma_R(\omega) \) versus \( \omega - \omega_0 \) for various values of the Stokes transition frequency \( \omega_2 \).

From Eqs. (5.13) and (5.73) we see that \( \sigma_R(\omega) = 0 \) for \( \omega \in G \). Thus in all plots of \( \sigma_R(\omega) \) versus \( \omega - \omega_0 \), the Rayleigh spectrum will vanish completely in the "spectral gap" region \( \omega - \omega_0 \in G_s \equiv (\omega_v - \omega_0, \omega_c - \omega_0) \). From Eqs. (5.70) and (5.77) we observe that the imaginary part \( \Sigma''(\omega) \) of the self energy exhibits square root singularities at \( \omega = \omega_v + \omega_1 \) and \( \omega = \omega_c + \omega_1 \) leading to "dark lines" at \( \omega - \omega_0 = \omega_v - \omega_2 \) and \( \omega - \omega_0 = \omega_c - \omega_2 \) in \( \sigma_R(\omega) \). These dark lines represent complete quenching of spontaneous emission at the respective frequencies and are separated exactly by the width \( \Delta = \omega_c - \omega_v \) of the gap. They are shifted to higher frequencies when \( \omega_2 \) is decreased.

The dark lines here are artifacts of the isotopic dispersion relation (5.74) which lead to the atomic form factor \( z(\omega) \) being singular at the band edges \( \omega_v \) and \( \omega_c \) as shown in Eq. (5.77). In real photonic crystals, the dispersion relation is anisotropic and \( z(\omega) \) is
not singular at the band edges[67]. Moreover, in realistic situations, relaxation processes in the medium should be taken into consideration[5, 67]. One way to do this is to rewrite Eq. (5.77) in the form

$$ z(\omega) = \begin{cases} \frac{|\omega - \omega_0|}{\sqrt{(\omega - \omega_v)(\omega - \omega_c) + \kappa^2}}, & \omega \not\in G \\ 0, & \omega \in G, \end{cases} \quad (5.79) $$

where $\kappa$ is a constant characteristic of the medium, introduced phenomenologically to account for relaxation processes. Thus in realistic situations, the Rayleigh spectrum will exhibit significant suppressions at $\omega = \omega_v + \omega_{10}$ and $\omega = \omega_c + \omega_{10}$ but not dark lines.

The dark lines at $\omega = \omega_v + \omega_{10}$ and $\omega = \omega_c + \omega_{10}$ split $\sigma_R(\omega)$ into triplets. These are “band A” which lies to the left of $\omega_v + \omega_{10}$, “band B” which lies between $\omega = \omega_v + \omega_{10}$ and $\omega = \omega_c + \omega_{10}$, and “band C” which lies to the right of $\omega_c + \omega_{10}$. Band B measures the fractionalized steady-state atomic population on the excited state $|2\rangle$ due to the presence of the photonic band gap[65] and, therefore, can be used to experimentally probe this steady-state population. On the other hand, bands A and C measure the fraction of the excited state population $|2\rangle$ that decays to the lower levels $|1\rangle$ and $|0\rangle$.

Spectral splitting was derived by John and Wang[5] in the effective mass approximation to the dispersion relation (5.74) near the upper band edge $\omega_c$, totally disregarding the effect of the lower band edge $\omega_v$. One dark line and splitting into a doublet (bands B and C) was reported in the absence of an external field (vacuum Rabi splitting). The splitting was caused entirely by strong interaction between the atom and its own radiation field. More recently, dark lines have also been reported in Refs.[163, 186].

For illustration, we consider an isotropic PBG with a gap to mid-gap ratio of $r = 15\%$. In units of the mid-gap frequency $\omega_0$, the band width is $\Delta = .15$ and the band edge frequencies are at $\omega_v = 1 - \frac{r}{2} = 0.925$ and $\omega_c = 1 + \frac{r}{2} = 1.075$ so that $G = (0.925, 1.075)$. Also we choose $\gamma_{20} = \gamma_{21} = 0.01\omega_20$ for the decay constants. We fix the Rayleigh transition frequency to $\omega_20 = 1.2\omega_0 = 1.29$ so that $\omega_20$ is far from the band edge by more than the width $\Delta$ of the gap. Having fixed $\omega_20$, we plot the Rayleigh spectrum $\sigma_R(\omega)$ versus $\omega - \omega_20$. 

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for various values of the Stokes transition frequency \( \omega_{21} \) in the neighborhood of the upper band edge \( \omega_e \). In all of these plots \( \sigma_R(\omega) = 0 \) in the region \( G_s = (-0.340, -0.215) \).

We are mainly interested in the behavior of \( \sigma_R(\omega) \) in the neighborhood of the resonance \( \omega - \omega_{20} = 0 \). In regions far from resonance, the value of \( \sigma_R(\omega) \) is negligibly small compared with its values in the resonance region. For instance, for our choice \( \omega_{20} = 1.2\omega_e \), the spectral gap \( G_s = (-0.340, -0.215) \) lies far to the left of the resonance region \( \omega - \omega_{20} = 0 \) and is not shown in the plots.

First we consider the case when \( \omega_{21} \) also lies far above the upper band edge. We take \( \omega_{21} = 1.15\omega_e = 1.236 \) so that \( \omega_{21} \) is removed from the upper band edge \( \omega_e \) by the width \( \Delta \) of the gap. We then have \( \omega_{10} = \omega_{20} - \omega_{21} = 0.054 \) and \( \gamma_{20} = \gamma_{21} = \omega_{10}/10 = 0.0054 \). Thus the dark lines of \( \sigma_R(\omega) \) occur at \( \omega = \omega_v + \omega_{10} = 0.979 \) and \( \omega = \omega_e + \omega_{10} = 1.129 \) or at \( \omega - \omega_{20} = -0.311 \) and \( \omega - \omega_{20} = -0.161 \). The left dark line lies within the spectral gap \( G_s = (-0.340, -0.215) \). The spectrum corresponding to this choice of \( \omega_{21} \) is shown by the dot-dashed curve in Fig. (5.4). The left and right dark lines occur far to the left of the resonance region \( \omega - \omega_{20} = 0 \) and are not shown in the plot. From the plot we see that, for \( \omega_{21} = 1.15\omega_e \), \( \sigma_R(\omega) \) practically consists only of band C, implying that all the population on the excited level \( |2\rangle \) eventually decays to the lower levels \( |1\rangle \) and \( |0\rangle \). The plot also shows that band C is a Lorentzian with full width at half maximum \( \Gamma = \gamma_{20} + \gamma_{10} = 0.011 \). On the scale of this figure, the Rayleigh spectrum derived from Eq. (5.73) is indistinguishable from the free space spectrum (5.64).

The effect of the band gap is negligible for transition frequencies removed from a band edge by at least the width \( \Delta \) of the gap. This can be explained as follows. When \( \omega_{21} \) lies far above the band edge \( \omega_e \), we can apply the Wigner-Weisskopf (WW) approximation for the Stokes channel and write \( \Sigma'_S = 0, \Sigma''_S = \gamma_{21}/2 \). Since \( z(\omega) = 1 \), Eq. (5.73) reduces to the free space equation (5.64). Thus when both transition frequencies \( \omega_{20} \) and \( \omega_{21} \) lie far above the upper band edge \( \omega_e \), we expect the Rayleigh line to be a Lorentzian of full width at half maximum \( \Gamma = \gamma_{20} + \gamma_{10} \) centered at the frequency \( \omega = \omega_{20} \), just as in free space. On the other hand, when \( \omega_{21} \) is near the upper band edge, the density of electromagnetic
Figure 5.4: The spectrum of spontaneous emission $\sigma_R(\omega)$ for the Rayleigh transition $|2\rangle \rightarrow |0\rangle$ in the case of the PBG described by the isotropic dispersion relation (5.74). We take $r = 15\%$, $\omega_{20} = 1.2\omega_c$, and $\gamma_{20} = \gamma_{21} = 0.01\omega_{20}$. Thus $\omega_{20}$ is removed from the band edge $\omega_c$ by more than the width of the gap. We plot $\sigma_R(\omega)$ for different values of the Stokes frequency $\omega_{21}$. The dot-dashed curve represents both equations (5.73) and (5.64) when $\omega_{21} = 1.15\omega_c$. Thus the effect of the gap is negligible for transition frequencies removed from the band edge by at least the width $\Delta$ of the gap.

modes changes rapidly in the vicinity of the atomic transition frequency $\omega_{21}$, rendering the WW approximation to the self energy $\Sigma_S(\epsilon)$ inadequate. In this case, we must perform an exact integration in Eq. (5.69) which is then used in Eq. (5.73) to evaluate the spectrum $\sigma_R(\omega)$.

Next we consider the case when $\omega_{21}$ is close to the upper band edge $\omega_c$. We take $\omega_{21} = 1.05\omega_c = 1.129$ so that $\omega_{10} = 0.161$. Thus the left and right dark lines occur at $\omega - \omega_{20} = -0.204$ and $\omega - \omega_{20} = -0.054$, respectively. The spectrum for this case is shown by the solid curve in Fig. (5.4). The left dark line occurs far to the left of the resonance region $\omega - \omega_{20} = 0$ and is not shown in the plot. For $\omega_{21} = 1.05\omega_c$, $\sigma_R(\omega)$ practically consists only of two bands, bands B and C. The existence of band B shows that there is a fractionalized steady-state population on the excited level $|2\rangle$ even though $\omega_{20}$ lies outside the gap. The line width of band B is much smaller than the natural line width $\gamma_{20}$ of the spontaneous transition $|2\rangle \rightarrow |0\rangle$ whereas the line width of band C is roughly the same as that in free space. When $\omega_{21} = 1.05\omega_c$ most of the initial population on $|2\rangle$ decays to the lower levels and only a very small part is retained as a fractionalized steady-state
population. As $\omega_{21}$ is pushed further towards $\omega_c$, the dark lines are shifted to the right and the width of the band B increases, implying that more and more of the initial population is now retained as a fractionalized steady-state population. The dotted curve in Fig. 5.4 represents the spectrum when $\omega_{21}$ coincides with the band edge, $\omega_c$. In this case the left dark line occurs at $\omega - \omega_{20} = -0.15$ and is not shown in the figure, whereas the right dark line occurs at the resonance $\omega = \omega_{20}$.

Finally we consider the case when $\omega_{21}$ lies inside the gap. We take $\omega_{21} = \omega_o = 1$, the midgap. Then $\omega_{10} = 0.29$ and the left and right dark lines occur at $\omega - \omega_{20} = -0.075$ and $\omega - \omega_{20} = -0.075$, respectively, equidistant from the resonance frequency $\omega = \omega_{20}$. The spectrum for this case is shown by the dashed curve in Fig. (5.4). From the plot we see that by far the dominant contribution to $\sigma_R(\omega)$ comes from band B, implying that for $\omega_{21} = \omega_o$ most of the initial population on $|2\rangle$ is retained as a fractionalized steady-state population, and only a very small part decays to the lower levels.

5.4 System in a Dispersive Medium

In this section we briefly discuss the case when the three-level atom is embedded within a frequency dispersive medium (FDM). It is well known that a frequency gap for propagating electromagnetic modes exists in many natural dielectrics and semiconductors, such as NaCl, KCl, GaAs, GaP, InP and InAs. Unlike in artificial PBG materials, where a suppression of the photon density of states over a narrow frequency range results from multiple photon scattering by a periodic array of scatterers, the gaps in dispersive media are caused by photon coupling to elementary excitations of the media such as optical phonons[187, 188].

The AC electric field associated with electromagnetic radiation causes the charged ions in an ionic crystal (such as NaCl) to oscillate about their equilibrium positions. In an optical mode, oppositely charged ions in each primitive cell undergo oppositely directed displacements, giving rise to a non-vanishing polarization density $\mathbf{P}$. Associated with this polarization density there will in general be a macroscopic electric field $\mathbf{E} = \text{Re}\{\mathbf{E}_0\} e^{i\mathbf{k} \cdot \mathbf{r}}$. In a longitudinal optical mode $\mathbf{P}$ is parallel to $\mathbf{k}$, whereas in a transverse optical mode $\mathbf{P}$
is perpendicular to $k$. The frequency dependent dielectric constant $\varepsilon(\omega)$ of the crystal is given by [17]

$$\varepsilon(\omega) \equiv \frac{k^2}{\omega^2} = \varepsilon_\infty + \frac{\varepsilon_\infty - \varepsilon_0}{(\omega^2/\omega_T^2) - 1}.$$  \hspace{1cm} (5.80)

Here $\varepsilon_0 \equiv \varepsilon(\omega = 0)$ stands for the static dielectric constant of the crystal, while $\varepsilon_\infty$ is the dielectric constant at optical frequencies, and therefore is related to the index of refraction $n$ by $n^2 = \varepsilon_\infty$. $\omega_T$ is the frequency of the long wavelength ($k \to 0$) transverse optical modes, and is related to the frequency $\omega_L$ of the longitudinal optical mode by

$$\omega_L^2 = \frac{\varepsilon_0}{\varepsilon_\infty} \omega_T^2, \hspace{1cm} (5.81)$$

$\omega_L$ always exceeding $\omega_T$. In terms of $\omega_L$ and $\omega_T$, Eq. (5.80) assumes the form

$$\varepsilon(\omega) = \varepsilon_\infty \left( \frac{\omega^2 - \omega_L^2}{\omega^2 - \omega_T^2} \right). \hspace{1cm} (5.82)$$

Clearly, $\varepsilon(\omega)$ is negative ($k$ is imaginary) in the frequency range $\omega_T < \omega < \omega_L$ and, therefore, no radiation in this frequency range can propagate in the crystal. The width of this so called polaritonic gap is thus $\Delta = \omega_L - \omega_T$, and its gap-to-midgap ratio is

$$\frac{\Delta}{\omega_o} = 2 \left( \frac{\sqrt{\varepsilon_0/\varepsilon_\infty} - 1}{\sqrt{\varepsilon_0/\varepsilon_\infty} + 1} \right), \hspace{1cm} (5.83)$$

where $\omega_o = \omega_T + \Delta/2$ is the central frequency of the gap, and we have used Eq. (5.81).

For instance, NaCl has $\omega_T = 3.18 \times 10^{13}$ Hz (which is in the infra-red), $\varepsilon_0 = 5.90$ and $\varepsilon_\infty = 2.34$[17], so that $\omega_L = 1.59\omega_T$, $\omega_o = 1.29\omega_T$, $\Delta = 12$ meV, and $\Delta/\omega_o = 45\%$. On the other hand, LiF has $\omega_T = 5.75 \times 10^{13}$ Hz, $\varepsilon_0 = 9.01$ and $\varepsilon_\infty = 1.96$ so that $\omega_L = 2.14\omega_T$, $\omega_o = 1.57\omega_T$, $\Delta = 43$ meV, and $\Delta/\omega_o = 72.6\%$. 

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Figure 5.5: The photon density of states in a dispersive medium described by the dispersion relation (5.84). The dashed curve represents the free space density of photon states. We see that $\rho(\omega)$ is highly singular at the lower band-edge $\omega_\nu$ behaving like $\rho(\omega) \sim (\omega_\nu - \omega)^{-4}$, but is identically zero at the upper band edge, $\rho(\omega_\nu) = 0$. This should be contrasted with the PBG case shown in Fig. 5.2 where $\rho(\omega)$ exhibits square-root singularities at both band edges. In the inset is shown the photon spectrum for a dispersive medium described by the dispersion relation (5.84).

5.4.1 Model Dispersion Relation

A simple model dispersion relation for a frequency dispersive medium can be written as \[189\]

$$\omega_{\pm}(k) = \frac{1}{2} \left[ (\omega_L + k) \pm \sqrt{(\omega_L - k)^2 + 4k\Delta} \right], \quad (5.84)$$

where $\omega_L$ is the frequency of the longitudinal optical mode and $\Delta = \omega_L - \omega_T$ is the width of the polaritonic gap. The $+(-)$ sign applies for the upper (lower) branch of the photon spectrum. The dispersion relation (5.84) is plotted in the inset in Fig. 5.5. It exhibits an isotropic band gap, with center frequency $\omega_\sigma = \omega_L - \Delta/2$ and band-edge frequencies $\omega_\nu = \omega_L - \Delta$ and $\omega_c = \omega_L$.

As shown in Appendix (F), the dispersion relation (5.84) gives

$$z(\omega) = \begin{cases} \frac{\omega^2 - 2\omega_\omega + \omega_\nu \omega_c}{(\omega - \omega_c)^2 + \kappa^2}, & \omega \not\in G \\ 0, & \omega \in G, \end{cases} \quad (5.85)$$

for the form factor in a dispersive medium. Here we have added a phenomenological
damping constant $\kappa$ to account for relaxation processes in the medium. From Eq. (5.85) we see that at the upper band edge $z(\omega) = \omega/\Delta$, whereas near the lower band edge $z(\omega) \sim (\omega - \omega_{\nu})^{-2}$. Unlike the PBG case, where $z(\omega)$ exhibits singularities at both band edges, $z(\omega)$ in a dispersive medium is singular only at the lower band edge $\omega_{\nu}$ and the square singularity here is much stronger than the square-root singularity of the isotropic PBG. From Eqs. (5.85) and (5.12) we obtain

$$\rho(\omega) = \begin{cases} \frac{\omega^2(\omega - \omega_{\nu})^2}{2\pi^2} \left[ \frac{\omega^2 - 2\omega\omega + \omega^2_{\nu}}{(\omega - \omega_{\nu})^4} \right], & \omega \notin G \\ 0, & \omega \in G \end{cases}$$

for the density of photon modes in a dispersive medium [see Appendix (F)].

The density of states (5.86) is plotted in Fig. (5.5). For regions far removed from the band gap, Eq. (5.86) shows that $\rho(\omega) \sim \omega^2/2\pi$, as expected. Near the lower band edge $\omega_{\nu}$, $\rho(\omega)$ behaves as $\rho(\omega) \sim (\omega_{\nu} - \omega)^{-4}$ for $\omega < \omega_{\nu}$. On the other hand, near the upper band-edge $\omega_c$, $\rho(\omega)$ behaves as $\rho(\omega) \sim (\omega - \omega_{c})^2$ for $\omega > \omega_{c}$. Thus $\rho(\omega)$ is highly singular at the lower band-edge $\omega_{\nu}$ but is identically zero at the upper band edge, $\rho(\omega_c) = 0$. These near band-edge behaviors of $\rho(\omega)$ can easily be understood from the slope of the dispersion curve (5.84). For both the upper and lower branches we obtain

$$\left( \frac{d\omega}{dk} \right)_{\pm} = \frac{1}{2} \left[ \frac{1 \pm \frac{(2\Delta - \omega_L) + k}{\sqrt{(\omega_L - k)^2 + 4\Delta k}}}{\sqrt{(\omega_L - k)^2 + 4\Delta k}} \right],$$

where the $+(-)$ sign applies for the upper(lower) branch. For $k \to \infty$, where the lower band edge occurs, we have $(d\omega/dk)_+ \to 0$ which means that $\rho(\omega)$ is singular at $\omega_{\nu}$. On the other hand at $k = 0$, where the upper band edge occurs, $(d\omega/dk)_- = \Delta/\omega_L$, which means that the group velocity is finite at the upper band edge $\omega_c$ (unless the width $\Delta$ of the gap is zero). The photon density of states which behaves as $\rho \sim k/(d\omega/dk)_+$ [see Eq. (F.5)] will then be zero at $\omega_c$, since $k = 0$ there. The strong singularity of $\rho(\omega)$ at the lower band edge can be explained by the fact that we have allowed the wave vector $k \to \infty$ while, in reality, only wave vectors within the first Brillouin zone are relevant. This unrealistic singularity in
Figure 5.6: The real part $\Sigma'(\epsilon)$ (solid curve) of the self energy for a frequency dispersive medium as a function of the scaled frequency $\epsilon/\omega_c$. The quantities $r$, $\omega_{10}$, $\omega_{20}$, $\gamma_{20}$ and $\gamma_{21}$ are the same as those in Fig. 5.3. The dotted line represents the real part of the free space self energy which is identically zero [see Eq. (5.37)]. In the inset is shown a plot of the imaginary part $\Sigma''(\epsilon)$ (solid curve). The dashed line in the inset represents the imaginary part of the free space self energy given by Eq. (5.38). The scale used for plotting $\Sigma''(\epsilon)$ is much smaller than that used for plotting $\Sigma'(\epsilon)$.

the photon density of states is removed when we take relaxation processes in the medium into account.

Using Eq. (5.85) in Eq. (5.30), we obtain the imaginary part $\Sigma''(\epsilon)$ of the self energy for a dispersive medium described by the isotropic dispersion relation (5.84). Likewise the real part $\Sigma'(\epsilon)$ is determined by substituting Eq. (5.85) into Eq. (5.29) and evaluating (numerically) the integral. Both the real and imaginary parts of the self energy are plotted in Fig. 5.6, where we choose $r = 15\%$, $\omega_{10} = \Delta/10$, $\omega_{20} = 0.95\omega_c$ and $\gamma_{20} = \gamma_{21} = \Delta/100$. The frequency intervals $G$ and $G'$ are given by $(0.925, 1.073)$ and $(0.95, 1.075)$, respectively. According to Eq. (5.53), $\Sigma''(\epsilon) = 0$ for $\epsilon \in G'$ and, according to Eqs. (5.30) and (5.85), $\Sigma''(\epsilon)$ exhibits singularities at $\omega_v = 0.925$ and $\omega_v + \omega_{10} = 0.94$. Likewise, the real part $\Sigma'(\epsilon)$ is singular at $\omega_v$ and at $\omega_v + \omega_{10}$. In the frequency range $(\omega_v, \omega_v + \omega_{10})$, $\Sigma'(\epsilon)$ monotonically increases from $-\infty$ to $+\infty$ whereas outside this range $\Sigma'(\epsilon)$ asymptotically approaches the free space value of zero.

The differences between a PBG and a FDM described by the isotropic dispersion relations (5.74) and (5.84), respectively, can be observed by contrasting Figs 5.3 and 5.6. In the PBG
case, both $\Sigma'(\epsilon)$ and $\Sigma''(\epsilon)$ are singular at $\omega_v, \omega_c, \omega_v + \omega_0$ and $\omega_c + \omega_0$. In the FDM case, however, $\Sigma'(\epsilon)$ and $\Sigma''(\epsilon)$ are not singular at $\omega_c$ and $\omega_c + \omega_0$ but rather exhibit relatively large values (compared to the free space case) at these frequencies. In both the PBG and FDM cases $\Sigma''(\epsilon) = 0$ for $\epsilon \in G' = (0.94, 1.075)$. However, in this frequency range, the real part $\Sigma'(\epsilon)$ behaves differently in the FDM from the PBG case. As we traverse $G'$ from left to right (i.e., from $\epsilon = 0.94$ to $\epsilon = 1.075$), in the PBG case, $\Sigma'(\epsilon)$ monotonically increases from a very large negative value to a very large positive value passing through the free space value of $\Sigma'(\epsilon) = 0$. On the other hand, in the FDM case, $\Sigma'(\epsilon)$ increases from $-\infty$ to about $\Sigma'(\epsilon) = -20$ and is entirely negative in the frequency range $G'$.

5.4.2 Discrete Modes and Continuous spectrum in a Dispersive Medium

In this section we briefly discuss the discrete bound states and the continuous spectra of section (5.2) when the atom is embedded in a frequency dispersive medium described by the dispersion relation (5.84). First we investigate the condition for the occurrence of a photon-atom bound state. As shown in Eq. (5.57), a discrete bound state is given by the eigenvalue $\epsilon$ satisfying $\Sigma'(\epsilon) = \omega_20 - \epsilon$ provided that $\epsilon \in G' = (0.94, 1.075)$. However, as shown in Fig. 5.6, in the frequency range $G'$, $\Sigma'(\epsilon) < -20$. Thus to satisfy the equality $\Sigma'(\epsilon) = \omega_20 - \epsilon$, we must have $\epsilon > \omega_20 + 20$ which means that $\epsilon \not\in G'$. In other words, Eq. (5.57) does not have a solution within the frequency range $G' = (0.94, 1.075)$ where $\Sigma'' = 0$. In conclusion, for the choice of parameters in section (5.3.1) used to support a photon-atom bound state in a PBG, a frequency dispersive medium does not support a photon-atom bound state.

Next we investigate equation (5.73) in the case of FDM. We choose the relevant parameters $\Delta, \omega_20, \gamma_21$, and $\gamma_21$ just like in the PBG case (see Fig. 5.4). In Fig. 5.7 we plot the Rayleigh spectrum $\sigma_R(\omega)$ versus $\omega - \omega_20$ for various values of the Stokes transition frequency $\omega_21$ ($\omega_21 = 1.15\omega_c, 1.05\omega_c, \omega_c$ and $\omega_v$). On the scale of the figure, all cases give the same spectral distribution which is “half-Lorentzian” cut off at $\omega - \omega_20 = 0.365$ (that is at $\omega = 0.925 = \omega_v$). The peak value of this spectrum is less than that of free space (shown
The spectrum of spontaneous emission $\sigma_R(\omega)$ for the Rayleigh transition $|2\rangle \rightarrow |0\rangle$ in the case of FDM described by the isotropic dispersion relation (5.84). The parameters $r = 15\%$, $\omega_0$, $\gamma_0$, and $\gamma_21$ have the same values as in Fig. 5.4. We plot $\sigma_R(\omega)$ for different values of $\omega_21$ ($\omega_21 = 1.15\omega_c$, 1.05$\omega_c$, $\omega_c$, and $\omega_c$) of the Stokes frequency. On the scale of this figure, all cases give the same spectral distribution cut off at $\omega = \omega_c$ and a peak value which is less than the free space case (shown in the inset) by about three orders of magnitude.

Figure 5.7 is the FDM analogue of Fig. 5.4. Comparing the two figures, we notice a number of differences. In the FDM case (unlike in the PBG case) there is no spectral splitting when $\omega_21$ lies close to the upper band edge $\omega_c$. This can be explained by comparing the density of states near $\omega_c$ in the PBG and FDM cases, Figs. 5.2 and 5.5, respectively. In the PBG case the photon density of states near $\omega_c$ (on the upper side) is much greater than that of free space leading to strong atom-field interaction, resulting in level splitting. On the other hand, in the FDM case the photon density of states in the neighborhood of $\omega_c$ is less than that of free space. In this case atom-field interaction in this frequency region is partially suppressed. Another difference between Figs. 5.4 and 5.7 is that, in the PBG case the spectral distribution $\sigma_R(\omega)$ is very sensitive to small changes in $\omega_21$, whereas in the FDM case all considered values of $\omega_21$ give essentially the same result. This can again be explained by referring to Figs. 5.2 and 5.5. As shown in Eq. (5.49) $\sigma_R(\omega)$ is proportional to $\rho(\omega)$ and therefore reflects the variation of the photon density of states with frequency. In the FDM case $\rho(\omega)$ varies little near $\omega_c$ whereas in the PBG case it is singular at $\omega_c$. 

in the inset) by about three orders of magnitude.
Finally, the peak of the spectral distribution is much larger in the PBG than in the FDM case. This can be explained by the fact that $\sigma_R(\omega) \propto \rho(\omega)$, as shown in Eq. (5.49).

The sensitivity of the Raman scattering cross-section near the band edge of an isotropic dispersion relation is due to the rapid variation of the density of states near the band edge. A divergence in the density of states at both band edges of the isotropic PBG model, and at the lower band edge of the FDM model leads to large vacuum Rabi splitting[5]. This vacuum Rabi splitting is absent near the upper band edge of the FDM. Similarly, in a more realistic anisotropic PBG model, vacuum Rabi splitting is absent. However, both Rabi splitting and non-Markovian radiative dynamics re-appear when an infinitesimal electromagnetic field is applied to the system. This may occur in the case of superradiance (collective light emission from a number of atoms)[185] or in the case of a very weak external field[147]

5.5 Discussion and Conclusions

Raman scattering from a three-level atom in a PBG material sensitively depends on the positions of the Stokes transition frequency $\omega_{21}$ and the Rayleigh transition $\omega_{20}$ relative to the band edge frequency $\omega_c$. When both $\omega_{21}$ and $\omega_{20}$ are far above $\omega_c$, the corresponding spectra are both Lorentzian as in free space. However, if either of the transition frequencies is close to $\omega_c$, the spectra will be markedly different from the free-space case. For example, if $\omega_{20}$ lies far above $\omega_c$, while $\omega_{21}$ is near $\omega_c$, the Rayleigh spectrum is split into triplets. The middle band which lies between $\omega_o + \omega_{10}$ and $\omega_c + \omega_{10}$ measures the fractionalized steady-state atomic population on the excited level |2$\rangle$. As $\omega_{21} \to \omega_c^+$, the dark lines shift upward in frequency and the width of the middle band increases. In this manner, resonance Raman scattering can be used as a direct experimental probe of the photon-atom bound state.
Chapter 6

Concluding Remarks and Future Perspectives

In this thesis the quantum electrodynamic properties of a three-level atom embedded in a PBG material were investigated. Specifically, the combined effects of coherent control by an external driving field and photon localization facilitated by a PBG on spontaneous emission from a three-level atom embedded in a PBG material were investigated. It was demonstrated that quantum information is stored in the model system considered, and that the nature of this information is strongly dependent on the externally prescribed initial conditions, suggesting an application of the model system as a single-atom optical memory device. Moreover, the protected electric dipole within the photonic band gap provides a basis for a qubit to encode information for quantum computations.

The resonance Raman scattering of a laser photon from such a three-level atom embedded in a PBG material was also studied in this thesis. It was demonstrated that the spectrum of the scattered light provides a distinctive experimental signature of atom-photon interactions near a photonic band edge.

The investigation in this thesis was based on the "effective- mass" dispersion model in the coherent control case, and the isotropic dispersion model in the Raman scattering case. The effective mass dispersion model is a good approximation if the photonic band gap is
relatively wide (compared to the detuning of the atomic transition from the atomic band edge), and if the relevant atomic transitions are near the band edge. Moreover, an isotropic dispersion model is expected to lead to results that are qualitatively similar to those of a more realistic anisotropic dispersion relation. However, in order to make quantitative comparison with experiment, it is important to investigate both the coherent control and the Raman scattering problems using a full anisotropic dispersion relation of a realistic photonic band structure.

Another interesting direction for future research is the “two-particle problem” involving the Raman scattering of a laser photon from a three-level atom which is initially in excited state. We have already seen in the coherent control problem that a driven three-level atom with a transition frequency near the edge of a photonic band gap can serve as a single-atom optical memory device, in the sense that the steady-state information written on the atom keeps memory of the externally prescribed initial conditions, such as the phase and intensity of the driving laser. The Raman scattering of a laser photon from such a three-level atom in the steady state may reveal the information written on the atom. The two-particle problem can then be viewed as a “reading” problem for the information “written” by the coherent control problem.

A possible scheme for a quantum optical logic gate is to use a “flying qubit” (polarized photon which comes in two possible circular polarization states $|L\rangle$ and $|R\rangle$) which then interacts with an atom that has a nondegenerate ground state coupled to a triply degenerate excited state by means of an optical transition that lies just inside the PBG. This would then require a generalization of our model to include the polarization degeneracy of the photon-atom bound state in the PBG material.

It is also useful to investigate the coherent control of spontaneous emission, not from a single three-level atom, but from two neighbouring three-level atoms coupled via resonant dipole-dipole interaction. Quantitative results on the energy transfer between the two atoms found from such an investigation may give us a qualitative picture on the effects of the presence of many atoms within a cubic wavelength of the atom of interest.
Appendix A

Hamiltonian of a Three Level Atom Interacting with a Quantized Radiation Field

In this appendix we derive the Hamiltonian of a three-level atom interacting with a quantized electromagnetic field in the electric dipole and in the rotating wave approximations.

A.1 Operator Description of Isolated Atoms

Consider an isolated atom\(^{1}\) with no radiation present. Such an atom is described by a Hamiltonian operator \(H_{a}\) which is a function of both the position \(\mathbf{r}\), and the momentum \(\mathbf{p}\) of the constituent particles of the atom, and has no explicit time dependence. This Hamiltonian satisfies the eigenvalue equation

\[
H_{a}|i\rangle = \hbar \omega_{i}|i\rangle ,
\]

(A.1)

where \(\hbar \omega_{i}\) are the energy eigenvalues and \(|i\rangle\) are the eigenvectors. The states \(|i\rangle\) span the Hilbert space of the atomic system. Since \(H_{a}\) is an observable, its basis vectors \(|i\rangle\) form a complete orthonormal set\(^{[156]}\). This is expressed by the closure (completeness) and

\(^{1}\)We use the terms “atom” and “molecule” equivalently in most of our discussions.
orthonormality relations

\[ \sum_i |i\rangle\langle i| = 1, \quad \langle i|j \rangle = \delta_{ij}, \quad (A.2) \]

where 1 represents the identity operator, and \( \delta_{ij} \) is the Kronecker delta function (\( \delta_{ij} = 1 \) for \( i = j \), and \( \delta_{ij} = 0 \) for \( i \neq j \)). We define the atomic operator \( \sigma_{ij} \) by

\[ \sigma_{ij} \equiv |i\rangle\langle j|. \quad (A.3) \]

These operators provide the multilevel generalization of Dicke’s spin operators for two level atoms\([168, 169]\). From the orthonormality relation (A.2) we obtain

\[ \sigma_{ij}|k\rangle = \delta_{jk}|i\rangle, \quad (A.4) \]

which, in turn, leads to the commutation relation

\[ [\sigma_{ij}, \sigma_{lk}] = \sigma_{ik}\delta_{jl} - \sigma_{lj}\delta_{ki}. \quad (A.5) \]

Thus, operator \( \sigma_{ij} \) acting on level \( |j\rangle \) transforms it to level \( |i\rangle \), and, therefore, is a raising (lowering) operator for \( i > j \) (\( i < j \)). On the other hand, the operator \( \sigma_{ii} = |i\rangle\langle i| \) gives the population of level \( |i\rangle \), that is, the probability to find the atom on level \( |i\rangle \).

Using Eq. (A.1) and the closure relation (A.2), we can expand the atomic Hamiltonian \( H_a \) in terms of the atomic operators \( \sigma_{ij} \) as

\[ H_a = H_a \sum_i |i\rangle\langle i| = \sum_i H_a |i\rangle\langle i| = \hbar \sum_i \omega_i \sigma_{ii}. \quad (A.6) \]

Similarly, by using the closure relation twice, any operator \( Q(p, r) \) which is a function of \( p \) and \( r \) can be expanded in terms of the atomic operators \( \sigma_{ij} \) as

\[ Q = \left( \sum_i |i\rangle\langle i| \right) Q \left( \sum_j |j\rangle\langle j| \right) = \sum_{i,j} Q_{ij} \sigma_{ij}, \quad (A.7) \]

where the expansion coefficients \( Q_{ij} = \langle i|Q|j\rangle \) are just the matrix elements of \( Q \) in the
energy representation. If $Q$ is Hermitian (as any observable should be), we have $Q_{ij} = Q_{ji}^*$.

A.2 Minimal Coupling Hamiltonian

In this section we consider an atom interacting with a quantized radiation field. For simplicity, the source of the radiation field (charges and currents) is not considered. We also assume the atom to have a single electron of charge $e$ and mass $m$ in a potential $V(r)$, where $r$ is the position vector of the electron. The momentum conjugate to $r$ is[32]:

$$ p = m \frac{d}{dt} r + eA. $$

(A.8)

We choose to work in the Coulomb gauge which is defined by the condition

$$ \nabla \cdot A = 0. $$

(A.9)

This gauge has particular advantages for slowly moving particles in bound states[32].

The non-relativistic minimal coupling Hamiltonian for a one-electron atom interacting with the electromagnetic radiation field may be written in mks units as [32, 35]

$$ H = \frac{1}{2m} (p - eA)^2 + eV(r) + H_r. $$

(A.10)

In the Coulomb gauge (A.9), we may write (A.10) as

$$ H = \frac{p^2}{m} + eV(r) + H_r - \frac{e}{m} A \cdot p + \frac{e^2}{2m} A^2. $$

(A.11)

The first two terms on the right-hand side of Eq. (A.11) give the Hamiltonian $H_a$ of the free atom:

$$ H_a = \frac{p^2}{2m} + eV(r). $$

(A.12)

This atomic Hamiltonian is given in terms of its stationary states $\{ |i \rangle \}$ by Eq. (A.6). The third term $H_r$ in Eq. (A.11) represents the energy of the quantized (source-free) radiation
field in the absence of the atom. It is given by

$$H_r = \sum_{k,\lambda} \hbar \omega_k \left( a_{k\lambda}^\dagger a_{k\lambda} + 1/2 \right),$$

(A.13)

where $k$ and $\lambda (=1,2)$ represent, respectively, the wave vector and the polarization index for the mode $\{k\lambda\}$ of the radiation field. The operators $a_{k\lambda}$ and $a_{k\lambda}^\dagger$ are, respectively, the Schrödinger-picture annihilation and creation operators for mode $\{k\lambda\}$ of the field obeying the commutation rules

$$[a_{k\lambda}, a_{k'\lambda'}^\dagger] = 0, \quad [a_{k\lambda}^\dagger, a_{k'\lambda'}] = 0, \quad [a_{k\lambda}, a_{k'\lambda'}] = \delta_{kk'} \delta_{\lambda\lambda'}.$$ (A.14)

In Eq. (A.13), the term $1/2 \sum_{k,\lambda} \hbar \omega_k$ accounts for the ground state or zero-point energy of the quantized radiation field. This energy is a constant, albeit infinite, and therefore has no effect whatsoever on the system dynamics. Thus it can be dropped out of the Hamiltonian $H_r$, which is tantamount to shifting the energy origin. Accordingly we write

$$H_o \equiv H_a + H_r = \sum_i \hbar \omega_i \sigma_i + \sum_{k,\lambda} \hbar \omega_k a_{k\lambda}^\dagger a_{k\lambda}$$ (A.15)

as the Hamiltonian of the unperturbed system.

The fourth term in Eq. (A.11), which is of first order in the coupling constant $e$, we denote by

$$H_1 = -\frac{e}{m} A \cdot p.$$ (A.16)

This term represents the interaction between the electron momentum $p$ and the radiation field $A$. It is small compared with $H_a$ and $H_r$, but is large compared with the last term which is of order $e^2$. This last term is denoted by

$$H_2 = \frac{e^2}{2m} A^2.$$ (A.17)

It represents the energy of mutual interaction between different modes of the radiation field through the coupling of the electron to the field. In this thesis we neglect the small term
The Hamiltonian (A.11) is given in the Schrödinger picture, where observables are represented by stationary (time-independent) operators. In this picture, the vector potential \( \mathbf{A}(\mathbf{r}) \) may be expanded in plane waves of mode \{\mathbf{k}\lambda\} as[32]

\[
\mathbf{A}(\mathbf{r}) = \sum_{\mathbf{k},\lambda} \left( \frac{\hbar}{2\epsilon_0\omega_k V} \right)^{1/2} \mathbf{e}_{\mathbf{k}\lambda} \left( a_{\mathbf{k}\lambda} e^{i\mathbf{k}\cdot\mathbf{r}} + a_{\mathbf{k}\lambda}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}} \right). 
\]

Here \( V \) represents the quantization volume, \( \epsilon_0 \) the Coulomb constant, and \( \mathbf{e}_{\mathbf{k}\lambda} \) are the two transverse (polarization) unit vectors satisfying

\[
\hat{\mathbf{k}} \cdot \mathbf{e}_{\mathbf{k}\lambda} = 0, \quad \mathbf{e}_{\mathbf{k}\lambda} \cdot \mathbf{e}_{\mathbf{k}\lambda'} = \delta_{\lambda\lambda'}, \quad (\lambda = 1, 2), 
\]

where \( \hat{\mathbf{k}} \) is the unit vector in the direction of \( \mathbf{k} \). The transversality condition \( \hat{\mathbf{k}} \cdot \mathbf{e}_{\mathbf{k}\lambda} = 0 \) expresses the fact that the mode amplitudes are perpendicular to the propagation direction and is a direct consequence of Eq. (A.9). On the other hand, condition \( \mathbf{e}_{\mathbf{k}\lambda} \cdot \mathbf{e}_{\mathbf{k}\lambda'} = \delta_{\lambda\lambda'} \) shows that unit vectors \{\mathbf{e}_{\mathbf{k}1}, \mathbf{e}_{\mathbf{k}2}, \hat{\mathbf{k}}\} form a right-handed triad.

According to Eq. (A.7), the electron momentum operator \( \mathbf{p} \) can be expanded as

\[
\mathbf{p} = \sum_{i,j} p_{ij} \sigma_{ij}, \quad (A.21)
\]

where \( p_{ij} = \langle i | \mathbf{p} | j \rangle \). Using the fact that

\[
[H_x, \mathbf{r}] = -\frac{i\hbar}{m} \mathbf{p}, \quad (A.22)
\]

we obtain

\[
p_{ij} \equiv \langle i | \mathbf{p} | j \rangle = -\frac{m}{i\hbar} \langle i | [H_x, \mathbf{r}] | j \rangle = \frac{im}{\epsilon} \omega_{ij} d_{ij}, \quad (A.23)
\]

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where

$$\omega_{ij} = \omega_i - \omega_j$$  \hspace{1cm} (A.24)

is the atomic transition frequency between levels $|i\rangle$ and $|j\rangle$, and

$$d_{ij} = \langle i | e r | j \rangle$$  \hspace{1cm} (A.25)

is the electric dipole moment transition matrix element between states $|i\rangle$ and $|j\rangle$. We have $d_{ij} \neq 0$, only if levels $|i\rangle$ and $|j\rangle$ are of opposite symmetry so that the transition $|i\rangle \leftrightarrow |j\rangle$ is dipole allowed. In particular, $d_{ii} = 0$. Using Eq. (A.23) in Eq. (A.21), we obtain

$$p = \frac{im}{e} \sum_{i,j} \omega_{ij} d_{ij} \sigma_{ij}$$  \hspace{1cm} (A.26)

as the expansion for the momentum operator $p$ in terms of the atomic operators $\sigma_{ij}$. This expansion and Eq. (A.19) can then be used in Eq. (A.16) to obtain

$$H_1 = -\frac{e}{m} A(r) \cdot p = -i \frac{\hbar}{\epsilon_0} \sum_{k, \lambda} \sum_{i,j} g_{k, \lambda}^{ij} \sigma_{ij} \left( a_{k, \lambda} e^{ik \cdot r} + a_{k, \lambda}^\dagger e^{-ik \cdot r} \right).$$ \hspace{1cm} (A.27)

Here

$$g_{k, \lambda}^{ij} = \frac{\omega_{ij} d_{ij}}{\hbar} \left( \frac{\hbar}{2 \epsilon_0 \omega_k V} \right)^{1/2} \hat{e}_{k, \lambda} \cdot \hat{d}_{ij}$$  \hspace{1cm} (A.28)

represents the frequency dependent coupling constant between the atomic transition $|i\rangle \rightarrow |j\rangle$ and the mode $\{k, \lambda\}$ of the radiation field. Also $d_{ij}$ and $\hat{d}_{ij}$ represent the magnitude and unit vector of the atomic dipole moment $d_{ij}$ for the transition $|i\rangle \rightarrow |j\rangle$.

In the optical regime of the spectrum where photon wavelengths are long compared to atomic dimensions ($\lambda_{\text{photon}} \sim 10^3 \text{Å}$, whereas $r_{\text{atom}} \sim 1 \text{Å}$), it is useful to make the electric dipole approximation ($k \cdot r \approx 0$) in Eq. (A.27). Using this approximation in Eq. (A.18) we obtain

$$H = \sum_i \hbar \omega_i \sigma_{ii} + \sum_{k, \lambda} \hbar \omega_k a_{k, \lambda}^\dagger a_{k, \lambda} - i \hbar \sum_{k, \lambda} \sum_{i,j} g_{k, \lambda}^{ij} \sigma_{ij} \left( a_{k, \lambda} + a_{k, \lambda}^\dagger \right),$$ \hspace{1cm} (A.29)

as the total Hamiltonian for the atom-field system in the electric-dipole approximation.
A.3 The $\Xi$, $\Lambda$, and $V$ configurations

Our discussion so far applies to a general $n$-level atom. In this section we restrict our attention to a three-level atom. We denote the ground level of a three level atom by $|0\rangle$, and the two excited levels by $|1\rangle$ and $|2\rangle$. A three level atom can be in one of three distinct configurations. These are the $\Xi$ (xi) or Cascade configuration, the $\Lambda$ (lambda) configuration, and the $V$ configuration[see Fig. (A.1)]. Direct dipole transitions are forbidden between levels $|2\rangle$ and $|0\rangle$ for $\Xi$ system, between levels $|1\rangle$ and $|0\rangle$ for $\Lambda$ system, and between levels $|2\rangle$ and $|1\rangle$ for $V$ system. Thus, each of the three configurations of Fig. (A.1) has two dipole allowed transitions, and two associated coupling constants. These coupling constants are $g^{21}_{k\lambda}$ and $g^{10}_{k\lambda}$ for the $\Xi$ system, $g^{20}_{k\lambda}$ and $g^{21}_{k\lambda}$ for the $\Lambda$ system, and $g^{20}_{k\lambda}$ and $g^{10}_{k\lambda}$ for the $V$ system.

The dipole operator is an odd vector operator with vanishing matrix elements between states of the same parity. Thus, $d_{ij} = 0$ for $i = j$. It follows from Eq. (A.28) that $g^{01}_{k\lambda} = 0$. Thus, for the $\Xi$ configuration, the interaction Hamiltonian (A.27) assumes the form

$$H_1 = -i\hbar \sum_{k,\lambda} \left( g^{21}_{k\lambda} \sigma_{21} + g^{12}_{k\lambda} \sigma_{12} + g^{10}_{k\lambda} \sigma_{10} + g^{01}_{k\lambda} \sigma_{01} \right) \left( a_{k\lambda} + a_{k\lambda}^\dagger \right).$$

(A.30)
Since the dipole operator \( d = e \mathbf{r} \) is Hermitian we have, \( d_{ij} = d_{ji}^* \). We also assume that the dipole transition matrix elements \( d_{ij} \) are real, as it is always possible to make them so by choosing the relative phases of the state vectors properly[70]. Then, the coupling constants \( g_{ji}^{ij} \) will also be real, and Eq. (A.28) shows that \( g_{k\lambda}^{ij} = -g_{kj}^{ji} \) since, according to Eq. (A.24), \( \omega_{ij} = -\omega_{ji} \). Thus

\[
H_1 = -i\hbar \sum_{k,\lambda} \left[ g_{k\lambda}^{21} (\sigma_{21} - \sigma_{12}) + g_{k\lambda}^{10} (\sigma_{10} - \sigma_{01}) \right] (a_{k\lambda} + a_{k\lambda}^\dagger)
\]  

(A.31)

Next we invoke the rotating wave approximation (RWA). This approximation admits only processes corresponding to the emission of a photon and the simultaneous lowering of an atomic state, or vice versa. Thus, in the RWA, terms (such as \( \sigma_{01} a_{k\lambda} \) and \( \sigma_{10} a_{k\lambda}^\dagger \)) which describe virtual processes of excitation (de-excitation) of the atom with simultaneous creation (annihilation) of a photon are neglected[70]. Under the RWA, Eq. (A.31) reduces to

\[
H_1 = i\hbar \sum_{k,\lambda} \left[ g_{k\lambda}^{21} \left( a_{k\lambda}^\dagger \sigma_{12} - \sigma_{21} a_{k\lambda} \right) + g_{k\lambda}^{10} \left( a_{k\lambda}^\dagger \sigma_{01} - \sigma_{10} a_{k\lambda} \right) \right].
\]  

(A.32)
Appendix B

Non-Markovian Memory Kernels

In this appendix we will derive the various expressions for the Green's functions used in Chapters (3) and (4). We start from Eq. (4.10)

\[ G_{lm}(t - t') = \sum_{k\lambda} g^0_{k\lambda} g_{k\lambda}^{m0} e^{-i\mu_{k}^{m0}(t - t')}, \quad (l, m = 1, 2), \tag{B.1} \]

where \( \mu_{k}^{m0} \) is given by Eq. (4.6). When \( l = m \), Eq. (B.1) reduces to Eq. (3.35). Substituting for \( g^0_{k\lambda} \) and \( g_{k\lambda}^{m0} \) from Eq. (4.2) we obtain

\[ G_{lm}(t - t') = \frac{\zeta}{V} \sum_{k\lambda} (\hat{e}_{k\lambda} \cdot \hat{d}_{l0})(\hat{e}_{k\lambda} \cdot \hat{d}_{m0}) \frac{1}{\omega_{k}} e^{-i\mu_{k}^{m0}(t - t')}, \tag{B.2} \]

where

\[ \zeta = \sqrt{\left(\frac{\omega_{l0}^{2} d_{l0}^{2}}{2\hbar\varepsilon_{o}}\right) \left(\frac{\omega_{m0}^{2} d_{m0}^{2}}{2\hbar\varepsilon_{o}}\right)}. \tag{B.3} \]

Assuming that the modes of the field are closely spaced in frequency, we make the continuum approximation for the field modes and replace the summation over \( k \) by an integral:

\[ \sum_{k} \rightarrow \frac{V}{(2\pi)^3} \int d^3k, \tag{B.4} \]

where \( d^3k \equiv k^2dkd\Omega \), \( d\Omega \) being the space angle element. Thus

\[ G_{lm}(t - t') = \frac{\zeta}{(2\pi)^3} \frac{8\pi}{3} \int \left[ \frac{3}{8\pi} \sum_{\lambda} (\hat{e}_{k\lambda} \cdot \hat{d}_{l0})(\hat{e}_{k\lambda} \cdot \hat{d}_{m0}) \right] \frac{1}{\omega_{k}} e^{-i(\omega_{k} - \omega_{m0})(t - t')} d^3k. \tag{B.5} \]
This is a general result valid for any dispersion relation $\omega_k$. When the dispersion relation is isotropic (i.e., when $\omega_k$ depends only on the magnitude $k$ of $k$), Eq. (B.5) reduces to

$$G_{lm}(t - t') = \frac{\zeta}{(2\pi)^3} \frac{8\pi}{3} \eta_{lm} \int_0^\Lambda \frac{1}{\omega_k} e^{-i(\omega_k - \omega_{m0})(t - t')} k^2 dk,$$  (B.6)

where

$$\eta_{lm} = \frac{3}{8\pi} \int \sum_\lambda \left( \hat{e}_{k\lambda} \cdot \hat{d}_{l0} \right) \left( \hat{e}_{k\lambda} \cdot \hat{d}_{m0} \right) d\Omega,$$  (B.7)

and $\Lambda = mc/\hbar$ is the Compton wave number of the electron. We have introduced the cut-off $\Lambda$ in the photon wave vector $[184]$ as the contributions of extremely high energy photons cannot be important. The non-relativistic approximation for the electron is not valid for photons of energy $\hbar \omega \sim mc^2$.

We consider a coordinate system defined by the unit vectors $\{\hat{e}_{k1}, \hat{e}_{k2}, \hat{k}\}$. Defining $\{\alpha_{l0}, \beta_{l0}, \theta_{l0}\}$ as the direction angles of the dipole moment unit vector $\hat{d}_{l0}$, we obtain

$$\eta_{lm} = \frac{3}{8\pi} \int \left( \cos \alpha_{l0} \cos \alpha_{m0} + \cos \beta_{l0} \cos \beta_{m0} \right) d\Omega.$$  (B.8)

If the dipoles $\hat{d}_{l0}$ and $\hat{d}_{m0}$ are parallel or anti-parallel (so that $\alpha_{l0} = \pm \alpha_{m0} = \alpha$, $\beta_{l0} = \pm \beta_{m0} = \beta$, and $\theta_{l0} = \pm \theta_{m0} = \theta$), the law of direction cosines gives

$$\cos \alpha_{l0} \cos \alpha_{m0} + \cos \beta_{l0} \cos \beta_{m0} = \cos^2 \alpha + \cos^2 \beta = 1 - \cos^2 \theta.$$  (B.9)

Using this in Eq. (B.8) we obtain $\eta_{lm} = 1$. It follows that

$$\eta_{lm} = \delta_{lm} + \eta(1 - \delta_{lm}),$$  (B.10)

where $\delta_{lm}$ is the Kronecker delta function, and

$$\eta = \frac{3}{8\pi} \int \left( \cos \alpha_{20} \cos \alpha_{10} + \cos \beta_{20} \cos \beta_{10} \right) d\Omega.$$  (B.11)

Thus $\eta = 1$, when $\hat{d}_{l0} = \pm \hat{d}_{m0}$, that is, when the dipoles associated with the transitions $l\to |0\rangle$ and $m\to |0\rangle$ are parallel or antiparallel.
For vacuum we use the isotropic dispersion relation $\omega_k = ck$. The emitted radiation is centered about the atomic transition frequency $\omega_k = \omega_{m0}$, and the quantity $\omega_k$ varies very little around $\omega_k = \omega_{m0}$. We can, therefore, replace $k^2/\omega_k$ in Eq. (B.6) by $\omega_{m0}/c^2$ and extend the upper limit of integration to $\infty$ to obtain

$$G_{lm}(t - t') = \frac{\zeta \omega_{m0}}{(2\pi c)^3} \frac{8\pi}{3} \eta_{lm} \int_0^\infty e^{-i(\omega_k - \omega_{m0})(t - t')} d\omega_k.$$  \hspace{1cm} (B.12)

On the other hand, we have

$$\delta(t) = \frac{1}{2\pi} \int_{-\infty}^\infty d\omega e^{i\omega t}$$ \hspace{1cm} (B.13)

as the integral expression for the Dirac delta function. Thus

$$G_{lm}(t - t') = \eta_{lm} \sqrt{\gamma_{l0}} \gamma_{m0} (\omega_{m0}/\omega_{l0}) \delta(t - t'),$$ \hspace{1cm} (B.14)

where

$$\gamma_{j0} = \frac{1}{4\pi \epsilon_0} \frac{4\omega_{j0}^3 d_{j0}^2}{6\hbar c^3}, \, \, \, (j = 1, 2)$$ \hspace{1cm} (B.15)

is half the vacuum spontaneous emission rate $\Gamma_{j0} = 2\gamma_{j0}$ for the transition $|j\rangle \rightarrow |0\rangle$.

Assuming that the upper levels $|2\rangle$ and $|1\rangle$ are close together so that $\omega_{m0}/\omega_{l0} \approx 1$, we finally obtain

$$G_{lm}(t - t') = \eta_{lm} \sqrt{\gamma_{l0}} \gamma_{m0} \delta(t - t').$$ \hspace{1cm} (B.16)

The dipole moment of a hydrogenic (that is one-electron) atom can be approximated by $d_{20} \sim e a_0$, where $e$ is the magnitude of the electronic charge, and $a_0 \sim 0.5\text{Å}$ is the Bohr radius. Moreover, for optical transition frequencies $\omega_{20} \sim 5 \times 10^{15}$ Hz. Using these in Eq. (B.15) we obtain $\gamma_{j0} \sim 10^8 s^{-1}$.

For a PBG material described by the isotropic "effective mass" dispersion relation (3.57), Eq. (B.5) takes the form

$$G_{lm}(t - t') = \frac{\zeta}{(2\pi)^3} \frac{8\pi}{3} \eta_{lm} e^{i\delta_{m0}(t - t')} \int_{k_0}^{\Lambda} \frac{k^2 e^{-i\Lambda(k - k_0)^2(t - t')}}{\omega_c + A(k - k_0)^2} dk.$$

where $\delta_{m0} = \omega_{m0} - \omega_c$ is the detuning of the atomic transition frequency $\omega_{m0}$ from the
band-edge frequency $\omega_c$. The integral in Eq. (B.17) can be approximated by replacing $k$ by $k_0$ outside of the exponential and extending the wave-vector integration to infinity, which then reduces to a complex Fresnel integral given by\[190\]

$$
\int_0^\infty e^{-iAt^2(t-t')}du = \frac{e^{-i\pi/4}}{2\sqrt{A}} \sqrt{\frac{\pi}{t-t'}}.
$$

(B.18)

Using Eqs. (B.3) and (B.18) in Eq. (B.17) we obtain

$$
G_{lm}(t-t') = \eta_{lm} \sqrt{\beta_{10}^{3/2} \beta_{m0}^{3/2} \frac{e^{i[\delta_{m0}(t-t')-\pi/4]}}{\sqrt{\pi(t-t')}}},
$$

(B.19)

where

$$
\beta_{j0}^{3/2} = \frac{1}{4} \left( \frac{\gamma_j}{\omega_j} \right) \left( \frac{c\omega_c}{A^{1/2}} \right).
$$

(B.20)

This expression may be further simplified by assuming that the upper levels $|2\rangle$ and $|1\rangle$ are close together so that $\omega_2 \approx \omega_1$. We then have

$$
\sqrt{\beta_{10}^{3/2} \beta_{20}^{3/2}} \approx \beta^{3/2} \equiv \frac{1}{4} \left( \frac{\gamma_2}{\omega_2} \right) \left( \frac{c\omega_c}{A^{1/2}} \right).
$$

(B.21)

If we use the approximation $A \approx c^2/\omega_c$ we finally obtain

$$
\beta^{3/2} \approx \frac{1}{4} \left( \frac{\gamma_2}{\omega_2} \right) \omega_c^{3/2}.
$$

(B.22)

At optical frequencies $\gamma_2 \sim 10^8 \text{ Hz}$ and $\omega_2 \sim 2\pi \times 10^{15} \text{ Hz}$ so that $\beta \sim 10^{-6}\omega_c$. Thus, if the band-edge $\omega_c$ is also in the optical regime, we have $\beta \sim 10^{10} \text{ Hz}$. As shown in Appendix (E), for a two level atom placed inside a photonic crystal of band edge frequency nearly resonant with the atomic transition frequency, the upper level splits into a doublet because of the strong interaction between the atom and its own localized radiation. $\beta$ gives the magnitude of this frequency splitting for the isotropic dispersion relation (3.57). Thus for a band-gap in the optical regime, $\beta \sim 10^{10} \text{ Hz}$ is larger than the ordinary vacuum Lamb shift ($\sim 10^9 \text{ Hz}$) of the $2p_{1/2}$ level of hydrogen relative to the $2s_{1/2}$ level.

For a PBG material described by the anisotropic "effective mass" dispersion relation (3.58),
the general expression (B.5) takes the form

\[ G_{lm}(t - t') = \frac{\zeta}{(2\pi)^3} \frac{8\pi}{3} e^{i\theta_{m0}(t - t')} \int \left[ \frac{3}{8\pi} \sum_k \left( \hat{e}_{k\lambda} \cdot \hat{d}_{l0} \right) \left( \hat{e}_{k\lambda} \cdot \hat{d}_{m0} \right) \right] \frac{e^{-iA(k - k_0)^2(t - t')}}{\omega_c + A(k - k_0)^2} d^3k. \]  

(M.23)

Making the substitution \( q = k - k_0 \), so that \( d^3q = q^2 dq dq \), performing the angular integration, and extending the wave-vector integration to infinity we obtain

\[ G_{lm}(t - t') = \frac{\zeta}{(2\pi)^3} \frac{8\pi}{3} \eta_{lm} e^{i\theta_{m0}(t - t')} \frac{1}{A} \int_0^\infty \frac{e^{-iAq^2(t - t')}}{\omega_c/A + q^2} q^2 dq, \]  

(B.24)

where \( \eta_{lm} \) is given by Eq. (B.8). For large \( t - t' \), the integral in Eq. (B.24) is dominated by the stationary phase point \( q = 0 \). Thus, the integral can be approximated by putting \( q = 0 \) in the denominator, and using

\[ \int_0^\infty x^2 e^{-ax^2} dx = \sqrt{\pi}a^{-3/2}/4, \]  

(B.25)

to obtain

\[ G_{lm}(t - t') = -\eta_{lm} \sqrt{\alpha_{j0} \alpha_{m0}} \frac{e^{i[\theta_{m0}(t - t') + \pi/4]}}{\sqrt{4\pi(t - t')^3}} \omega_c(t - t') \gg 1, \]  

(B.26)

where

\[ \alpha_{j0} = \frac{1}{4} \left( \frac{\gamma_{j0}}{\omega_{j0}} \right) \left( \frac{c^3}{\omega_c A^{3/2}} \right), \]  

(B.27)

and we have used Eq. (B.15). Eq. (B.26) may be further simplified by assuming that the upper levels \( |2\rangle \) and \( |1\rangle \) are close together so that \( \omega_{20} \approx \omega_{10} \). We then have

\[ \sqrt{\alpha_{10} \alpha_{20}} \approx \alpha \equiv \frac{1}{4} \left( \frac{\gamma_{20}}{\omega_{20}} \right) \left( \frac{c^3}{\omega_c A^{3/2}} \right). \]  

(B.28)

The full expression of \( G_{lm}(t - t') \), including its short-time behavior, is given in Ref.[158].

Eq. (B.28) shows that the value of \( \alpha_{j0} \) strongly depends on the curvature \( A = (1/2)(\partial^2 \omega/\partial k^2)_{k = k_0} \) of the dispersion curve \( \omega(k) \) at the band edge, \( k = k_0 \). For the anisotropic dispersion relation we cannot use the approximation \( A \approx c^2/\omega_c \) because the dispersion curve, in general, exhibits different curvatures in different directions. Instead we can use \( A \approx f c^2/\omega_c \) where
$f$ is a dimensionless number, whose value depends on the nature of the dispersion relation near the band edge $\omega_c$. This reduces Eq. (B.28) to

$$\alpha^2 = \frac{1}{16 f^3} \left( \frac{\gamma_{20}}{\omega_{20}} \right)^2 \omega_c.$$  \hspace{1cm} (B.29)
Appendix C

Time Dependence of the Atomic Amplitudes and The Strong Field Limit

The solutions $c_{1,2}(t)$ of Eqs. (3.33) and (3.34), in the case of PBG, are found from the inverse Laplace transform of the expressions for $\tilde{c}_{1,2}(s + i\delta)$ given by Eqs. (3.63) and (3.64) through the complex inversion formula

$$e^{-it}c_j(t) = \frac{1}{2\pi i} \int_{\epsilon-i\infty}^{\epsilon+i\infty} e^{st}\tilde{c}_j(s + i\delta)ds.$$ \hspace{1cm} (C.1)

Here the real number $\epsilon$ is chosen so that $s = \epsilon$ lies to the right of all the singularities (poles and branch points) of the functions $\tilde{c}_{1,2}(s + i\delta)$. It is apparent from Eq. (3.65) that $s = 0$ is a branch point of both $\tilde{c}_{1,2}(s + i\delta)$. In order to evaluate (C.1), we consider the contour $C$ shown in Fig. (C.1) where the branch cut of the integrand is chosen to lie along the negative real axis.

According to the residue theorem

$$\frac{1}{2\pi i} \oint_C e^{st}\tilde{c}_j(s + i\delta)ds = R_{\text{sum}},$$ \hspace{1cm} (C.2)
where $R_{\text{sum}}$ is the sum of the residues of the integrand at the poles enclosed by the contour $C$. Omitting the integrand, we obtain

$$R_{\text{sum}} = \frac{1}{2\pi i} \oint_C = \frac{1}{2\pi i} \left( \int_{AB} + \int_{BDE} + \int_{EH} + \int_{HJK} + \int_{KL} + \int_{LNA} \right). \quad (C.3)$$

In the limit $r \to 0$ and $R \to \infty$ (so that $T \to \infty$), the second, the fourth and the sixth integrals on the RHS of Eq. (C.3) approach zero and, according to Eq. (C.1), the first integral gives $e^{-\sqrt{t}c_j(t)}$. Thus

$$e^{-\sqrt{t}c_j(t)} = R_{\text{sum}} - \lim_{R \to \infty, r \to 0} \frac{1}{2\pi i} \left( \int_{EH} + \int_{KL} \right). \quad (C.4)$$

First we calculate $c_2(t)$. Along EH, $s = xe^{i\pi} = -x$. Using this in Eq. (3.63) we obtain

$$\lim_{R \to \infty, r \to 0} \int_{EH} e^{st}c_2(s + i\delta)ds = \int_0^\infty \frac{[(-x + i\delta)\cos \theta - \Omega e^{i\phi} \sin \theta]e^{-xt}}{(-x + i\delta)^2 - a e^{-i\pi/4}(-x + i\delta)\sqrt{x + \Omega^2}}dx. \quad (C.5)$$

Similarly, along KL, $s = xe^{-i\pi} = -x$. Using this in Eq. (3.63) we obtain

$$\lim_{R \to \infty, r \to 0} \int_{KL} e^{st}c_2(s + i\delta)ds = -\int_0^\infty \frac{[(-x + i\delta)\cos \theta - \Omega e^{i\phi} \sin \theta]e^{-xt}}{(-x + i\delta)^2 + a e^{-i\pi/4}(-x + i\delta)\sqrt{x + \Omega^2}}dx. \quad (C.6)$$
Using Eqs. (C.5) and (C.6) in Eq. (C.4) we obtain

\[
e^{-i\delta t}c_2(t) = R_{\text{sum}} + \frac{\alpha e^{i\pi/4}}{\pi} \int_0^\infty \frac{g_2(x)e^{-(x-i\delta)t}}{Z(x)} \, dx.
\]

where

\[
g_2(x) = [(-x + i\delta)\cos \theta - \Omega e^{i\theta} \sin \theta](-x + i\delta)\sqrt{x}, \tag{C.8}
\]

\[
Z(x) = [(-x + i\delta)^2 + \Omega^2]^2 + i\alpha^2(-x + i\delta)^2x. \tag{C.9}
\]

Next we evaluate the total residue \(R_{\text{sum}}\). From Eqs. (3.63) and (3.65) we have

\[
e^{st} \tilde{c}_2(s + i\delta) = [(s + i\delta)\cos \theta - \Omega e^{i\theta} \sin \theta]e^{st} \prod_{j=1}^4 \frac{\sqrt{s + e^{i\pi/4}u_j}}{s - iu_j^2} \tag{C.10}
\]

Clearly, the function \(e^{st}\tilde{c}_2(s + i\delta)\) has simple poles at \(s = iu_j^2, (j = 1, \ldots, 4)\). The residue \(R_k\) at \(s = iu_k^2\) is then

\[
R_k \equiv \lim_{s \to iu_k^2} (s - iu_k^2) e^{st} \tilde{b}_3(s + i\delta)
\]

\[
= \left[(u_k^2 + \delta)\cos \theta + i\Omega e^{i\theta} \sin \theta\right] e^{iu_k^2t} \frac{(\sqrt{u_k^2 + u_1}) \cdots (\sqrt{u_k^2 + u_4})}{(u_k^2 - u_l^2)(u_k^2 - u_m^2)(u_k^2 - u_n^2)} (k \neq l \neq m \neq n). \tag{C.11}
\]

Numerical examinations show that the roots \(u_{1,3}\) are real (\(u_1\) is positive but \(u_3\) is negative). The roots \(u_{2,4}\) are complex conjugates of each other with a negative real part (\(u_2\) and \(u_4\) lie in the third and second quadrants, respectively). Thus, the negative root \(u_3\) lies outside the contour \(C\) so that the residue at \(u_3\) is \(R_3 = 0\). For the complex root \(u_4\) (which has a positive imaginary part), the factor \(e^{iu_4^2t}\) increases exponentially in time and therefore is unphysical. Thus, for this root we choose the negative branch of the square root function and set \(\sqrt{u_4^2 + u_4} = 0\) so that the residue at \(u_4\) is \(R_4 = 0\). On the other hand, for the positive root \(u_1\) and the complex root \(u_2\) we choose the positive branch of the square root function.
function and set $\sqrt{u_j^2} = u_j$, $(j = 1, 2)$. The residues at $u_1$ and $u_2$ are

$$R_j = P_j Q_j e^{iu_j^2 t}, \quad (j = 1, 2), \quad (C.12)$$

where

$$P_j = \frac{2u_j}{(u_j - u_l)(u_j - u_m)(u_j - u_n)}, \quad (l, m, n = 1, ..., 4, j \neq l \neq m \neq n). \quad (C.13)$$

$$Q_j = (u_j^2 + \delta)\cos \theta + i\Omega e^{i\phi} \sin \theta. \quad (C.14)$$

The sum of the residues of the function $e^{st} \bar{c}_2(s + i\delta)$ is then

$$R_{\text{sum}} = \sum_{k=1}^{4} R_k = \sum_{j=1}^{2} P_j Q_j e^{iu_j^2 t}. \quad (C.15)$$

Using this in Eq. (C.7) we finally arrive at the desired result (3.76):

$$c_2(t) = \sum_{j=1}^{2} P_j Q_j e^{i(u_j^2 + \delta)t} + \frac{\alpha e^{i\pi/4}}{\pi} \int_{0}^{\infty} \frac{g_2(x)e^{-(x-i\delta)t}}{Z(x)} dx. \quad (C.16)$$

Following exactly the same procedure we also find that

$$c_1(t) = \sum_{j=1}^{2} P_j R_j e^{i(u_j^2 + \delta)t} + \frac{\alpha \Omega e^{i(\phi - \pi/4)}}{\pi} \int_{0}^{\infty} \frac{g_1(x)e^{-(x-i\delta)t}}{Z(x)} dx. \quad (C.17)$$

where $P_j$ and $Z(x)$ are the same as those for $c_2(t)$ and

$$R_j = (u_j^2 + \alpha u_j + \delta)e^{i\phi} \sin \theta - i\Omega e^{i\phi} \cos \theta, \quad (C.18)$$

$$g_1(x) = \{(-x + i\delta)\cos \theta - \Omega e^{i\phi} \sin \theta\} \sqrt{x}. \quad (C.19)$$

For a control laser field so strong that $\Omega \gg \alpha^2, \delta$, the roots given by Eqs. (3.67) and (3.68) are approximately given by

$$u_1 \sim \sqrt{\Omega}, \quad u_2 \sim -\sqrt{\Omega}, \quad u_2 = u_4^* \sim -\sigma_2 - i\sqrt{\Omega}, \quad \text{for} \ \Omega \gg \alpha^2. \quad (C.20)$$
Using these in Eqs. (C.13), (C.14), and (C.18) we obtain,

\[ P_1 \sim 1/2\Omega \]  \hspace{1cm} (C.21)
\[ Q_1 \sim \Omega [\cos \theta + ie^{i\phi} \sin \theta ] \]  \hspace{1cm} (C.22)
\[ R_1 \sim -i\Omega e^{i\phi_c} [\cos \theta + ie^{i\phi} \sin \theta ] \]  \hspace{1cm} (C.23)

which can then be used in Eqs.(3.84) and (3.86) to obtain

\[ n_{2s} \approx n_{1s} \approx \frac{1}{4} (1 - \sin 2\theta \sin \phi) \], \[ n_{cs} \approx \frac{ie^{-i\phi_c}}{4} (1 - \sin 2\theta \sin \phi) \]  \hspace{1cm} (C.24)

for the steady-state values of populations and coherences in the limit of a strong driving field.
Appendix D

Quantum Beat Solutions

D.1 The Vacuum Case

In this appendix Eq. (4.16), which gives the solutions to Eqs. (4.13) and (4.14) in the quantum beats case, will be derived. For $\Omega = 0$, Eqs. (4.13) and (4.14) reduce to

\begin{align}
    \dot{c}_1(t) &= -\gamma_0 c_1(t) - \eta \tilde{\gamma} e^{-i\omega_2 t} c_2(t), \\
    \dot{c}_2(t) &= -\gamma_2 c_2(t) - \eta \tilde{\gamma} e^{i\omega_2 t} c_1(t).
\end{align}

These equations constitute a linear homogeneous system with time-dependent coefficients. Solution of these equations is facilitated if we introduce new functions $f_j(t)$ by

\begin{equation}
    c_j(t) = e^{-\gamma_0 t} f_j(t), \quad (j = 1, 2)
\end{equation}

in terms of which the equations can be re-written as

\begin{align}
    \dot{f}_1(t) &= -\eta \tilde{\gamma} e^{-i\lambda t} f_2(t), \\
    \dot{f}_2(t) &= -\eta \tilde{\gamma} e^{i\lambda t} f_1(t).
\end{align}
where \( \lambda \) is a complex constant given by

\[
\lambda = \gamma_{20} - \gamma_{10} + i\omega_{21}. \tag{D.6}
\]

Taking the derivative with respect to time of Eq. (D.4) and using Eq. (D.5) we obtain

\[
f_2(t) - \lambda f_2(t) - (\eta \bar{\gamma})^2 f_2(t) = 0. \tag{D.7}
\]

This is a second order ordinary linear differential equation with constant coefficients. Its characteristic equation is

\[
q^2 - \lambda q - (\eta \bar{\gamma})^2 = 0 \tag{D.8}
\]

with solutions

\[
q_{1,2} = \frac{\lambda}{2} \pm \sqrt{\left(\frac{\lambda}{2}\right)^2 + (\eta \bar{\gamma})^2}. \tag{D.9}
\]

The general solution of Eq. (D.7) is thus

\[
f_2(t) = A_1 e^{\eta_1 t} + A_2 e^{\eta_2 t}, \tag{D.10}
\]

where \( A_{1,2} \) are arbitrary constants. Going back to Eq. (D.5) we obtain

\[
f_1(t) = -\frac{1}{\eta \bar{\gamma}} f_2(t). \tag{D.11}
\]

Substituting the derivative of Eq. (D.10) into Eq. (D.11), we obtain

\[
f_1(t) = -\frac{1}{\eta \bar{\gamma}} \left( A_1 q_1 e^{\eta_1 t} + A_2 q_2 e^{\eta_2 t} \right) e^{-\lambda t}. \tag{D.12}
\]

We now use the given initial values for the amplitudes \( c_j(t) \) to determine the constants \( A_{1,2} \). From Eq. (D.3) we see that \( f_j(0) = c_j(0) \). Using this in Eqs. (D.10) and (D.10), and solving the resulting coupled equations for \( A_{1,2} \) we obtain

\[
A_j = \frac{q_k c_2(0) + \eta \bar{\gamma} c_1(0)}{q_k - q_j}, \quad (k \neq j). \tag{D.13}
\]
Now that we have the solutions for \( f_j(t) \), we use Eq. (D.3) to obtain

\[
c_2(t) = e^{-\gamma_2 t} \sum_{j=1}^{2} A_j e^{q_j t}, \quad c_1(t) = e^{-(\gamma_2 + i\omega_2 t)} \sum_{j=1}^{2} B_j e^{q_j t},
\]

(D.14)

where

\[
B_j = -q_j A_j / \eta \gamma.
\]

(D.15)

This completes the solution of Eqs. (D.1) and (D.2).

When \( \eta = 0 \), Eqs. (D.1) and (D.2) have the exponentially decaying solutions given by

\[
c_j(t) = c_j(0) e^{-\gamma_2 t}, \quad (j = 1, 2).
\]

(D.16)

Comparing these solutions with the general solutions (D.14), we see that

\[
A_1 = 0, \quad A_2 = c_2(0), \quad B_1 = c_1(0), \quad B_2 = 0; \quad \text{for } \eta = 0.
\]

(D.17)

Next we show that the amplitudes \( c_j(t) \) given by Eq. (D.14) decay to zero in the long time limit:

\[
\lim_{t \to \infty} c_j(t) = 0.
\]

(D.18)

From Eq. (D.14), we see that Eq. (D.18) will be satisfied only if

\[
\text{Re}\{q_{1,2}\} < \gamma_2,
\]

(D.19)

where \( \text{Re}\{\} \) denotes the real part of the quantity in braces. Condition (D.19) can easily be proved if we assume that \( \gamma_{10} \approx \gamma_2 \) so that \( \tilde{\gamma} \approx \gamma_2 \). This a reasonable assumption, especially if the upper levels \(|2\rangle \) and \(|1\rangle \) are close to each other but far from the ground level \(|0\rangle \) so that \( \omega_{21} \ll \omega_2, \omega_{10} \); as is usually the case in the V configuration. In this approximation, Eq. (D.6) shows that \( \lambda = i\omega_{32} \) so that Eq. (D.9) gives

\[
q_{1,2} = \begin{cases} 
\pm \omega_{21}/2 & \text{if } \eta \gamma_2 \geq \omega_{21}/2, \\
\pm i\omega_{21}/2 & \text{if } \eta \gamma_2 < \omega_{21}/2,
\end{cases}
\]

(D.20)
where
\[ x = \sqrt{\left| (\eta \gamma_20)^2 - \left( \frac{\omega_{21}}{2} \right)^2 \right|}. \] (D.21)

Since \( \eta \leq 1 \), it follows that \( x < \gamma_{20} \). From Eq. (D.20) we see that \( \text{Re}\{q_{1,2}\} = \pm x \) if \( \eta \gamma_{20} \geq \omega_{21}/2 \) or \( \text{Re}\{q_{1,2}\} = 0 \) if \( \eta \gamma_{20} < \omega_{21}/2 \). Thus in both cases condition (D.19) is easily satisfied.

### D.2 The PBG Case

In this section Eqs. (4.36) and (4.37), which are solutions to Eqs. (4.33) and (4.34) when \( \eta = 1 \), and \( \delta_{20} = -\delta_{10} = \delta \geq 0 \), will be derived. In this special case, Eqs. (4.33) and (4.34) reduce to

\[
\dot{h}_1(t) = i\delta h_1(t) - \int_0^t G(t - t') [h_1(t') + h_2(t')] dt',
\] (D.22)

\[
\dot{h}_2(t) = -i\delta h_2(t) - \int_0^t G(t - t') [h_1(t') + h_2(t')] dt'.
\] (D.23)

Upon taking the Laplace transforms of these equations, we obtain

\[
\tilde{h}_1(s) = \frac{(s + i\delta)c_1(0) - \rho e^{i\pi/4} \sqrt{s}}{D(s)},
\] (D.24)

\[
\tilde{h}_2(s) = \frac{(s - i\delta)c_2(0) + \rho e^{i\pi/4} \sqrt{s}}{D(s)},
\] (D.25)

where

\[
\rho = \alpha [c_2(0) - c_1(0)],
\] (D.26)

\[
D(s) = s^2 + 2\alpha e^{i\pi/4} \sqrt{s} - \delta^2 \prod_{j=1}^4 (\sqrt{s} - e^{i\pi/4} v_j),
\] (D.27)

and \( \alpha \) is defined in Eq. (3.61). Here \( v_j (j = 1, ..., 4) \) are the roots of the quartic equation

\[ x^4 + 2\alpha x^3 - \delta^2 = 0, \] (D.28)
Eq. (D.33) shows that the quantity $u$ is always negative. Thus $\xi_1$ and $\sigma_{1,2}$ are positive whereas $\xi_2$ is negative. Thus, the roots

$$v_{1,3} = -|\sigma_1|/2 \pm \sqrt{(\sigma_1/2)^2 + \xi_2},$$

(Eq. D.29)

$$v_2 = v_4^* = -\sigma_2/2 - i\sqrt{\xi_1 - (\sigma_2/2)^2},$$

(Eq. D.30)

$$\sigma_{1,2} = \alpha \pm \sqrt{\alpha^2 + u},$$

(Eq. D.31)

$$\xi_{1,2} = u/2 \pm \sqrt{(u/2)^2 + \delta^2},$$

(Eq. D.32)

$$u = -(2\alpha^2\delta^2)^{1/3} \left[(A + 1)^{1/3} - (A - 1)^{1/3}\right],$$

(Eq. D.33)

$$A = \left[1 + (4/27)(2\delta^2/\alpha^2)^6\right]^{1/2}.$$  

(Eq. D.34)

Eq. (D.33) shows that the quantity $u$ is always negative. Thus $\xi_1$ and $\sigma_{1,2}$ are positive whereas $\xi_2$ is negative. Thus, the roots

$$v_{1,3} = -|\sigma_1|/2 \pm \sqrt{(\sigma_1/2)^2 + |\xi_2|}$$

are both real. Moreover numerical analysis shows that $\xi_1 - (\sigma_1/2)^2 \geq 0$ for all $\delta$, the equality sign holding for $\delta = 0$, that is, when the upper levels $|2\rangle$ and $|1\rangle$ are degenerate (The effective mass approximation means that $\delta$ cannot be too large). Thus, Eq. (D.30) can be re-written as

$$v_2 = v_4^* = -\sigma_2/2 - i|\xi_1 - (\sigma_2/2)^2|.$$  

(Eq. D.36)

This shows that the roots $v_2$ and $v_4$ are complex conjugates of each other. Eqs. (D.24) and (D.25) can now be inverted [following the procedure described in Appendix (C)] to give

$$c_2(t) = \sum_{j=1}^{2} S_j T_j e^{i(v_j^2 + \delta)t} + \frac{e^{i\pi/4}}{\pi} \int_0^\infty \frac{f_2(x) e^{-(x-i\delta)t} dx}{W(x)},$$

(Eq. D.37)

$$c_1(t) = \sum_{j=1}^{2} S_j U_j e^{i(v_j^2 - \delta)t} + \frac{e^{i\pi/4}}{\pi} \int_0^\infty \frac{f_1(x) e^{-(x+i\delta)t} dx}{W(x)},$$

(Eq. D.38)

where

$$S_j = \frac{2v_j}{(v_j - v_l)(v_j - v_m)(v_j - v_n)}, \quad (l, m, n = 1, ..., 4, j \neq l \neq m \neq n).$$  

(Eq. D.39)
\[ T_j = (v_j^2 - \delta)c_2(0) + v_j \rho = (v_j^2 + \alpha v_j - \delta)c_2(0) - \alpha v_j c_1(0), \quad (D.40) \]
\[ U_j = (v_j^2 + \delta)c_2(0) - v_j \rho = (v_j^2 + \alpha v_j + \delta)c_1(0) - \alpha v_j c_2(0), \quad (D.41) \]
\[ f_2(x) = \left[ -\rho(x^2 + \delta^2) + 2\alpha c_2(0)(x + i\delta)x \right] \sqrt{x}, \quad (D.42) \]
\[ f_1(x) = \left[ \rho(x^2 + \delta^2) + 2\alpha c_1(0)(x - i\delta)x \right] \sqrt{x}, \quad (D.43) \]
\[ W(x) = (x^2 + \delta^2)^2 + i4\alpha^2 x^3. \quad (D.44) \]
Appendix E

Two-level Atom in the Anisotropic Model

In this appendix we consider a special case of our model three-level system when single photon spontaneous emission for the transition $|1\rangle \rightarrow |0\rangle$ is assumed forbidden. This means that the population of level $|1\rangle$ cannot decay directly to level $|0\rangle$. Its only decay mechanism is indirectly through level $|2\rangle$ via the coupling $\Omega$. Now, if the upper levels $|2\rangle$ and $|1\rangle$ are not driven by a control laser field so that $\Omega = 0$, level $|1\rangle$ will be completely decoupled from the rest of the system and our model system of Fig. (3.1) is effectively a two level system consisting of levels $|2\rangle$ and $|0\rangle$.

In this case Eqs. (3.39) and (3.40) reduce (assuming that the atom is initially on the upper level $|2\rangle$ so that $\theta = 0$) to $\tilde{c}_1(s) = 0$ and

$$\tilde{c}_2(s) = \frac{1}{s + \tilde{G}(s)}. \quad (E.1)$$

Using Eq. (3.62) for $\tilde{G}(s)$ we obtain

$$\tilde{c}_2(s + i\delta) = 1/D(s), \quad (E.2)$$
where

\[ D(s) = s + \alpha e^{i\pi/4} \sqrt{s} + i\delta = \prod_{j=1}^{2} \left( \sqrt{s} - e^{i\pi/4} v_j \right). \]  \hspace{1cm} (E.3)

Here \( v_j(j = 1, 2) \) are the roots of the quadratic equation

\[ x^2 + \alpha x + \delta = 0 \]  \hspace{1cm} (E.4)
given by

\[ v_{1,2} = -\frac{\alpha}{2} \pm \sqrt{\left( \frac{\alpha}{2} \right)^2 - \delta}. \]  \hspace{1cm} (E.5)

The amplitude \( c_2(t) \) is found from the inverse Laplace transform of \( c_2(s + i\delta) \) through the inversion integral of Eq. (C.1). Following the method of Appendix (C) and using the contour of Fig. (C.1) yields the following results:

(a) If \( \delta < 0 \) (upper level \( \mid \delta \rangle \) is inside the gap), the roots (E.5) are given by

\[ r_{1,2} = -\frac{\alpha}{2} \pm \sqrt{\left( \frac{\alpha}{2} \right)^2 + |\delta|}. \]  \hspace{1cm} (E.6)

Thus \( r_1 \) is positive, whereas \( r_2 \) is negative and lies outside the contour of integration. In this case we obtain

\[ c_2(t) = c_1 e^{i(r_1^2 + \delta)t} + I(\delta, t), \hspace{0.5cm} \delta < 0, \]  \hspace{1cm} (E.7)

where

\[ c_1 = 2r_1/(r_1 - r_2), \]  \hspace{1cm} (E.8)

\[ I(\delta, t) = \frac{\alpha e^{i\pi/4}}{\pi} \int_0^\infty \frac{\sqrt{x}e^{(-x+i\delta)t}}{(-x + i\delta)^2 + i\alpha^2 x} dx. \]  \hspace{1cm} (E.9)

(b) If \( 0 \leq \delta \leq (\alpha/2)^2 \), both roots \( v_{1,2} \) are negative and lie outside the contour of integration. In this case we obtain

\[ c_2(t) = I(\delta, t), \hspace{0.5cm} 0 \leq \delta \leq (\alpha/2)^2, \]  \hspace{1cm} (E.10)

where \( I(\delta, t) \) is given by Eq. (E.9).
(c) If $\delta > (\alpha/2)^2$, the roots (E.5) are complex conjugates of each other given by

$$q_1 = q_2^* = -\frac{\alpha}{2} - i\sqrt{|\delta| - \left(\frac{\alpha}{2}\right)^2}. \quad (E.11)$$

In this case the residue corresponding to $q_2$ is zero, and we obtain

$$c_2(t) = d_1 e^{i(q_1^2 + \delta)t} + I(\delta, t), \quad \delta > (\alpha/2)^2, \quad (E.12)$$

where

$$d_1 = \frac{2q_1 \cos \theta}{q_1 - q_2}. \quad (E.13)$$

Since the root $r_1$ is positive while the root $q_1$ is complex with a negative real part, the first term on the RHS of Eq. (E.7) is a non-decaying oscillatory term while that of Eq. (E.12) decays in time and tends to zero as $t \to \infty$. The term $I(\delta, t)$ also decays in time and tends to zero as $t \to \infty$. The steady-state population on the upper level $|2\rangle$ is then given by

$$n_{2\text{s}} \equiv \lim_{t \to \infty} |c_2(t)|^2 = \begin{cases} \frac{4r_1^2}{(r_1 - r_2)^2} & \text{if } \delta < 0 \\ 0 & \text{if } \delta \geq 0. \end{cases} \quad (E.14)$$

Thus for the two level system (consisting of the ground level $|0\rangle$ and the excited level $|2\rangle$) placed inside a PBG structure described by the "effective mass" anisotropic dispersion relation Eq. (3.58), fractionalized steady state inversion occurs only for $\delta < 0$, that is for $\omega_2 < \omega_c$. On the other hand, for such a two-level system in the isotropic model Eq. (3.57), fractionalized inversion was shown[65] to occur even when $\omega_2$ is slightly greater than $\omega_c$ (that is, even when the excited state lies outside (but not far from) the band gap).
Appendix F

Photon Density of states

The photon density of states in a medium is defined by

\[ \rho(\omega) = \frac{1}{L^d} \sum_k \delta (\omega - \omega(k)) , \]  

(F.1)

where \( L^d \) is the sample volume in \( d \)-dimensional space and \( \omega(k) \) is the photon dispersion relation in the medium. In the limit of large \( L \), we may replace the sum by the integral:

\[ \rho(\omega) = \frac{1}{(2\pi)^d} \int d^d k \, \delta (\omega - \omega(k)) . \]  

(F.2)

When the dispersion relation is isotropic (that is, when \( \omega(k) \) depends only on the magnitude \( k \) of \( k \)) we can perform the angular integration in Eq.(F.2) to obtain

\[ \rho(\omega) = \frac{S_d}{(2\pi)^d} \int k^{d-1} \, dk \, \delta (\omega - \omega(k)) . \]  

(F.3)

where \( S_d \) is the surface area of a unit sphere in \( d \)-dimensions (\( S_d = 2, 2\pi \) or \( 4\pi \) for \( d = 1, 2 \) or 3, respectively). Using Eq. (5.12) in Eq. (F.3), we obtain

\[ \delta (\omega - \omega(k)) = z(\omega) \delta (k - k(\omega)) . \]  

(F.4)
Inserting this in Eq. (F.3) we finally obtain

$$\rho(\omega) = \frac{S_d}{(2\pi)^d} k^{d-1}(\omega) z(\omega)$$  \hspace{1cm} (F.5)$$

for the density of states in $d$-dimensions.

In free space $k(\omega)$ and $z(\omega)$ are given by Eqs. (3.42), and (5.35) and using them in Eq. (F.5) we obtain

$$\rho(\omega) = \frac{\omega^2}{2\pi^2}, \quad (d = 3)$$  \hspace{1cm} (F.6)$$

for the free space photon density of states in three dimensions.

For a PBG, we use the dispersion relation (5.74) in Eq. (5.12), and obtain

$$z(\omega) = \frac{|\omega - \omega_0|}{\sqrt{(\omega - \omega_u)(\omega - \omega_c)}}.$$  \hspace{1cm} (F.7)$$

Clearly, $z(\omega)$ exhibits square root singularities at the band edge frequencies $\omega_u$ and $\omega_c$. From Eqs. (5.74) we obtain

$$k(\omega) = \sqrt{\omega_u \omega_d} \pm \sqrt{(\omega - \omega_c)(\omega - \omega_u)}$$  \hspace{1cm} (F.8)$$

where the $+-(-)$ sign applies for $\omega > \omega_c$ ($\omega < \omega_u$). Eq. (F.8) shows that, for the frequency range $G \equiv (\omega_u, \omega_c)$ spanning the gap, the wave vector $k(\omega)$ is complex, indicating that the wave is non-propagating. Outside the PBG (that is, for $\omega \notin G$) Eqs. (F.7), (F.8) and (5.12) give

$$\rho(\omega \notin G) = \frac{|\omega - \omega_0|}{2\pi^2} \left[ \frac{\omega^2 - 2\omega_0\omega + 2\omega_u\omega_c}{\sqrt{(\omega - \omega_u)(\omega - \omega_c)}} \pm 2\sqrt{\omega_u\omega_c} \right],$$  \hspace{1cm} (F.9)$$

where the $+-(-)$ sign applies for $\omega > \omega_c$ ($\omega < \omega_u$).

For a dispersive medium, described by the dispersion relation (5.84), we obtain

$$k(\omega) = \omega \left( \frac{\omega - \omega_c}{\omega - \omega_u} \right),$$  \hspace{1cm} (F.10)$$

where $\omega_u = \omega_L - \Delta$ and $\omega_c = \omega_L$ are the band-edge frequencies. Using (F.10) in Eq. (5.12),
we obtain
\[ z(\omega) = \frac{\omega^2 - 2\omega_0 \omega + \omega_0 \omega_c}{(\omega - \omega_0)^2} \]  \hspace{1cm} (F.11)
for the form factor of a frequency dispersive medium. Comparing Eqs. (F.11) and (F.7) we notice that, unlike in the PBG case where \( z(\omega) \) is singular at both band edge frequencies, in the FDM case \( z(\omega) \) is singular only at the lower band edge frequency \( \omega_v \). Moreover, the singularity at \( \omega_v \) is much stronger \( z(\omega) \sim (\omega - \omega_v)^{-2} \) than that in the PBG case.

The wave number \( k \), being the magnitude of a vector \( (k) \), is positive definite. However, Eq. (F.10) shows that, for frequencies \( \omega \in G \), \( k(\omega) \) is negative, indicating that this range corresponds to the PBG region. Outside the photonic band gap, Eqs. (F.10), (F.11) and (5.12) give
\[ \rho(\omega \not\in G) = \frac{\omega(\omega - \omega_c)^2}{2\pi^2} \left[ \frac{\omega^2 - 2\omega_0 \omega + \omega_0 \omega_c}{(\omega - \omega_0)^4} \right] \]  \hspace{1cm} (F.12)
for the density of states in a frequency dispersive medium.
Bibliography


[107] O. Toader and S. John, to be published.


[114] N. M. Lawandy and G. Kweon, “Molecular and free electron spontaneous emission in periodic three-dimensional dielectric structures,” in Ref.[23].


