Quantum Theory of Phonon-Mediated Decoherence and Relaxation of Two-Level Systems in a Structured Electromagnetic Reservoir

by

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Abstract

In this thesis we study the role of nonradiative degrees of freedom on quantum optical properties of mesoscopic quantum dots placed in the structured electromagnetic reservoir of a photonic crystal. We derive a quantum theory of the role of acoustic and optical phonons in modifying the optical absorption lineshape, polarization dynamics, and population dynamics of a two-level atom (quantum dot) in the “colored” electromagnetic vacuum of a photonic band gap (PBG) material. This is based on a microscopic Hamiltonian describing both radiative and vibrational processes quantum mechanically. Phonon sidebands in an ordinary electromagnetic reservoir are recaptured in a simple model of optical phonons using a mean-field factorization of the atomic and lattice displacement operators. Our formalism is then used to treat the non-Markovian dynamics of the same system within the structured electromagnetic density of states of a photonic crystal. We elucidate the extent to which phonon-assisted decay limits the lifetime of a single photon-atom bound state and derive the modified spontaneous emission dynamics due to coupling to various phonon baths. We demonstrate that coherent interaction with undamped phonons can lead to enhanced lifetime of a photon-atom bound state in a PBG by (i) dephasing and reducing the transition electric dipole moment of the atom and (ii) reducing the quantum mechanical overlap of the state vectors of the excited and ground state (polaronic shift). This results in reduction of the steady-state atomic polarization but an increase in the fractionalized upper state population in the photon-atom bound state. We demonstrate, on the other hand, that the lifetime of the photon-atom bound state in a PBG is limited by the lifetime of phonons due to lattice anharmonicities (break-up of phonons into lower energy phonons) and purely nonradiative decay. We demonstrate how these additional damping effects limit the extent of the
polaronic (Franck-Condon) shift of the atomic excited state. We also derive the modified polarization decay and dephasing rates in the presence of such damping. This leads to a microscopic, quantum theory of the optical absorption lineshapes. Our model and formalism provide a starting point for describing dephasing and relaxation in the presence of external coherent fields and multiple quantum dot interactions in electromagnetic reservoirs with radiative memory effects.
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Chapter 1

Introduction

1.1 Mesoscopic quantum optics in photonic crystals

The interaction of small systems (few degrees of freedom) with a reservoir consisting of large number of degrees of freedom (possibly infinite) is a central paradigm in physics and arises in numerous areas of research. In quantum optics examples of small systems are simple atoms (modelled as two-level or multilevel systems) and the large reservoir consists of the radiation field of an unstructured electromagnetic reservoir. Recent advances in the synthesis of ultra-high quality photonic crystals and 3D photonic band gap (PBG) materials have given rise to the possibility of engineering atom-photon interactions resulting in a structured electromagnetic reservoir. One of the dramatic consequences of tailoring atom-photon interaction[1, 2] in PBG materials is the inhibition of spontaneous emission of light from atoms[3, 4, 5, 6, 7] and the formation of photon-atom bound state[8, 9, 10, 11, 12]. When the atomic resonance lies very close to a photonic band edge or other sharp features in the electromagnetic density of states, the radiative dynamics can exhibit long time memory (non-Markovian) effects. Important new phenomena such as the ability to population-invert a two-level system by coherently pumping on resonance have been suggested[13, 14, 15, 16, 17, 18]. These effects rely on the microfabrication of PBG materials with resolution on the scale of a few nanometers in the vicinity of a quantum dot. Applications of the photon-atom bound state to quantum information processing have also been proposed[19, 20] and experimental studies of quantum entanglement of photons propagating through photonic crystals have recently been initiated[21, 22]. Their successful implementation rely on robust control of various decoherence channels (interaction with radiative and lattice degrees of freedom).
present in these mesoscopic systems[23, 24]. Decoherence is a fundamental obstacle to quantum information processing and refers to the loss of entanglement due to interaction of the system with external reservoirs. The interaction of two closely spaced atoms is also modified in the structured reservoir of a photonic crystal[25, 26, 27] which can have applications in construction of qubits for quantum information processing[28, 29, 19, 20].

Experimental verification of theoretical predictions has been made possible due to developments in fabrication of photonic crystals and embedding them with light emitters[30, 31, 32, 5]. Photonic crystals can modify the radiative decay rate of atoms due to the change in local photonic density of states at the position of the embedded light sources[4, 32]. The experiment[4] was performed using CdSe nanocrystals as light sources in titania inverse opal photonic crystals. CdSe nanocrystals typically have a diameter between 3 nm and 6 nm and show spectral features associated with quantum confinement. The colour of the emission spectra depends strongly on the size of the CdSe nanocrystal with pronounced red shift of the spectrum with increasing nanocrystal size due to the change in the excitonic energy levels. When these nanocrystals were embedded inside a photonic crystal, the lifetime of the nanocrystals was found to change by a factor-of-three depending on the lattice parameter of the photonic crystal. Changing the lattice parameter of the photonic crystal varies the local photonic density of states in the vicinity of the resonant frequency of the nanocrystal. If the local photonic density of states increases relative to the free space density of states, the nanocrystal undergoes faster radiative decay and vice versa.

Quantum optics in photonic crystal structures is strongly influenced by the local photonic density of states in the vicinity of the atomic transition frequency and can be classified into two regimes. The photonic density of states is suppressed inside the photonic band gap which results in reduced atom-photon coupling leading to reduced decoherence for any quantum information theoretic application[6]. Introduction of defects in the photonic band gap can also create localized modes. These localized defect modes can be used as qubits to store quantum information. They can also be designed to obtain very high quality factors and small modal volumes which can lead to lasing in the presence of a suitable gain medium. Linear defects embedded in photonic crystals can serve as waveguides for integrated optics[22]. Embedded point defects can also act as microcavities[33, 34].

Outside the band gap there is a large enhancement of the photonic density of states relative to the unstructured electromagnetic reservoir. This enhanced local density of
states can result in superradiance if the resonant frequency of any embedded light source falls in this region. It can dramatically increase the Purcell factor raising the intriguing possibility of strong-coupling regime of cavity quantum electrodynamics (QED)[35, 36, 37, 38, 39, 40]. This has lead to enormous research and given rise to the rapidly emerging field of solid-state cavity QED[41, 30, 42, 43, 44] with excitons localized in the embedded quantum dots acting as atoms. These systems are also of importance for construction of single photon sources[45] on demand with substantially improved efficiency and speed[46, 47]. They can also be used as entangled photon sources on demand. Consequently these systems offer enormous potential for quantum optics on a chip. These semiconductor quantum dots, also called artificial atoms, can be effectively described by a few degrees of freedom. They also interact with the underlying solid-state lattice which can modify their optical properties. Consequently the combined interaction of these semiconductor nanostructures with the radiation field of a structured electromagnetic reservoir and lattice vibrations provide new challenges in quantum optics[48, 49]. In the light of these developments it is timely to develop and study analytical tools applicable to the study of mesoscopic quantum optics in photonic crystals.

In this thesis we study in detail the role of nonradiative degrees of freedom on quantum optical properties of mesoscopic quantum dots placed in the structured electromagnetic reservoir of a photonic crystal. Two standard approaches to this problem either involve a stochastic description of dephasing or a semiclassical approach to the optical lineshape (see Chapter 2). In the stochastic description of dephasing, the interaction of the quantum dot with phonons is treated as a modulation of the two-level system transition frequency. In the semiclassical approach to the optical lineshape problem, the lattice modes are treated quantum mechanically assuming a parabolic adiabatic potential for the phonons. Following second quantization the lattice modes are described as quantized harmonic oscillators. We provide a full quantum mechanical treatment of the role of phonons and photons and elucidate in detail the role of lattice degrees of freedom in modifying the optical absorption lineshape, polarization dynamics, and population dynamics of a two-level atom (quantum dot). This is based on a microscopic Hamiltonian which includes both radiative and vibrational processes. Previous results of the optical lineshape problem based on semiclassical arguments[50] are rederived in the homogenous reservoir of free space using a simple model of optical phonons and a mean-field factorization of the atomic and lattice displacement operators. We then use our formalism to describe non-Markovian dynamics of the same system within the structured electro-
Chapter 1. Introduction

magnetic density of states of a photonic crystal. We provide a detailed analysis of the effects of phonon-assisted decay processes on the lifetime of a single photon-atom bound state and also derive the modified spontaneous emission dynamics due to coupling to various phonon baths. We find that the interaction of the quantum dot with undamped phonons can increase the lifetime of a photon-atom bound state in a photonic band gap material. However this description is incomplete and additional processes need to be incorporated to obtain an experimentally consistent picture. We find that the lifetime of the photon-atom bound state in a photonic band gap material is limited by the lifetime of phonons and purely nonradiative decay of the excited electronic state. Lattice anharmonicities and higher order phonon processes damp the phonon modes which in turn modify the quantum dot dynamics. All these processes provide a microscopic and consistent quantum mechanical theory of the optical lineshape which is valid in a general electromagnetic reservoir.

The theoretical predictions made in this thesis are also experimentally verifiable. Using microscopic considerations for the physically relevant acoustic phonon models (in the presence of phonon-assisted decay of the excited state and finite phonon damping), we obtain explicit expressions for polarization and population decay rates of a single self-assemble quantum dot. For example, in the case of phonon damping, the lifetime of phonons is described by the Landau-Rumer process of relaxation of a transverse acoustic mode into a thermal longitudinal mode. The phonon damping rate can now be evaluated for the quantum dot in consideration which gives specific decay rates of polarization and population dynamics of the excited state. These decay rates can now be compared to experimental data. Similar considerations also apply to the case of direct nonradiative decay. Other quantities of interest for experimental verification are the temperature dependence of the polaron shift and the width of the absorption spectrum. Finally we note that the general formalism presented in this thesis also provides a starting point for describing dephasing and relaxation in the presence of external coherent driving fields. It can also be used to discuss multiple quantum dot interactions in electromagnetic reservoirs with radiative memory effects.

1.2 Quantum dots

In this section we discuss important features and physical characteristics of semiconductor quantum dots which have made them very attractive for applications in the rapidly
Figure 1.1: Density of states $\rho_{3D}(E)$ of a three-dimensional bulk semiconductor, $\rho_{2D}(E)$ of a quasi two-dimensional quantum well, $\rho_{1D}(E)$ of a quasi one-dimensional quantum wire and a quasi zero-dimensional quantum dot structure $\rho_{0D}(E)$. The density of states as a function of energy are $\rho_{3D}(E) \propto E^{1/2}$, $\rho_{2D}(E) \propto \sum_j \Theta(E - E_j)$ where $E_j$ are the energy eigenvalues due to confinement along one direction, $\rho_{1D}(E) \propto \sum_k (E - E_k)^{-1/2}$ where $E_k$ are the energy eigenvalues due to confinement along two directions and $\rho_{0D}(E) \propto \sum_i \delta(E - E_i)$ where $\Theta(E - E_j)$ is the step function and $\delta(E - E_i)$ is the Dirac-delta distribution.
emerging field of mesoscopic quantum optics[51]. Semiconductor quantum dots have very sharp nearly \( \delta \)-function like electronic density of states. The charge carriers in quantum dots are excitons (electron-hole bound pairs) confined in all three directions which is effectively a zero-dimensional system. The confinement region has typical lateral dimensions of 1 to 30 \( nm \) corresponding to about \( 10^3 \) to \( 10^6 \) atoms. Confinement of the charge carriers results in discretization of the energy spectrum with primary level separation of the order of 10 to 100 meV[52]. Quantum dots exhibit optical spectra with very narrow linewidths and also have very high optical nonlinearity in contrast to higher-dimensional heterostructures. In Fig(1.1) we illustrate the density of states of a bulk semiconductor and semiconductor heterostructures of different dimensions.

The exciton energy spectra and the primary level splitting depend on the boundary conditions on the exciton wave function. An important parameter which characterizes quantum dots is the ratio of the characteristic carrier confinement length \( L \) to the exciton Bohr radius \( a_B \) of the exciton in the bulk semiconductor material. Two possible regimes of carrier confinement are relevant[52]. In the strong confinement regime, characterized by \( L/a_B << 1 \), carriers are confined in small pyramidal or lens-shaped islands with typical extensions of \( 10 \times 10 \times 5nm^3 \). In the opposite limit, the weak confinement regime, the carriers are localized at monolayer fluctuations of a semiconductor quantum well. Carriers in the weak confinement regime are confined in a region of volume \( 100 \times 100 \times 5nm^3 \).

Primary level splitting (transition energy from a single exciton state to the state with no excitons) in weakly confined quantum dots is \( 0.1 \) – \( 1meV \) and \( 10 \) – \( 100meV \) in strongly confined quantum dots. Exciton decoherence time in weakly confined quantum dots as measured using coherence spectroscopy is 40ps. Exciton decoherence time in strongly confined quantum dots is of the order of nanoseconds as measured using four wave mixing and is 750ps from coherence spectroscopy measurements[52]. Such long decoherence times are unusual in bulk semiconductors and other finite-dimensional semiconductor heterostructures. This is attributed to the sharp atom-like spectra and strong suppression of carrier-carrier and carrier-phonon scatterings in semiconductor quantum dots due to reduced phase space available for energy and momentum conservation.

Self-assembled quantum dots are produced by self-organization[53] using molecular beam epitaxy[54] or metal-organic chemical vapour deposition. The underlying process is the Stranski-Krastanov transition where strain relaxation due to lattice misfit between the monocrystal substrate and the growing layer results in phase separation (first-order transition). The pressure exerted on the growing monolayer leads to a strain-relaxed
Figure 1.2: Schematic sketch of (a) weak and (b) strong confinement regime of carriers in a semiconductor quantum dot. In the weak confinement regime the carriers are usually confined in a 2-dimensional quantum well layer with small fluctuations in the other direction. Typical sizes of these regions are $100 \times 100 \times 5 nm^3$. In the strong confinement regime, the carriers are tightly confined into a much smaller region of lower-bandgap material embedded in a larger bandgap material. These islands are usually pyramidal or lens-shaped and have sizes of the order of $10 \times 10 \times 5 nm^3$.

arrangement and the monolayer breaks up into crystallite islands. Useful examples of materials with lattice mismatch which can be used for the growing layer and the underlying substrate are InAs/GaAs and InP/InGaP. The lattice misfit values for InAs/GaAs is 7 percent and InP/InGaP is 4 percent[55]. For the case of InAs/GaAs, the monolayer of InAs grows as a strained 2D layer followed by the Stranski-Krastanov transition when the InAs layer exceeds 1.5 monolayers. The resulting quantum dot size and shape and its associated optical properties depend on factors such as temperature, growth rate and amount of growth material. Variations in size and shape are unavoidable which can lead to considerable inhomogeneity in the optical properties due to subtle changes in the excitonic energy levels of the quantum dot. Self-assembled quantum dots are well-suited for single dot spectroscopy as the dots are grown in a single layer. Moreover, the quantum dot density can be controlled over a wide range by changing the growth conditions.

We note that the analogy between free atoms and quantum dots, though useful, has serious limitations and it is important to put them in perspective. These differences originate primarily from the fact that semiconductor quantum dots are solid state devices[56]. A single quantum dot consists of thousands of atoms arranged in a finite size lattice embedded in a large bandgap host semiconductor. In contrast to free atoms which are bound
mainly by Coulomb interaction, quantum dots also interact with the lattice degrees of freedom. Unlike free atoms, interaction with phonon modes can substantially modify the optical spectra of quantum dots which typically show up as broad multiphonon resonances also known as phonon sidebands. Other important differences between quantum dots and atoms lie in the physical origin of eigenenergies and degeneracies of the eigenvalue spectrum. For example, consider the Hydrogen atom which consists of an electron and a proton bound by Coulomb interaction. The energy levels follow precisely from the solution of the Schrodinger equation and the spectrum of the emitted light (due to transition between energy levels) is very sharp (apart from the broadening of the energy levels due to quantum fluctuations of the electromagnetic vacuum). On the other hand, discretization of the energy spectrum of a quantum dot results from carrier confinement and size quantization[57]. In any given ensemble, size fluctuations in quantum dots are inevitable since nucleation and crystal growth happen in conditions removed from thermodynamic equilibrium and involve multiple kinetic processes. Fluctuations in sizes of quantum dots in an ensemble lead to inhomogeneous broadening effects in the optical spectra. In addition, the temperature dependence of the band gaps of the underlying semiconductor results in a thermal drift of the resonant energies of the quantum dots. The shell structure of electronic levels and their degeneracies also depend on the shape of the quantum dot and variations in shape can induce additional inhomogeneities in the ensemble. These effects show up in the optical lineshape and the fluorescence spectra[58, 59, 60, 61]. In the following section we discuss the interaction of quantum dots with the radiative and lattice degrees of freedom.

1.3 Interaction of quantum dot with radiation and lattice degrees of freedom

Semiconductor quantum dots have found numerous applications in applied research[62, 63]. Among these is the proposal to use them as physical qubits for quantum information processing. Quantum dots are regarded as "artificial atoms" in which information can be stored in the charge or spin degree of freedom and coherently driven by high electric fields[64, 65]. Dephasing times for excitons in self-assembled quantum dots at low temperatures are known to be radiatively limited (dephasing rate is half the relaxation rate as in the simple case of a two-level system interacting only with an unstructured
electromagnetic reservoir of free space) suggesting that the long time (\( \sim ns \)) decoherence is due to radiative dephasing. This allows for sufficiently high numbers of coherent manipulations with picosecond pulses making them ideal systems for quantum information processing. It is also important to consider important sources of decoherence in these systems which can effect their use as qubits. The primary source of decoherence results from the interaction of the quantum dot with the phonon degrees of freedom. Another major source of decoherence is the decay of an excited quantum dot due to radiative recombination of the electron-hole pair and emission of a photon. The recombination rate can be controlled by embedding them in photonic crystals or microresonators[66]. Quantum dots can also be grown into the host dielectric structure of the photonic crystal backbone. In this thesis, we consider in detail a single quantum dot embedded deep inside a photonic crystal with a transition frequency in the vicinity of the band edge of the photonic band gap. We compute its optical lineshape and describe in detail the role of phonons in its observed spectrum[67, 68, 69]. We also discuss the polarization and population dynamics of the quantum dot. In this section we provide a brief summary of some important aspects of the interaction of quantum dots with radiation and phonon degrees of freedom. We discuss in detail model Hamiltonians and provide a framework to include relevant physical processes which are central to the main part of the thesis.

### 1.3.1 Quantum dot photon interaction

The optical excitation (exciton) in a quantum dot can be considered as a two-level system where the excited state corresponds to the presence of one exciton (electron-hole pair) and the ground state corresponds to an empty dot (no electron and hole in the quantum dot). The two-level approximation is valid if the transition frequencies of the dot are well separated and the exciton dynamics is only confined to the two lowest energy levels. This has been experimentally demonstrated by optically exciting the two lowest electronic energy levels in single quantum dots and coherently driving them by applying external electromagnetic fields inducing the well known Rabi oscillations[64, 65]. Rabi oscillation here refers to the damped oscillation of the coherence of an initially excited atom due to an externally applied electromagnetic field. The two-level model is a widely used paradigm in quantum optics and laser spectroscopy[70] and has also been utilized in various models in condensed matter and, more recently, in the study of decoherence and dephasing in the context of quantum information processing[71, 52]. It is also a useful
model to describe the spectral signatures of simple systems interacting with lattice modes
in diverse areas of chemical physics and quantum dissipative systems [72, 73, 74].

If a quantum dot is modelled as a two-level atom with sharp energy spectrum, the
optical lineshape in an unstructured electromagnetic reservoir is a Lorentzian (in units
of \( \hbar = 1 \)):

\[
\chi(\omega) = \chi_0 \frac{\Gamma_0(\omega)}{2\pi (\omega - \omega_0)^2 + \left(\frac{\Gamma_0(\omega_0)}{2}\right)^2}
\]  

(1.1)

where \( \chi_0 \) is a constant and \( \Gamma_0(\omega_0) = 1/\tau_{\text{rad}} \) where \( \tau_{\text{rad}} \) is the radiative lifetime. The
radiative lifetime \( \tau_{\text{rad}} \) depends on the size, shape and material properties of the quantum
dot and is in the nanosecond range in free space for Ga(In)As/GaAs dots. Therefore the
optical lineshape has energy width \( \Gamma_0(\omega_0) \approx 1\mu eV \).

In the case of quantum dot ensembles, the optical lineshape associated with sharp
atom-like spectra of single quantum dots is washed out by inhomogenous broadening.
Assuming that every dot has its own intrinsic transition frequency \( \omega_0 \), the optical line-
shape \( \chi(\omega) \) is now given by an average over a probability density \( P(\omega_0 - \langle \omega_0 \rangle) \) that the
dot has a transition frequency \( \omega_0 \):

\[
\langle \chi(\omega) \rangle = \int d\omega_0 \chi(\omega) P(\omega_0 - \langle \omega_0 \rangle)
\]

(1.2)

The probability function \( P(\omega) \) is usually a Gaussian function which is broader than the
Lorentzian spectrum of a single quantum dot. The averaged optical lineshape \( \langle \chi(\omega) \rangle \) is
also a Gaussian with an energy width \( \hbar \Delta \omega \approx 50\text{meV} \) in Ga(In)As/GaAs dots.

Due to the detrimental effects of inhomogenous broadening, any plausible application
of quantum dot hardware design and quantum control rely strongly on single quantum
dot spectroscopy [75]. Moreover, for the robust analytical understanding of the funda-
mental optical properties of quantum dots, one has to eliminate all spurious signals which
smear the single quantum dot optical spectra. However, single dot spectroscopy is an
experimental challenge which is at the forefront of experimental research. Novel methods
capable of distinguishing and isolating single constituents from an ensemble of quan-
tum dots are being developed. From an experimentalist’s perspective single quantum
dot spectroscopy is very similar to single molecule spectroscopy. Selection of a single
entity from an ensemble can be performed either in real space or in the frequency do-
main. Single quantum dot resolution is simplified in real space and depends on sample
preparation with low density ensembles of quantum dots being much easier to resolve.
One way to get access to a single quantum dot in real space is microscopy. Far-field methods like confocal microscopy are capable of spatial resolutions in the range of $1\mu m$ and more specialized versions like the solid immersion lens technique extend this range down to $0.25\mu m$. On the other hand near field methods, either performed with *insitu* nanoapertures or scanning probes, can extend this range down to $50\,nm$ in special cases. Also cathodoluminescence and STM (scanning tunneling microscope)-induced luminescence have been shown to be powerful tools, suitable to reach the limit of single dot spectroscopy[76].

We now briefly discuss the interaction of a quantum dot with the photonic reservoir as used in this thesis. Later on we demonstrate how this description can be used to obtain the optical lineshape Eq(1.1) including nonradiative effects. As discussed above, a quantum dot can be considered as a two-level system if only the two lowest states of the exciton energy manifold is considered. The Hilbert space of the two-level system is denoted by basis states $|g\rangle$ ($|e\rangle$) denoting the ground (excited) state and they obey completeness and orthonormality relations: $\sum_{i=g,e} |i\rangle \langle i| = I$ and $\langle i|j\rangle = \delta_{ij}$ where $I$ is the identity and $\delta_{ij}$ is the Kronecker delta. The state $|g\rangle$ contains no excitons and $|e\rangle$ is the excited state with one exciton. The two-level system operators (in the pseudospin representation) are denoted $\sigma_{ij}$ and obey the commutation relation:

$$[\sigma_{ij}, \sigma_{kl}] = \delta_{jk} \sigma_{il} - \delta_{il} \sigma_{kj} \quad (1.3)$$

The Hamiltonian of the two-level system $H_0$ can then be written as: $H_0 = \hbar \omega_e \sigma_{ee} - \hbar \omega_g \sigma_{gg}$. Using the completeness relation we rewrite the Hamiltonian as: $H = \hbar \omega_0 \sigma_{ee}$ where $\omega_0 = \omega_e - \omega_g$ and we have omitted the constant energy term since it does not affect the system dynamics.

The electromagnetic field in a volume $V$ (for example, volume of the Wigner-Seitz cell in a photonic crystal) following second quantization can be represented by a set of infinitely many discrete modes indexed by $k$ and the creation (annihilation) operators $\hat{a}_k^\dagger$ ($\hat{a}_k$) which obey the commutation relation:

$$[\hat{a}_k, \hat{a}_{k'}^\dagger] = \delta_{k,k'} \quad (1.4)$$
Figure 1.3: Schematic sketch of the ground state of a semiconductor nanocrystal as an excitonic two-level system. The state $|0\rangle$ is the ground state of the quantum dot where no excitons are present. $|1\rangle$ represents the excited state of the two-level atom with one exciton. Transitions between both states are dipole-allowed and the excited state can emit a photon of energy $\omega = \omega_e - \omega_g$ following electron-hole recombination and decay to the ground state. The two-level excitonic system can also be driven by strong optical fields resulting in Rabi oscillations.

The Hamiltonian for the free field is

$$H_{em} = \sum_k \hbar \omega_k \hat{a}_k^+ \hat{a}_k$$

(1.5)

where $\omega_k$ is the frequency of the $k^{th}$ mode. Detailed derivations of the quantization of the electromagnetic field can be found in numerous textbooks[70, 77]. The Hamiltonian of a two-level atom interacting with infinitely many oscillators of the radiation field can be shown to be given by[70, 77]:

$$H = \hbar \omega_0 \hat{\sigma}_{ee} + \sum_k \hbar \omega_k \hat{a}_k^+ \hat{a}_k + \hat{\sigma}_{eg} \sum_k \lambda_k (\hat{a}_k + \hat{a}_k^+) + \hat{\sigma}_{ge} \sum_k \lambda_k^* (\hat{a}_k + \hat{a}_k^+)$$

(1.6)

where the two-level atom radiation coupling constant $\lambda_k$ is given by:

$$\lambda_k = i \frac{\omega_0 |\vec{D}_{ge}|}{\hbar} (\frac{\hbar}{2 \epsilon_0 \omega_k V})^{\frac{3}{2}} \hat{e}_k \cdot \vec{u}_d$$

(1.7)

and $\vec{u}_d$ is a unit vector parallel to the atomic dipole moment $\vec{D}_{ge}$. $\hat{e}_k$ is the transverse polarization unit vector of the radiation field and $V$ is the quantization (sample) volume. Since the quantum dot-photon coupling constant is smaller than the atomic transition
frequency by several orders of magnitude we can assume that only transition that conserve energy are important. This is the rotating wave approximation[70, 77] following which Hamiltonian Eq(1.6) takes the form:

$$H = \hbar \omega_0 \sigma_{ee} + \sum_k \hbar \omega_k \hat{a}_k \hat{a}^\dagger_k + \sum_k (\lambda_k \sigma_{eg} \hat{a}_k + \lambda_k^* \hat{a}^\dagger_k \sigma_{ge})$$ (1.8)

### 1.3.2 Quantum dot phonon interaction

The interaction of quantum dots with electromagnetic radiation has been discussed in the previous subsection. We now present some important aspects of electron-phonon interaction in quantum dots. Unlike “natural” atoms, quantum dots interact strongly with lattice degrees of freedom which introduce additional structure in their optical line-shapes. Experimental evidence suggests that electron-phonon interaction in quantum dots is very different from that in bulk semiconductors and other semiconductor heterostructures. Primary method by which excited electrons relax in bulk semiconductors and quantum wells made of III-IV and II-VI semiconductors is through interaction with longitudinal optical phonons[57]. A secondary channel is through scattering with acoustic phonons either via the deformation potential or piezo-electric scattering[78, 79, 56]. Electron-phonon interactions in bulk semiconductors and quantum well heterostructures are efficiently computed in the Fermi-golden rule framework wherein matrix elements of the relevant transitions are computed and integrated over all the final states with identical energy as the initial one[80].

A straightforward application of the Fermi-golden rule framework to quantum dots predicts poor relaxation of carriers in quantum dots (phonon-bottleneck effect)[81, 82, 83, 84]. Phonon bottleneck effect follows directly from the application of the Fermi golden rule[81, 85, 86] to the electronic transitions in a quantum dot. For example, consider the case of optical phonons. The emission of a longitudinal optical phonon by an electron in a quantum dot is possible only if the electronic energy difference between the initial and final state is equal to the energy of the emitted phonon due to requirements of energy conservation. The negligible dispersion of longitudinal optical phonons and the rapidly decaying form factor of the electron phonon coupling with increasing \( \vec{k} \) negates the possibility of any longitudinal optical phonon emission due to energy non-conservation. However acoustic phonons have much broader spectrum but the deformation potential decreases rapidly with increasing phonon wavevectors. The emission of energetic acoustic
phonons is rapidly suppressed when the electron energy separation scale exceeds a few meV’s. Hence Fermi golden rule predicts that for a typical InAs/GaAs quantum dot, emission of either optical or acoustic phonons is energetically impossible and an excited electron cannot relax by mechanisms applicable in bulk semiconductors and quantum wells. This prediction of an inhibited relaxation is not in agreement with experiments.

Experimental studies (see [52] and references therein) show that for most types of self-assembled quantum dots, the primary level splitting (energy difference between the ground and excited state) is of the order of $10 - 100$ meV. Unlike in bulk semiconductors, decoherence does not proceed as a simple exponential decay where the rate can be determined by a few time constants. Typically at low temperatures ($< 20K$), the polarization in self-assembled quantum dots is found to decay in two stages: an initial rapid drop in the coherence which is non exponential and lasts a few picoseconds followed by an almost exponential decay. This results in strong non-Lorentzian optical lineshapes consisting of a strong relatively sharp line superimposed on a broad background (see Fig(1.4)).

The role played by phonons in quantum dots depends subtly on the energy difference between electronic levels. This can take the form of real acoustic phonon-assisted transitions which change the occupation of the energy levels or by virtual transitions which lead to pure dephasing or by combinations of both of them[88]. Phonon assisted transitions resulting in transfer of electrons between energy levels are of minor importance in quantum dots, especially in the strong confinement limit and at low temperatures, and is dominated by virtual processes[78]. Decoherence caused by carrier-phonon interactions that do not change the carrier occupation and do not cause electronic relaxation is known as pure dephasing. Decoherence in this scenario is attributed to the formation of a composite exciton-phonon excitation (polaron) and a rapid transfer of coherence from the quantum dot to the surrounding phonon cloud[89, 90].

A simple mathematical model to capture the dephasing of the quantum dot (in the absence of any relaxation) is given by the Independent – Boson model. The quantum dot is modelled as a two-level system and the phonon reservoir consists of a bath of harmonic oscillators. The two-level atom phonon interaction is turned on only when the system is in the excited state. The Hamiltonian (Independent – Boson model) is given by:

$$H = \omega_0 \hat{\sigma}_{ee} + \sum_{q} \Omega_q \hat{c}_q^\dagger \hat{c}_q + \hat{\sigma}_{ee} \sum_{q} \eta_q (\hat{c}_q + \hat{c}_q^\dagger)$$

(1.9)

The energy of the two-level atom is $\omega_0$ (in units of $\hbar = 1$) and the phonon dispersion is
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Figure 1.4: Plot of normalized luminescence spectra of a quantum dot in an unstructured reservoir as a function of temperature. The quantum dot is a CdTe structure grown by epitaxial methods. At 5K the spectrum has a Lorentzian profile with a linewidth of 180\( \mu \)eV. With increasing temperatures there is a noticeable redshift of the spectrum attributed to the leakage of the electronic wavefunction into the semiconductor shell surrounding the quantum dot core. Note the appearance of sidebands about the zero-phonon line with increasing temperatures. As temperature increases, the lineshape deviates from a Lorentzian and a broad background appears on both sides of the zero-phonon line. This broadband is due to exciton acoustic phonon interaction. At high temperatures the zero phonon line disappears into the acoustic phonon sideband. From [87].

\( \Omega_q, \eta_q \) is the two-level atom phonon coupling constant. The phonon operators \( \hat{c}_i (\hat{c}_i^\dagger) \) are bosonic and the two-level system operators obey the commutation relation Eq(1.3). It is clear from Hamiltonian Eq(1.9) that the phonons do not change the population of the atom (\( \hat{\sigma}_{ee} \) commutes with the Hamiltonian). However it still leads to decoherence (pure dephasing) as elucidated in the following discussion.

The simplest way to proceed further is to employ a canonical transformation which leads to a diagonal Hamiltonian \( \tilde{H} \):

\[
\tilde{H} = e^S H e^{-S} = (\omega_0 - \Delta)\hat{\sigma}_{ee} + \sum_q \Omega_q \hat{c}_q^\dagger \hat{c}_q
\]

(1.10)

where \( S = \hat{\sigma}_{ee} \sum_q \frac{m_q}{\Omega_q} (\hat{c}_q^\dagger - \hat{c}_q) \). \( \Delta = \sum_q \frac{\eta_q^2}{\Omega_q} \) is the polaron shift and accounts for the
renormalization of the two-level resonant frequency due to phonon absorption and emission. Since the Hamiltonian Eq(1.10) is diagonal it is now straightforward to calculate the dipole autocorrelation function (the lineshape function):

\[ \langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(0) \rangle = e^{-i(\omega_0-\Delta)t}e^{-(\Phi(0)-\Phi(t))} \]  

(1.11)

where \( \Phi(t) \) is defined as:

\[ \Phi(t) = \sum_q \frac{\eta_q^2}{\Omega_q^2} [(N_q + 1) \exp(-i\Omega_q t) + N_q \exp(i\Omega_q t)] \]  

(1.12)

and \( N_q = \langle \hat{c}_q^\dagger \hat{c}_q \rangle = [\exp(\beta\Omega_q) - 1]^{-1} \) (See Chapter 3 for more details).

We now evaluate the phonon correlation function for a specific model of acoustic phonons. We adopt a model used in earlier literature[52, 91] to describe GaAs-based quantum dots. Assuming a spherical dot model with acoustic deformation potential interactions, the dot-phonon coupling is:

\[ q = \left( \frac{\rho c_l}{D e D h} \right)^{1/2} \exp\left( -\frac{q^2 L^2}{4} \right) \]  

(1.13)

where \( \rho = 5379 \, kg \, m^{-3} \) is the mass density of GaAs, \( c_l = 5110 \, m/s \) is the longitudinal sound velocity, \( D_e = -14.6 \, eV \) and \( D_h = -4.8 \, eV \) the deformation potentials for electrons and holes and \( L = 5 \, nm \) is the electronic localization length in the quantum dot. The exponential in Eq(1.13) introduces an effective cutoff in \( q \). Hence an explicit cutoff at the Debye wavevectors is not required. Typical material parameters for GaAs with a quantum dot electronic localization length \( L = 5nm \) lead to characteristic time and energy (temperature) scales of \( \tau = L/c_l \approx 1ps \) and \( \hbar/\tau \approx 0.7meV \) (7.8K) respectively. For small \( q \)’s an effective linear dispersion relation \( \Omega_q = c_l q \) is assumed and Eq(1.12) for \( \Phi(t) \) is now given by:

\[ \Phi(t) = \alpha_p \int_0^{\infty} dx x \exp\left( -\frac{x^2}{2} \right) \left[ i \sin(x\bar{t}) + \cos(x\bar{t}) \coth\left( \frac{\hbar\beta x}{2\tau} \right) \right] \]  

(1.14)

where \( \bar{t} = \frac{t}{\tau} \) and the dimensionless dot-phonon coupling (Huang-Rhys factor) \( \alpha_p \) is defined as:

\[ \alpha_p = \frac{(D_e - D_h)^2}{4\pi^2\hbar^2c_l^3L^2} \approx 0.033 \]  

(1.15)
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Figure 1.5: Plot of $|\exp(-(\Phi(0) - \Phi(t)))|$ ($\Phi(t)$ defined in Eq(1.14)) for undamped acoustic phonons as a function of $t$ for $\alpha_p = 0.033$ and $\alpha_p = 0.066$ ($\alpha_p$ defined in Eq(1.15)) and for temperatures ranging from $T = 3.9K$ (upper curves) to $T = 78K$ (lower curves). The timescale is measured in units of $\tau \equiv L/c_l \simeq 1ps$. We see a rapid decay at early times which approaches a constant but reduced value at later times. The rapid decays at early times ($t \sim ps$) are a result of the rapid transfer of coherence from the quantum dot to the associated phonon reservoirs. The long time residual value $e^{-\Phi(0)}$ indicates incomplete dephasing of the quantum dot polarization due to undamped phonons in the Independent – Boson model.
In Fig(1.5) we plot $|e^{-(\Phi(0)-\Phi(t))}|$ as a function of time $t$ (in units of ps) for two values of dot-phonon coupling strength, $\alpha_p = 0.033$ and $\alpha_p = 0.066$. Pure dephasing due to acoustic phonons is clearly seen as a rapid loss of coherence at ps time scales. The coherence, however, does not decay to zero and approaches a constant value, implying that dephasing is only partial. The residual coherence decreases with temperature and dot-phonon coupling strength.

Having described pure dephasing in quantum dots using the Independent – Boson model, it is now important to find a mechanism to explain energy relaxation which is observed in quantum dots[92, 79]. The polaron states of the Independent – Boson model which were obtained using the polaron transformation are stationary states of the full electron-phonon system. It follows from standard quantum mechanical arguments that if the system is prepared in such a state it will neither evolve nor relax[93, 94]. If prepared in a linear superposition of the polaron states it would oscillate (in the sense of Rabi oscillations) between the multitude of polaron states but with no irreversible loss of energy. At low temperatures, the main mechanism for energy relaxation in a Independent – Boson model is the phonon instability due to anharmonic effects which result in its disassociation into other low energy phonons. This associates a finite lifetime to the phonons and is the primary mechanism for polaron decay. At higher temperatures phonon-assisted electronic transitions resulting in direct non-radiative decay also become important. These processes are not included in the Independent – Boson model which limits its applicability. In Chapter 4 we generalize the Independent – Boson model to include these processes phenomenologically by introducing a finite lifetime for the phonons themselves. We further allow for the possibility of direct non-radiative decay of the two-level system. This results in population and polarization decay due to phonons in addition to electromagnetic dephasing and relaxation.

The theory developed in this thesis focusses primarily on self-assembled InGaAs/GaAs quantum dots where phonon induced dephasing can be described by the Independent – Boson model[52] (especially at low temperatures). Experimental studies [95, 96, 97] in strongly confined quantum dots at low temperatures have clearly demonstrated that the polarization decay has a non-Lorentzian homogenous lineshape with a very narrow zero-phonon line superimposed on a broad acoustic-phonon spectrum. These InGaAs/GaAs quantum dot excitons have lifetimes as long as $T_1 = 900$ps in an unstructured electromagnetic reservoir of free space with dephasing times $T_2 = 630$ps at cryogenic temperatures [98, 99]. Since the dephasing times are almost as large as the decay times, dephasing in
these materials is radiatively limited. The exciton lifetime may be extended by placing these quantum dots inside the structured reservoir of the photonic crystal suppressing the radiative recombination of the electrons and holes due to the presence of a suitable photonic band gap. It is of interest to experimentally study the effects of suppression of radiative recombination which can result in much larger exciton lifetimes and longer decoherence times.

1.4 Optics in a structured electromagnetic reservoir

In this section we discuss some important aspects of optics in a structured electromagnetic reservoir which are important to understand the rest of the thesis. We also discuss some of the inherent difficulties in describing atom dynamics resulting from strong atom-photon interaction in photonic crystals (compared to free space) which help put in perspective the formalism that we have used throughout this thesis.

1.4.1 Photonic band structure

We now present a brief introduction to photonic band structures[100, 101]. They bear close resemblance to electronic band structures in solid-state physics. The electronic band structure in a solid is the energy spectrum of an electron and specifies the range of energies the electron is allowed. Diffraction of electronic De-Broglie waves in the periodic lattice structure of the crystal gives rise to the band structure of a given material and determines its physical characteristics (electronic and optical properties). Likewise, periodic variations in the dielectric function on length scales comparable to the wavelength of light result in scattering of light (optical Bragg diffraction). This can give rise to stopbands wherein propagation of light in certain specific directions is prohibited. In three-dimensional periodic dielectric material, strong scattering can lead to a full photonic band gap where no modes of the electromagnetic mode are allowed in a specific spectral region. In fact even the vacuum fluctuations of the electromagnetic field are absent in the bandgap leading to novel quantum electrodynamical effects. Restructured photonic density of states is of immense value in practical device applications of photonic crystals.

The propagation of electromagnetic waves in photonic crystals is described by Maxwell’s
equations with no free charge or current which are given by:

\[
\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \\
\n\nabla \cdot \vec{B} = 0 \\
\n\nabla \cdot \vec{D} = 0 \\
\n\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t}
\]

where \( \vec{E} \) and \( \vec{H} \) are the electric and magnetic fields and \( \vec{D} \) and \( \vec{B} \) are the displacement and magnetic-induction fields. The fields \( \vec{E} \) and \( \vec{H} \) are related to \( \vec{D} \) and \( \vec{B} \) respectively by the constitutive relations which are given by:

\[
\vec{D}(\vec{r}) = \epsilon(\vec{r})\vec{E}(\vec{r}) \\
\vec{B}(\vec{r}) = \mu_0 \vec{H}(\vec{r})
\]

where \( \epsilon(\vec{r}) \) is the dielectric function and \( \epsilon_0 \) and \( \mu_0 \) are the vacuum permittivities. The constitutive relations Eq(1.17) are written under the assumption that the field strengths are small enough that the linear relations Eq(1.17) are valid. For stronger field strengths higher order contributions need to be considered. We have also assumed that the material is isotropic so that \( \epsilon(\vec{r}) \) is a scalar. We also ignore any explicit dependence of \( \epsilon(\vec{r}) \) on frequency.

Several techniques have been developed to solve the above set of equations numerically on a computer to study the propagation of electromagnetic waves in a photonic crystal[102, 103]. It is beyond the scope of this chapter to discuss all of them in detail. In order to illustrate the salient features of a photonic band structure, we only discuss the plane-wave expansion method[102]. Numerical techniques based on the plane-wave expansion method are well developed owing to the stability and the reliability of the underlying algorithm and the minimal requirements of computer programming. The numerical implementation is based on Fourier transformation and matrix diagonalization. As drawbacks of this method we note that this method is not applicable to frequency dependent dielectrics. Moreover the computational cost becomes prohibitive for intricate photonic crystal structures since the use of a large number of plane waves renders matrix diagonalization very time consuming.

The periodicity of the photonic crystal (periodicity of the dielectric function) is char-
characterized in three-dimensional systems by the three primitive lattice vectors $\vec{a}_1$, $\vec{a}_2$ and $\vec{a}_3$. The dielectric function then obeys: $\epsilon(\vec{r}) = \epsilon(\vec{r} + n_1\vec{a}_1 + n_2\vec{a}_2 + n_3\vec{a}_3)$ for all integers $n_1$, $n_2$ and $n_3$. The linearity of the Maxwell equations allows us to expand the fields $\vec{E}(\vec{r})$ and $\vec{H}(\vec{r})$ into a series of harmonic modes. For light of a fixed frequency $\omega$, we rewrite the Maxwell’s equations as:

$$\vec{\nabla} \times [\vec{\nabla} \times \vec{E}(\vec{r})] = \epsilon(\vec{r})\frac{\omega^2}{c^2} \vec{E}(\vec{r})$$  \hspace{1cm} (1.18)$$

$$\vec{\nabla} \times \left[ \frac{1}{\epsilon(\vec{r})} \vec{\nabla} \times \vec{H}(\vec{r}) \right] = \frac{\omega^2}{c^2} \vec{H}(\vec{r})$$

Note that Eqs(1.18) have been cast in the form of an eigenvalue equation where the eigenfunctions provide the spatial distributions of the modes of the propagating field. It suffices to solve only one of these equations for the electric field or the magnetic field, the other field being uniquely determined through the Maxwell’s equations. For nonmagnetic materials, the operator $\vec{\nabla} \times \epsilon(\vec{r})^{-1}\vec{\nabla} \times$ acting on $\vec{H}(\vec{r})$ is Hermitean and its eigenvalues $\omega^2/c^2$ are real. In the following we focus on solving the eigenvalue equation for $\vec{H}(\vec{r})$.

The periodicity of the dielectric function implies that the allowed optical modes in the medium are the Bloch modes $\vec{u}_k(\vec{r})$ which satisfy the Bloch theorem[104]:

$$\vec{H}_k(\vec{r}) = e^{i\vec{k} \cdot \vec{r}} \vec{u}_k(\vec{r})$$ \hspace{1cm} (1.19)$$

The Bloch modes $\vec{u}_k(\vec{r})$ are characterized by the wavevector $\vec{k}$ and obey the periodicity condition of the photonic crystal: $\vec{u}_k(\vec{r}) = \vec{u}_k(\vec{r} + n_1\vec{a}_1 + n_2\vec{a}_2 + n_3\vec{a}_3))$. Using Eq(1.19) in the Maxwell’s equation for $\vec{H}_k(\vec{r})$ in Eq(1.18) we obtain:

$$\vec{\nabla} \times \frac{1}{\epsilon(\vec{r})} \vec{\nabla} \times e^{i\vec{k} \cdot \vec{r}} \vec{u}_k(\vec{r}) = \frac{\omega^2}{c^2} e^{i\vec{k} \cdot \vec{r}} \vec{u}_k(\vec{r})$$ \hspace{1cm} (1.20)$$

which can be rewritten as:

$$(i\vec{k} + \vec{\nabla}) \times \left[ \frac{1}{\epsilon(\vec{r})} (i\vec{k} + \vec{\nabla}) \times \vec{u}_k(\vec{r}) \right] = \frac{\omega^2}{c^2} \vec{u}_k(\vec{r})$$ \hspace{1cm} (1.21)$$

The transversality constraint now takes the form:

$$(i\vec{k} + \vec{\nabla}) \cdot \vec{u}_k(\vec{r}) = 0$$ \hspace{1cm} (1.22)$$
Figure 1.6: (a) Schematic of an inverted opal photonic crystal structure. (b) Total density of states of (a) for $\epsilon_{coating} = 11.9$. The radius of the core of the sphere is $|r| = \sqrt{2}/4|a|$ and the thickness of the coating shell is $0.09|a|$ where $|a|$ is the face centred cubic lattice constant. (c) Band structure of the inverse opal structure. The gaps opens between bands 8 and 9 and is centered at $|a|/\lambda_{vac} = 0.82$ (from PhD. thesis of Ovidiu Toader: *Photonic bandgap materials: architectures for microfabrication and their optical properties*).
We expand the dielectric function $\epsilon(\vec{r})$ and the field $\vec{H}(\vec{r})$ in a 3D-Fourier series over the reciprocal lattice vectors $\vec{G}$ as:

$$\epsilon(\vec{r})^{-1} = \eta(\vec{r}) = \sum_{\vec{G}} \eta_{\vec{G}} e^{i\vec{G} \cdot \vec{r}} \tag{1.23}$$

$$\vec{u}_{\vec{k}}(\vec{r}) = \sum_{\vec{G}} \vec{c}_{\vec{k}, \vec{G}} e^{i\vec{G} \cdot \vec{r}}$$

where the sum is over all reciprocal vectors $\vec{G}$ and $\vec{c}_{\vec{k}, \vec{G}} = \frac{1}{V} \int d^3\vec{r} e^{-i\vec{G} \cdot \vec{r}} \vec{u}_{\vec{k}}(\vec{r})$ with $V$ the unit-cell volume. Substituting Eq(1.23) into the eigenvalue equation for $\vec{H}(\vec{r})$ in Eq(1.18) we obtain a linear set of eigenvalue equations for all $\vec{G}$:

$$\sum_{\vec{G}'} \left[ -\eta_{\vec{G}' - \vec{G}} (\vec{k} + \vec{G}) \times [i(\vec{k} + \vec{G}')] \times \vec{c}_{\vec{k}, \vec{G}'} \right] = \frac{\omega_n^2}{c^2} \vec{c}_{\vec{k}, \vec{G}} \tag{1.24}$$

The eigenvalue problem is restricted to a single unit cell of the photonic crystal due to the periodic boundary condition which results in a discrete set of eigenvalues. For each wavevector $\vec{k}$ we find an infinite set of modes with discretely spaced frequencies which we label by a band index $n$. The frequency for each band $n$ varies continuously with $\vec{k}$. The set of continuous functions $\omega_n(\vec{k})$, indexed in order of increasing $n$, characterize the photonic band structure in the photonic crystal.

Eq(1.24) is an infinite set of linear equations for the infinite set of unknowns $\vec{c}_{\vec{k}, \vec{G}}$. The task now reduces to truncate this infinite set of equations by restricting the sum to a finite set of plane waves (e.g., a sphere around the origin) assuming that the terms corresponding to larger $|G|$ values are small. Once we have truncated the sum to a finite set, Eq(1.24) reduces to a finite matrix eigenequation which can be subsequently solved using fast Fourier transform algorithms. The truncation is usually a difficult task due to poor convergence of the Fourier transforms and numerical skill is required to simplify computations. The accuracy of the band structure computation is also determined by the number of plane waves used in matrix diagonalization following the truncation. Additional issues regarding the numerical aspects can be found in [102, 103].

For the rest of this thesis it is not necessary to consider the full photonic band structure of a periodic crystal since the underlying theme is atom-photon dynamics in the vicinity of the band edge. For theoretical purposes it is sufficient to consider an approximate photonic dispersion relation in the vicinity of the band edge. In this thesis we use the
Figure 1.7: (a) Schematic of an inverted diamond photonic crystal structure. (b) Total density of states of (a) for $\varepsilon_{\text{coating}} = 11.9$. The radius of the sphere is $|r| = 0.326|a|$ where $|a|$ is the face centred cubic lattice constant. (c) Photonic band structure of (a). The gaps opens between bands 2 and 3. The gap is centered at $|a|/\lambda_{\text{vac}} = 0.59$. (from PhD. thesis of Ovidiu Toader: *Photonic bandgap materials: architectures for microfabrication and their optical properties*)
isotropic dispersion relation outlined by John and Wang[9] which provides us with a simple analytic form for the photonic dispersion in the vicinity of the photonic band edge. In the following subsection we discuss some important features of atom-photon interaction near the photonic band edge and compare it an unstructured electromagnetic reservoir.

1.4.2 Atom-photon interaction in a photonic crystal

The interaction of atoms (two-level/few-level systems or their collections) with the radiation reservoir is a central theme in conventional quantum optics. The complete information about the dynamics of the atom is contained in its reduced density matrix \(\hat{\rho}(t)\) which is obtained by tracing the full density matrix (system + reservoir) over the radiation degrees of freedom. The reduced density matrix obeys a first-order dynamical equation, the master equation, and quantum expectation values of the atomic system are obtained from it. The structure of the master equation ensures the positivity of the reduced density matrix \(\hat{\rho}(t)\). Such an approach is well suited when the atom-photon interaction can be characterized as Markovian as in the infinitely broad featureless photonic reservoir of free space.

It is well established that atom-photon interaction depends on the behaviour of the density of states near the atomic transition frequency. If the density of states is a smoothly varying function of frequency (as in an unstructured reservoir of free-space), the temporal evolution of the dynamics of the atom at any instant of time \(t\) depends only on the present state of the atom (Markovian process) and all memory effects pertaining to its state at all previous times can be ignored. This regime is well described by the generalized Wigner-Weisskopf approach where the decay rate of the excited state of the atom is exponential and the spectrum is Lorentzian.

For the generic Hamiltonian Eq(1.8) describing the interaction of a two-level system with a bosonic (radiation) reservoir, the reduced density matrix \(\hat{\rho}\) (obtained by tracing out the reservoir degrees of freedom) can be shown to obey:

\[
\hat{\rho}(t) = -\int_0^t dt' Tr_R[\hat{H}_I(t'), [\hat{H}_I(t-t'), \hat{\rho}_R(0) \otimes \rho(t)]]
\]

(1.25)

where \(Tr_R\) denotes the trace over the reservoir and \(\hat{\rho}_R(0)\) is the reservoir density matrix at \(t = 0\). The interaction part of the Hamiltonian \(\hat{H}_I = \sum_k (\lambda_k \hat{\sigma}_e \hat{\alpha}_k + \lambda_k^* \hat{\alpha}_k^\dagger \hat{\sigma}_g)\) and \(\hat{H}_I(t)\) is
in the interaction representation defined using the non interacting part of the Hamiltonian given by $H_0 = \omega_0 \hat{\sigma}_{ee} + \sum_k \omega_k \hat{a}_k^\dagger \hat{a}_k$. Eq(1.25) is written in the Born approximation wherein the atom-photon interaction is considered perturbatively and only to second order in the atom-photon coupling constant. We assumed that the total density matrix (system + reservoir) at $t = 0$ is uncorrelated and written as $\hat{\rho}_R(0) \otimes \rho(0)$. We have also assumed that $tr_R(H_I(t)\hat{\rho}_R(0)) = 0$.

Eq(1.25) can be evaluated and simplified in the Markovian approximation wherein the reservoir correlation time is assumed to be very small compared to the time of observation. This requires the reservoir have non-denumerable infinite degrees of freedom and results in the system losing all memory of its past on typical timescales of observation. Eq(1.25) can then be recast into the form of a master equation in the Born-Markov approximation as:

$$\frac{\partial \hat{\rho}}{\partial t} = -i\omega_0 [\hat{\sigma}_{ee} - \hat{\sigma}_{gg}, \hat{\rho}] - \Gamma_0(\omega_0)[\hat{\sigma}_{eg}\hat{\sigma}_{ge}\hat{\rho} - 2\hat{\sigma}_{ge}\hat{\rho}\hat{\sigma}_{eg} + \hat{\rho}\hat{\sigma}_{eg}\hat{\sigma}_{ge}] \quad (1.26)$$

where $\Gamma_0(\omega_0)$ is the decay rate of the excited state due to spontaneous emission and is given by

$$\Gamma_0(\omega_0) = \frac{\omega_0^3|D_{ge}|^2}{3\pi \epsilon_0 \hbar c^3} \quad (1.27)$$

The master equation formalism, along with the quantum regression theorem, is sufficient to deal with standard computations in conventional quantum optics. The quantum regression theorem allows us to relate two-time correlation functions to the expectation value of a single-time operator which is straightforward to compute using the master equation Eq(1.26). For instance, consider the correlation function $P(t - t') = \langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(t') \rangle$. It is easy to show using Eq(1.26) that it is given by

$$|P(t - t')| = e^{-\frac{\Gamma_0(\omega_0)(t-t')}{2}} \quad (1.28)$$

The polarization decays exponentially and monotonically to zero with a rate which is half the decay rate of spontaneous emission. This exponential decay of the correlation function translates into a Lorentzian spectrum for the absorption lineshape which is characteristic of Markovian processes.

The situation is quite different, however, in the case of a structured electromagnetic reservoir of a photonic crystal. The photonic density of states in the structured electromagnetic reservoir of a photonic crystal reveals gaps for certain frequencies. The density of states also changes rapidly in the vicinity of the photonic band edge. In this case the
rate of change of the photonic density of states with frequency can be comparable to
the spontaneous emission rate and the Wigner-Weisskopf approach breaks down. This
invalidates the master equation used above in the case of smooth featureless density of
states of free space.

The absence of a simple master equation in the Born and Markov approximation as
in Eq(1.26) and the inapplicability of the quantum regression theorem in the structured
reservoir of a photonic crystal necessitates the use of other techniques[105, 106, 107, 108].
In this thesis we focus on the computation of the absorption lineshape and the two-
time correlation functions using the method of thermodynamic Green’s functions. We
also consider the generalization of the method of thermodynamic Green’s functions to
nonequilibrium problems, the method of generalized-Laplace transforms. However, these
methods are applicable to a restricted class of problems and lack the general applicability
of a master equation. They are limited to only single-photon processes and cannot be
readily generalized to compute multiple-time correlation functions (correlation functions
of atomic operators at more than two times).

The strongly non-Markovian behaviour leads to novel and nontrivial atom-photon
dynamics in a photonic crystal. The rapidly varying density of states in the vicinity of
a photonic band gap in the structured reservoir of a photonic crystal gives rise to effects
which are not readily observable in free space. If the resonant frequency of the atom
is in the vicinity of the band edge, atom-photon dynamics is strongly non-Markovian
with a non-exponential decay and a non-Lorentzian spectrum. Note that non-Markovian
atom-photon interaction is not restricted to photonic crystals and are also observed in
waveguides and microcavities. For instance, vacuum Rabi splitting of the excited state of
a two-level atom in a microcavity gives rise to a double peaked structure in the spectrum
as opposed to a single peaked Lorentzian spectrum. Similar effects are seen in a waveguide
if the atomic transition frequency is very close the cutoff frequency of the waveguide
(cutoff frequency of the waveguide is the frequency beyond which the waveguide no longer
contains any electromagnetic modes). Nonetheless photonic crystals offer a unique way
to control light which cannot be achieved in other systems. Some of these aspects will
be elaborated in detail in this thesis.

We now discuss a few important mathematical aspects of atom-photon interaction
in structured reservoirs. Consider a two-level system interacting with infinitely many
oscillators of the radiation reservoir given by the Hamiltonian Eq(1.8). The Fourier
transform of the correlation function \( P(t-t') = -i\Theta(t-t')\langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(t') \rangle \) is defined as
\[ P_\omega = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i\omega t} P(t) \]. It is easy to show that \( P_\omega \) (for instance, using the method of thermodynamic Green’s functions) is given by:

\[ P_\omega = \frac{1}{\omega - \omega_0 - \sum_k \frac{|\lambda_k|^2}{\omega - \omega_k}} \]  

(1.29)

Eq(1.29) is derived in the Born approximation and only includes single-photon processes. However, the Markov approximation has not been made. The term \( \sum_k \frac{|\lambda_k|^2}{\omega - \omega_k} \) is the self-energy contribution and is complex. The real part of the self-energy describes the renormalization of the resonant frequency of the two-level atom (Lamb shift) and the imaginary part is the linewidth associated with the decay of the excited state. To proceed further we need to specify a dispersion relation \( \omega_k \) for the radiation modes in the structured reservoir of a photonic crystal.

We consider a simple qualitative model proposed first by John and Wang[9]. Consider an infinite photonic crystal with a spherical Brilloiun zone and assume that a propagating mode is subject to the same periodic potential irrespective of the polarization or the direction of propagation. The photon dispersion relation for the isotropic model can then be shown to satisfy the following transcendental equation:

\[ 4n \cos(kL) = (1 + n)^2 \cos[(2na + b)\frac{\omega_k}{c}] - (1 - n)^2 \cos[(2na - b)\frac{\omega_k}{c}] \]  

(1.30)

which is an exact solution of the one-dimensional scalar wave equation:

\[ -\nabla^2 \phi(x) - \frac{\omega^2}{c^2} \epsilon(x) \phi(x) = \frac{\omega^2}{c^2} \phi(x) \]  

(1.31)

with the dielectric constant \( \epsilon(x) \) given by:

\[ \epsilon(x) = \sum_{m=-\infty}^{\infty} u(x - mL) \]  

(1.32)

and \( u(x) = n^2 - 1 \) for \(|x| < a\) and 0 otherwise. Here \( n \) is the refractive index of the dielectric scatterers and \( a \) is its radius. \( L \) is the lattice constant such that \( 2a + b = L \). The normal modes of the electromagnetic field are assumed to be plane waves and written as \( \hat{e}_{\nu k} e^{ik \cdot r}/\sqrt{V} \) where \( V \) is the volume of the dielectric sample and the polarization vectors satisfy the transversality condition \( \sum_{\nu = 1}^2 \hat{e}_{\nu k}^i \hat{e}_{\nu k}^j = (\delta_{ij} - \hat{k}_i \hat{k}_j) \). For the special case
Figure 1.8: Dispersion relation and density of states for the isotropic model. If the resonant frequency of the atom is near the upper band edge, the lower band edge can be ignored. In an effective mass approximation the dispersion relation can be approximated near the upper band edge by \( \omega_k = \omega_e + A(k - k_0)^2 \).

In an effective mass approximation to the dispersion relation Eq(1.33) near the upper band edge, we obtain:

\[
\omega_k = \omega_e + \omega_0^2 A(k - k_0)^2
\]

where

\[
\omega_e = \frac{c}{4na} \left[ 2n - \cos^{-1} \left( \frac{1 + n^2 - 6n}{1 + n^2 + 2n} \right) \right]
\]

and \( A = -2ac/\sin(4na\omega_e/c) \). \( k_0 \) is the wave vector corresponding to the band edge frequency and is given by \( k_0 = \frac{\pi}{2a(n+1)} \). The density of states corresponding to the dispersion relation Eq(1.34) is easily evaluated to:

\[
\rho(\omega) = \frac{V}{(2\pi)^3} \frac{k_0^2}{\sqrt{A}} \frac{\Theta(\omega - \omega_e)}{\sqrt{\omega - \omega_e}}
\]

where \( \Theta(\omega - \omega_e) \) is the Heavyside step function. Such a density of state \( \rho(\omega) \) can be realized in 1-d waveguide within a photonic band gap where the waveguide exhibits a 1-d gap itself. We see from Eq(1.36) that the density of states is zero for \( \omega < \omega_e \) and no electromagnetic modes can propagate for these frequencies. The correlation function
$P(\omega)$ in Eq(1.29) can now be evaluated to obtain:

$$P_\omega = \frac{1}{\omega - \omega_0 + i\frac{C}{\sqrt{\omega - \omega_0}}}$$  \hspace{1cm} (1.37)

with $C = \frac{\omega_0^2 |D_{ph}|^2 k_3^3}{12\pi \epsilon_0 \epsilon_\infty \omega_0^3}$. Introducing detuning $\delta = \omega_0 - \omega_e$ and a shift in frequency, we rewrite Eq(1.37) as:

$$P_{\omega+\omega_0} = \frac{1}{\omega + i\frac{C}{\sqrt{\omega + \delta}}}$$  \hspace{1cm} (1.38)

which can be rearranged as:

$$P_{\omega+\omega_0} = \frac{\omega(\omega + \delta) - iC\sqrt{\omega + \delta}}{\omega^2(\omega + \delta) + C^2}$$  \hspace{1cm} (1.39)

The dipole correlation function is determined by the nature of the three poles of $P_\omega$. We note two complex poles which are conjugates of each other and one real pole. The pole with positive imaginary part is unphysical whereas the pole with negative imaginary part is the transient solution and gives the decay part of the solution. The real pole is the nondecaying solution and is due to the formation of a photon-atom bound state.

![Figure 1.9: Correlation function $P(t)$ of a two-level in the structured reservoir of a photonic crystal for various values of detuning $\delta$ of the transition frequency from the band edge. $a = C^\frac{1}{2}$ where $C = \frac{\omega_0^2 |D_{ph}|^2 k_3^3}{12\pi \epsilon_0 \epsilon_\infty \omega_0^3}$. Non-zero correlation at long times is seen for detunings near the band edge. The correlation function decays exponentially, as in free space, if the detuning of the atomic transition frequency is far outside the gap.](image)

The two physical poles which contribute to the inversion integral can be understood
as follows. The resonant coupling of the atom to photons with vanishing group velocity results in self-dressing of the atom by its own localized radiation field which splits the atomic level into a doublet. The lower frequency component of the doublet is pushed inside the gap and forms a photon-atom bound state. The photon emitted by this component tunnels through the crystal for a finite length and is Bragg-reflected back to the emitting atom and is subsequently reabsorbed. The characteristic length scale for such a photon scattering is called the "localization length" which increases as the atomic transition frequency is shifted near the band edge. The second component of the doublet is pushed outside the band gap and into the continuum and decays by emitting into the allowed modes. This splitting of the excited state is analogous to the vacuum-Rabi splitting of atoms placed in microcavities which, however, are very weak. Moreover, in contrast to the Mollow splitting of the excited state in the presence of a strong external laser field, the splitting of the atomic level in the vicinity of the band edge is only due to the interaction of the atom with its own localized radiation field. Interference between the radiation fields emitted by the doublets leads to the oscillatory behaviour seen in Fig(1.9). Unlike in free space, the correlation function at long time is non-zero for $\delta < 0$ and has a nonexponential decay due to beating between the localized state and the decaying solution.

1.5 Outline of the thesis

We now present a short outline of this thesis. The central theme of this thesis is the study of the role of nonradiative degrees of freedom (phonons) on the optical lineshape of quantum dots placed inside the structured reservoir of a photonic crystal. Our focus is primarily on self-assembled quantum dots. These quantum dots have been studied in detail and experimental observations (in free space) of photoluminescence spectra of single quantum dots exhibit a narrow zero-phonon line on top of a broad background[91, 109, 110, 111] (due to acoustic phonons[112]).

In chapter II we first present a detailed discussion of two formulations of optical lineshape which can be applied to self-assembled quantum dots placed in an unstructured electromagnetic reservoir. However, these formalisms are of limited validity in the structured reservoir of a photonic crystal. We first discuss the stochastic approach to optical lineshape which was first proposed by Anderson[113, 114] and expanded by Kubo[115, 116]. The stochastic approach describes dephasing due to random fluctua-
tions of the excited state of the quantum dot. These random fluctuations results from
the interaction of the quantum dot with the lattice modes and leads to pure dephasing
with no population relaxation. We then present a semiclassical approach to the optical
lineshape based on the works of Kubo and Toyozawa[50] and Huang and Rhys[117]
specific to optical phonons). The semiclassical approach provides a correct description
of the optical lineshape due to phonons but fails to account for the effect of phonons
on the radiative component of the lineshape function. We also discuss a phenomenologi-
cal approach to describe polaron relaxation in this formulation following the works of
Mukamel[118, 119]. We then introduce a model Hamiltonian that provides a full quantum
mechanical description of photon and phonon processes which lead to dephasing
and decoherence of the quantum dot (modelled as a two-level system). This provides a
more fundamental picture than previous studies[120, 121, 122, 123, 124, 125] where in
the Born-Oppenheimer approximation[73, 74], the nuclear or lattice degrees of freedom
are assumed to be frozen. In the Condon approximation[126] the optical dipole transition
matrix element is also assumed to be independent of lattice coordinates. Our approach
treats the electronic, photonic and lattice degrees of freedom at a more fundamental
quantum level. We proceed by deriving an effective Hamiltonian using a mean-field the-
ory to decouple the radiative and lattice degrees of freedom. In this approximation the
effective dipole-moment incorporates the mean-field response of the lattice and is temper-
ature dependent. This allows us to develop a mean-field theory to describe the absorption
lineshape due to phonon sidebands which is valid for a general photonic reservoir (in the
Born approximation).

In chapter III we present a general formalism to calculate the absorption lineshape
incorporating the phonon sidebands in a general photonic reservoir. The absorption
lineshape is calculated using the method of thermodynamic Green’s functions (of the
two-level atom dipole operators). The Green’s functions are computed using the effective
mean-field Hamiltonian obtained in chapter II. A hierarchy of equations is generated
for the thermodynamic Green’s functions which is truncated to second order in the two-
level atom-photon coupling constant (Born approximation) without making the Markov
approximation. The Born approximation corresponds to including only single-photon
processes which is valid given the weak coupling between the atom and the photon reser-
voir. Next we calculate the polarization (dipole autocorrelation function) again utilizing
the mean-field approximation described in Chapter II. We then focus our attention to
the study of the population dynamics (lifetime) of the two-level system in the presence of
phonons. Population dynamics is a nonequilibrium problem wherein the density matrix is not of the canonical form. We use the Heisenberg equations of motion to describe the population dynamics in the Born approximation without making the Markov approximation. We also discuss a generalization (in Appendix B) of the method of thermodynamic Green’s functions to nonequilibrium problems, the method of generalized-Laplace transforms, and derive results which are identical to those obtained using the Heisenberg equations of motion. This serves as a useful introduction to this technique which we feel has not been explored sufficiently in the literature.

In chapter IV we consider specific models for electromagnetic reservoirs, the unstructured reservoir of free space and the structured reservoir of a photonic crystal, and obtain explicit expressions for the absorption spectra and the polarization and population dynamics using the general results obtained in the previous chapter. We first calculate the linear optical susceptibility in the presence of a finite temperature bath of optical phonons in a smooth, featureless photonic density of states. As a diagnostic of our model and its quantum mechanical solution, we recapture the standard picture of multiple-phonon emission and absorption sidebands[50]. Spectral signatures such as phonon sidebands emerge naturally in our formalism. We then apply our theory to a quantum dot placed inside a photonic crystal. We compute the absorption lineshape and demonstrate that phonon-mediated sidebands can lead to absorption of photons inside the photonic band gap. We then study the polarization dynamics of the two-level atom and show that, for undamped optical phonons, the atom can have residual coherence even at large times. This residual coherence is a result of the photon-atom bound state. Finally we study the population dynamics of the excited two-level atom in the presence of undamped optical phonons.

In chapter V we generalize the discussion in the previous chapters to undamped acoustic phonons and include additional processes which are not contained in the Independent—Boson model. We first study the polarization and population dynamics of the quantum dot coupled to a continuum of undamped acoustic phonons and placed inside the structured reservoir of a photonic crystal. The distinct and well-separated phonon sidebands merge into a continuum but with a δ-function peak at the quantum dot resonant frequency. This leads to pure dephasing since acoustic phonons do not cause polaron relaxation. We then generalize our model Hamiltonian to allow for nonradiative decay processes such as phonon-assisted electronic transitions from the excited state to the ground state which can result in population relaxation. This additional relaxation chan-
nel provides a more realistic picture of the spectral linewidths of the phonon sidebands and a broadening of the zero-phonon line. We then discuss a phenomenological generalization of the Independent – Boson model to include phonon-phonon scattering and other anharmonic processes[127]. These processes lead to damping of the phonons which allows the polaronic cloud to decay resulting in energy dissipation within the vibrational degrees of freedom. Using these models we study the modified polarization and population dynamics of the quantum dot. Finally we consider the case of damped optical phonons and obtain explicit expressions for nonradiative broadening of the phonon sidebands due to finite optical phonon lifetime.

Chapter VI presents a summary of the thesis and suggests possible applications of the theory developed here. In the appendices we provide additional details not presented in the main body of the thesis.
Chapter 2

General theory of optical susceptibility and lineshape

The main aim of this thesis is the study of the optical properties of a single quantum dot embedded in the solid backbone of a photonic crystal. Due to manufacturing techniques the host dielectric matrix of the photonic crystal will contain many quantum dots. However, at sufficiently low densities the quantum dots may be considered to be noninteracting. The spectra of a collection of almost identical quantum dots (non-identical due to fluctuations in growth conditions and different local conditions of the background dielectric) is invariably inhomogeneously broadened as each quantum dot contributes a zero-phonon line and a phonon sideband to the absorption and emission spectra. The individual zero-phonon line and phonon sidebands are slightly shifted in frequencies and have dissimilar spectral weights due to different transition energies and coupling strengths of the relevant excitonic transition. Therefore single quantum dot spectroscopy is paramount to study and differentiate the optical spectra of a single quantum dot from the inhomogenously broadened spectra. In the following two sections we present two formalisms used frequently in the literature to describe the optical lineshape of a single quantum dot in the unstructured electromagnetic reservoir of a homogenous medium. We point out the drawbacks of these models. We then introduce a full quantum mechanical Hamiltonian which takes into account all processes relevant to understanding the optical lineshape of a quantum dot in a general electromagnetic reservoir.
2.1 Stochastic formulation of the optical lineshape - Kubo and Anderson approach

We now present an analytical framework to study optical line shape of a single quantum dot in the unstructured reservoir of free space using a stochastic approach. The results discussed here are relevant to measurements of optical spectra of quantum dots by single-dot spectroscopy (eliminating the inhomogenous background). The formulation assumes that the quantum dot can be represented as a two-level system and the interaction of the quantum dot with lattice degrees of freedom is described as a modulation of the two-level system transition frequency. The modulation is mathematically modelled as a sudden and random jump (stochastic fluctuations) of the excited state frequency. The first theoretical model based on sudden and random frequency modulation was described by Anderson and Weiss in the context of electron paramagnetic resonance[113]. The fluctuating transition frequency was modelled stochastically as a Gaussian random process. Within this simplified stochastic model Anderson and Weiss demonstrated that the absorption lineshape was a Gaussian in the slow modulation limit whereas it was a "motionally narrowed" Lorentzian in the fast modulation limit. Another important model was introduced by Anderson where it was assumed that the transition frequency jumped randomly among a set of predetermined discrete values[114]. These two models were subsequently extended by Kubo and others[115, 116, 128] and applied to various systems including spectroscopy in the presence of vibrational dephasing.

The stochastic formulation based on the works of Anderson and Kubo can be adapted to study the optical lineshape of a single quantum dot as illustrated below. The electronic energy levels of a quantum dot embedded in the solid host are very sensitive to the local environment[129]. As a simple model to describe the role of the local solid-state environment we consider the transition of the quantum dot from the excited state with one exciton to the ground state with no excitons. The transition frequency $\omega(t)$, which is now time-dependent, can be considered as a sum of a static component $\omega_0$ and a dynamic component $\delta \omega(t)$. The static component determines the frequency origin for the spectral line whereas the fluctuating and dynamic component determines the line shape. The fluctuating component $\delta \omega(t)$ can originate from coupling to acoustic and optical phonon modes and are governed by the shape and size of the quantum dot. The electric dipole moment of the quantum dot also couples to the radiation reservoir
resulting in decay which we treat as a Markov-process in this section. We also make an important assumption that the modulation of the transition frequency of the excited state of the quantum dot is very rapid compared to the timescales involving radiative processes allowing us to ignore any interference between radiative transitions and non-radiative processes.

In the linear response regime, the absorption lineshape of the quantum dot can be written as \((\hbar = 1)\):\[\frac{\Gamma_0(\omega_0)}{2} e^{-\frac{\Gamma_0(\omega_0)}{2} t} \langle e^{-i \int_0^t d\tau \delta \omega(\tau)} \rangle_{sm} \] (2.1)

where \(\Gamma_0(\omega_0)\) is the lifetime of the two-level excited state solely due to radiative processes (Markovian interaction between the two-level system and the radiation reservoir is assumed). \(\langle \ldots \rangle_{sm}\) denotes the average over all stochastic realizations of the frequency trajectory \(\omega(\tau)\) within the interval \((0, t)\). The stochastic average introduces an additional polarization relaxation channel due to frequency fluctuations.

The simplest way to evaluate the trace over the stochastic ensemble is by performing a cumulant expansion\([115]\). We assume a Gaussian process for the stochastic fluctuations. Consequently all cumulants beyond second order vanish identically. The Gaussian correlation for the stochastic modulations of the excited state energy level can be justified using the central limit theorem. Many relatively weak random fluctuations contribute to the stochastic modulation and their sum assumes a normal distribution (central limit theorem). Note that the factorization of higher order correlation functions for a Gaussian random variable is formally identical with the factorization of higher order time-ordered correlation functions for harmonic degrees of freedom in a quantum mechanical description. As discussed in the next section, non-radiative (lattice) degrees of freedom are typically modelled as a bath of simple bosonic harmonic oscillators.

Performing the cumulant expansion, the stochastic average of the exponentials of the Gaussian random processes \(\langle e^{-i \int_0^t d\tau \delta \omega(\tau)} \rangle_{sm}\) can be expressed as:

\[\langle e^{-i \int_0^t d\tau \delta \omega(\tau)} \rangle_{sm} = e^{-\frac{1}{2} \int_0^t dt_1 \int_0^t dt_2 \langle \delta \omega(t_1) \delta \omega(t_2) \rangle} \] (2.2)

We assume that the correlation function for the stochastic modulation is exponential and given by:

\[\langle \delta \omega(t_1) \delta \omega(t_2) \rangle = \Gamma_{sm}^2 \exp(-\Omega |t_1 - t_2|) \] (2.3)
where $\Gamma_{sm}$ is the root mean square of the stochastic modulations of the excited frequency and $\Omega^{-1}$ is the correlation time of the modulations of the transition frequencies about the average $\omega_0$.

The trace over the stochastic ensemble can now be evaluated for the correlation function Eq(2.3). Using Eq(2.3) in Eq(2.2) and performing the integrals we obtain:

$$\langle e^{-i \int_0^t d\tau \delta \omega(\tau) \rangle}_{sm} = e^{-\Phi(t)}$$

(2.4)

where $\Phi(t)$ is given by:

$$\Phi(t) = \frac{\Gamma_{sm}^2}{\Omega^2} \left[ e^{-\Omega t} + \Omega t - 1 \right]$$

(2.5)

The full lineshape function in the presence of radiative decay is obtained by using Eq(2.5) in Eq(2.1):

$$\chi(t) \propto \int_0^\infty dt e^{i\omega t} e^{i\omega_0 t} e^{-\frac{\Gamma_{sm}}{2} t} e^{-\Phi(t)}$$

(2.6)

It is instructive to consider two limiting cases. For very short times $t \ll \Omega^{-1}$, the average over the stochastic ensemble reduces to:

$$\langle e^{-i \int_0^t d\tau \delta \omega(\tau) \rangle}_{sm} = e^{-\frac{1}{2} \Gamma_{sm}^2 t^2}$$

(2.7)

Hence the decay of the dipole autocorrelation function due to stochastic modulations at short times is Gaussian. For very long times, $t \gg \Omega^{-1}$, the stochastic average decays as:

$$\langle e^{-i \int_0^t d\tau \delta \omega(\tau) \rangle}_{sm} = e^{-\frac{\Gamma_{sm}^2}{\Omega^2} t}$$

(2.8)

The dephasing of the dipole autocorrelation function is now exponential and the optical lineshape function is a Lorentzian which is reminiscent of Markovian processes.

From the above presentation it is clear that stochastic modulations of the excited state energy level can capture pure dephasing exactly along the lines of the spin-boson model. The population of the excited state does not decay due to stochastic modulations and this approach should be relevant to compute optical lineshape of a quantum dot from a phenomenological perspective. The primary drawback of the stochastic approach is the lack of knowledge of the dependence of the dephasing rate on the microscopic parameters of the quantum dot. In the following section we present an alternate approach to the optical lineshape problem based on the work of Kubo[115, 116] and Toyozawa[50] and
Chapter 2. General theory of optical susceptibility

Huang and Rhys[117]. Later on in this chapter we write down a full model Hamiltonian which presents a complete quantum mechanical description of radiative and non-radiative processes and use it to compute the optical lineshape of a two-level atom.

2.2 Semiclassical formulation of the optical lineshape

- Kubo and Toyozawa approach

We now discuss the semiclassical formulation of the line shape (zero-phonon line and the phonon sidebands) of an optically active (absorbing and emitting) material embedded in a solid state matrix. A quantum dot embedded in the dielectric backbone of a photonic crystal is an example of such an optically active material. The coupling of the electron to the phonons produces additional structure in the optical lineshape (phonon sidebands) at frequencies which are integer multiples of phonon frequencies. In a semiclassical description, the electromagnetic field drives the electronic degree of freedom which in turn can drive the lattice modes due to the electron-phonon coupling. This gives rise to the phonon sidebands.

The idea of sidebands is best illustrated by the following example. Consider a classical Hamiltonian system describing a localized degree of freedom such as an exciton in a quantum dot which is nonlinearly coupled to the lattice modes (modelled as harmonic oscillators). The model Hamiltonian can be written as:

\[ H = \frac{1}{2} \left( \frac{d^2 x}{dt^2} \right)^2 + \frac{1}{2} \left( \frac{d^2 y}{dt^2} \right)^2 + \frac{1}{2} \omega_1^2 x^2 + \frac{1}{2} \omega_2^2 y^2 + \frac{1}{2} \gamma \omega_1^2 x^2 y \]  

(2.9)

Here \( x \) (\( y \)) denotes the electron (phonon coordinate) and \( \gamma \) is the nonlinear coupling between the two oscillator modes. The Hamiltonian equations of motion are straightforward to write:

\[ \frac{d^2 x}{dt^2} + \omega_1^2 x + \gamma \omega_1^2 xy = 0 \]  

(2.10a)

\[ \frac{d^2 y}{dt^2} + \omega_2^2 y + \frac{1}{2} \gamma \omega_1^2 x^2 = 0 \]  

(2.10b)

Substituting the zeroth-order solution \( x = x_0 \exp(\pm i \omega_1 t) \) and \( y = y_0 \exp(\pm i \omega_2 t) \) in the equations of motion Eq(2.10) we obtain, by successive approximation, a series of allowed frequencies \( |\omega_1 + n \omega_2| \) for the \( x \) oscillator. Imagine an external oscillating electromagnetic field \( Fe^{-i \omega t} \) driving the \( x \)-oscillator. Resonant absorption takes place not only at \( \omega_1 \)
but also at $|\omega_1 + n\omega_2|$. The external force excites a quantum $\hbar \omega_1$ of the $x$ oscillator which subsequently excites the vibrational degrees of freedom. Such structure in the absorption spectra resulting from the simultaneous excitation of another oscillator not directly subject to the external electromagnetic field is referred to as a sideband.

We now present a semiclassical theory of the optical lineshape including the sideband structure of an optically active material such as a quantum dot. As has been discussed previously, a quantum dot can be mathematically represented by a two-level system consisting of a ground state and an excited state. The two-level system is coupled to the electromagnetic field and also to the local environment of the host dielectric lattice. The optical lineshape is computed under the following assumptions. We treat the absorption and emission of photons by the quantum dot (optically active material) perturbatively (to second order in the dot-photon coupling constant) as the quantum dot light coupling is several orders of magnitude smaller than (i) quantum dot lattice coupling and (ii) quantum dot transition energy. The optical transition between the ground and the excited state is described using the "Condon" approximation\cite{50} where the electric dipole elements are assumed to be independent of phonon degrees of freedom. In a quantum mechanical description of the lattice dynamics, the initial lattice configuration is determined by quantum statistical mechanics and the lattice vibrational mode is a quantum harmonic oscillator. This corresponds to a parabolic adiabatic potential for the electron and equidistant phonon energy levels. In a purely classical theory the initial configuration is treated using classical statistical mechanics.

In the following we present a semiclassical description of the optical lineshape of a two-level atom adapted from reference\cite{50}. Let $H_L = K_L(\hat{p}) + U_L(\hat{q})$ be the Hamiltonian for the lattice vibrations of the medium in which the quantum dot is embedded and $H_e(\tilde{p}, \tilde{r}, \tilde{q})$ be the Hamiltonian for the localized electron in the excited state of the quantum dot in the presence of phonons. Here $\tilde{r}$ and $\tilde{p}$ denote the position and momentum coordinates of the electron while $\tilde{q} \equiv (q_1, q_2, \ldots)$ and $\tilde{p} \equiv (p_1, p_2, \ldots)$ denote those of the phonon modes. The excited electronic Hamiltonian $H_e$ depends upon the phonon coordinates $\tilde{q}$ since the "local" potential felt by the electron depends on the lattice displacement in its vicinity. The total Hamiltonian for the electron-phonon system in the excited state is then given by $H_{e}^{\text{tot}} = H_e + H_L$.

We now assume that the excited electronic state is separated from other higher electronic states by energies which are much larger than typical phonon energies such that no mixing of the energy levels is induced. We restrict our attention to the two low-
est energy levels of the electronic spectrum (two-level system). Hence in the adiabatic approximation\[50\], the Schrödinger equation for the electron and lattice vibrations can be solved as:

\[
\begin{align*}
H_e(\vec{p}, \vec{r}; \vec{q}) \Phi_e(\vec{r}; \vec{q}) & = \omega_e(\vec{q}) \Phi_e(\vec{r}; \vec{q}) \\
[K_L(\vec{p}) + W(\vec{q})] \chi_n(\vec{q}) & = E_n \chi_n(\vec{q})
\end{align*}
\] (2.11)

where \( W(\vec{q}) \equiv \omega_e(\vec{q}) + U_L(\vec{q}) \) is the adiabatic potential felt by the phonons and \( n \) denotes the vibrational state (\( \Phi_e(\vec{r}; \vec{q}) \) is the wavefunction of the excited electronic state and \( \chi_n(\vec{q}) \) is the \( n^{th} \) vibrational state). The wavefunction of the combined electron-phonon system is then given by the product of the wavefunctions of the electron and the phonons as \( \Phi_e(\vec{r}; \vec{q}) \chi_n(\vec{q}) \). The energy of the combined electron-phonon system in the excited state is \( E_n \).

In thermal equilibrium the phonon states are Boltzmann distributed with probabilities \( w_n \propto \exp(-\beta E_n) \). We also assume that the thermal energy \( K_B T \equiv \beta^{-1} \) is much smaller than the energy separation between the ground and the excited electronic state. The optical absorption lineshape now corresponds to the electronic transition from the ground state to the excited state.

The optical lineshape function \( f(t) \) is the autocorrelation function of the atomic dipole operator. It is related to the optical absorption spectra \( F(\omega) \) corresponding to the electronic transition from the ground state to the excited state through \( f(t) \equiv \int_{-\infty}^{\infty} d\omega F(\omega) \exp(-i\omega t) \). Let \( \lambda(\vec{q}) \) denote the electric dipole moment of the quantum dot which is a function of the phonon coordinates through its dependence on \( \vec{q} \). The density matrix of the phonon system is given by \( \rho(\beta) = \exp(-\beta H) \) (\( H \equiv K_L(\vec{p}) + W(\vec{q}) \) defined in Eq(2.11)). The autocorrelation function is given by\[119\]:

\[
f(t) = tr_L[\rho(\beta) \exp(iH_L t) \lambda(\vec{q}) \exp(-iHt) \lambda(\vec{q})] / tr_L[\rho(\beta)]
\] (2.12)

where \( tr_L \) denotes the trace operation over the lattice system (in units of \( \hbar = 1 \)). We now proceed with the computation of the generating function \( f(t) \) under the following approximations. We first make the harmonic approximation for the lattice vibrations (parabolic adiabatic potential):

\[
H_L = \sum_j \frac{1}{2}(\vec{p}_j^2 + \Omega_j^2 \vec{q}_j^2) = \sum_j (\hat{c}_j^\dagger \hat{c}_j + \frac{1}{2}) \hbar \Omega_j
\] (2.13)
where $\tilde{q}_j$ and $\tilde{p}_j (j = 1, 2, \ldots)$ represent the normal coordinates and the conjugate momentum of the phonon degrees of freedom and $\hat{c}_j$ ($\hat{c}_j^\dagger$) are the annihilation (creation) operators of the phonon modes (following second quantization) defined as:

$$\begin{align*}
\hat{c}_j &= (2\hbar \Omega_j)^{-1/2} (\Omega_j \tilde{q}_j + i \tilde{p}_j) \\
\hat{c}_j^\dagger &= (2\hbar \Omega_j)^{-1/2} (\Omega_j \tilde{q}_j - i \tilde{p}_j)
\end{align*}$$

We now expand $H_e(\tilde{p}, \tilde{r}; \tilde{q})$ in a Taylor series around $\tilde{q}_j = 0$ and keep only the linear term:

$$H_e(\tilde{p}, \tilde{r}; \tilde{q}) = H_e^0(\tilde{p}, \tilde{r}) + H'(\tilde{r}, \tilde{q})$$

Here $H'$ is the linear electron-phonon interaction and the coefficients are related by $\eta_j(\tilde{r}) = -\left(\frac{\hbar}{2m_j}\right)^{1/2} (\partial H_e / \partial \tilde{q}_j)|_{\tilde{q}=0}$. The full Hamiltonian in Eq(2.15) consisting of $H_e^0(\tilde{p}, \tilde{r})$ and $H'(\tilde{r}, \tilde{q})$ can be written in the second quantized form as:

$$\begin{align*}
H_e^0 &= \omega_e \hat{\sigma}_{ee} \\
H' &= \sum_j \eta_j \hat{\sigma}_{ee}(\hat{c}_j + \hat{c}_j^\dagger)
\end{align*}$$

where $\eta_j$ is the dot-phonon coupling constant. Note that the interaction Hamiltonian in Eq(2.16) is of the $x^2y$ form as in the Hamiltonian Eq(2.9). The sidebands in this second-quantized representation of our system are excited due to the coupling of the harmonic oscillators $\hat{c}_j$ to the two-level system through the coupling constant $\eta_j$.

We now calculate the eigenenergies $\omega_e(\tilde{q})$ of Eq(2.11) to first-order perturbation in $H'$. Adding $U_L(\tilde{q})$, the adiabatic potential is now found to be:

$$W(\tilde{q}) = \omega_e + \sum_j \left(\frac{1}{2} \Omega_j^2 \tilde{q}_j^2 - b_j \tilde{q}_j \right)$$

where $b_j = -\frac{\partial H_e}{\partial \tilde{q}_j}|_{\tilde{q}_j=0}$. Here we have assumed that $H'$ is small enough such that all higher order perturbation contributions starting with second order can be neglected. The adiabatic potential $W(\tilde{q})$ is now given by:

$$W(\tilde{q}) = E_0 + \sum_j \frac{1}{2} \Omega_j^2 (\tilde{q}_j - \Delta_j)^2$$
where

\[\Delta_j \equiv \frac{b_j}{\Omega_j^2}\]  \hspace{1cm} (2.19)

\[E_0 = \omega_c - \sum_j \frac{1}{2} \Omega_j^2 \Delta_j^2\]

\(\Delta_j\) represents the equilibrium displacement of the lattice when the electron is in the excited state. It corresponds to the configuration where the adiabatic potential \(W(q)\) takes the minimum value \(E_0\). \(\Delta_j\) is known as the polaron shift and is a renormalization in the energy of the excited state due to coupling to phonons.

We now evaluate the dipole autocorrelation function. In the Condon approximation, we neglect the \(\tilde{q}\) dependence of the dipole moment and take the constant (independent of phonon coordinates) \(|\lambda|^2\) out of the operation \(tr_L\) in Eq(2.12). The evaluation of the generating function now reduces to:

\[f(t) = |\lambda|^2 \langle e^{iH_L t} e^{-iH_t} \rangle_L\]  \hspace{1cm} (2.20)

where \(\langle ... \rangle_L\) denotes the trace over the vibrational states. The second quantized expressions for the phonon Hamiltonians \(H_L\) and \(H\) are given by \((\hbar = 1)\):

\[H_L = \sum_j \Omega_j \hat{c}^\dagger_j \hat{c}_j\]  \hspace{1cm} (2.21a)

\[H = E_0 + \sum_j \Omega_j (\hat{c}^\dagger_j - \frac{\eta_j}{\Omega_j})(\hat{c}_j - \frac{\eta_j}{\Omega_j})\]  \hspace{1cm} (2.21b)

We now introduce the unitary operator \(U = e^{S}\) where:

\[S \equiv \sum_j \frac{\eta_j}{\Omega_j}(\hat{c}^\dagger_j - \hat{c}_j)\]  \hspace{1cm} (2.22)

We note that the unitary transformation acts as follows: \(U^{-1}\hat{c}_j U = \hat{c}_j + \eta_j/\Omega_j\). In the Heisenberg representation \(U(t) \equiv \exp(iH_L t) U \exp(-iH_L t)\). Also \(U^{-1}HU = \tilde{\omega}_0 + H_L\) where \(\tilde{\omega}_0 = E_0 - \omega_g\). The lineshape generating function \(f(t)\) now reduces to:

\[f(t) = |\lambda|^2 \exp(-i\tilde{\omega}_0 t) \langle U(t) U^{-1}(0) \rangle_L\]  \hspace{1cm} (2.23)
\[ f(t) = |\lambda|^2 \exp(-i\omega_0 t - \Phi(0) + \Phi_+(t) + \Phi_-(t)) \]  
(2.24)

where

\[ \Phi_+(t) \equiv \int_0^\infty d\Omega' s(\Omega')(N(\Omega') + 1) \exp(-i\Omega't) \]  
(2.25)
\[ \Phi_-(t) \equiv \int_0^\infty d\Omega' s(\Omega') N(\Omega') \exp(i\Omega't) \]
\[ \Phi(0) \equiv \Phi_+(0) + \Phi_-(0) \]
\[ s(\Omega') \equiv \sum_j \frac{\eta_j^2}{\Omega_j^2} \delta(\Omega' - \Omega_j) \]

and \(N(\Omega) = [\exp(\beta\Omega) - 1]^{-1}\) is the number of phonons with energy \(\Omega\) in thermal equilibrium. In order to provide a physical interpretation of Eq(2.25) we first normalize the total intensity of the spectrum to unity such that \(f(0) \equiv |\lambda|^2 = 1\) and expand the generating function in a power series of \(\Phi_\pm(t)\). The term \(\Phi_+(t)^n \Phi_-(t)^{n'}\) represents a process where \(n\) phonons are emitted and \(n'\) phonons are simultaneously absorbed and corresponds to the phonon sideband at \(\omega = \tilde{\omega}_0 + \sum_{n'} \Omega_{n'} - \sum_n \Omega_n\). The term with \(n, n' = 0\) is the zero-phonon line at \(\omega = \tilde{\omega}_0\) with intensity \(\exp(-\Phi(0))\). The remaining intensity \(1 - \exp(-\Phi(0))\) is distributed over the sidebands which increases with temperature.

When the temperature of the phonon bath is close to absolute zero, \(N(\Omega) \simeq 0\), and the phonon emission sidebands appear only on the high energy side of the zero-phonon line. At higher temperatures there is enough thermal energy to excite many phonons and the probability of zero-phonon transition is close to zero. For quantum dots the probability of a zero-phonon electronic transition becomes likely at temperatures below 20K and depends on the exciton-phonon coupling and its material properties.

In the simple case of only one vibration mode with frequency \(\Omega_0\) (optical phonon), the lineshape generating function \(f(t)\) is easily expanded in a power series of \(\exp(i\Omega_0 t)\) and can be written as[117]:

\[ f(t) = |\lambda|^2 \exp(-(2N + 1)g) \sum_n e^{\frac{n\Omega_0}{2}} I_n(2g[N(N + 1)]^{1/2}) e^{-i\omega_0 t} e^{-i\Omega_0 t} \]  
(2.26)

where \(I_n(z)\) is the \(n^{th}\) order modified-Bessel function of the 1\(^{st}\) kind with complex argument \(z\) and \(g = \sum_q \eta_q^2 / \Omega_0^2\). In deriving Eq(2.26) we have used the relation \(e^{\frac{im\Omega_0}{2}} = \)
All throughout the above discussion of the lineshape, we have not included the radiative linewidth of the excited state. The exciton in the quantum dot can undergo electron-hole recombination and emit a photon. We denote the linewidth associated with this process by $\Gamma_0(\omega_0)$. Including the radiative linewidth of the excited state, the final expression for the optical lineshape is given by:

$$F(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega t} e^{-i\omega_0 t} e^{-\frac{\Gamma(\omega)}{2} t} e^{-(\Phi(0)-\Phi(t))} dt$$

where $\Phi(t)$ is defined in Eq(2.25). For a bath of optical phonons, $\Phi(t)$ is given by:

$$\Phi(t) = g[N e^{i\Omega t} + (N + 1) e^{-i\Omega t}]$$

The phonon sideband spectrum which in the case of optical phonons consisted of infinitely sharp discrete lines is now broadened by the radiative linewidth $\Gamma_0(\omega_0)/2$.

An important aspect of quantum dot dynamics that is not included either in the stochastic formulation or the semiclassical approach presented above is the decay of phonons due to anharmonic phonon-phonon interactions. As was discussed in the previous chapter, polaronic decay mechanisms (anharmonic processes and phonon-assisted electronic transitions) are important to obtain a complete and experimentally consistent picture. These processes introduce additional linewidth to the optical lineshape and can be introduced in a phenomenological way. We introduce damping rates $\gamma_j$ for the harmonic phonon modes $\Omega_j$ and assume that the phonon modes are underdamped $\gamma_j < \Omega_j$. The critically damped and overdamped cases ($\gamma_j \geq \Omega_j$) can be found in[118] and are not of importance to quantum dot physics. For simplicity we consider the case of only one vibrational mode $\Omega_0$ with damping $\gamma$. The following discussion is adapted from[118, 119]. The lineshape function $f(t)$ Eq(2.24) can now be modified to include phonon damping as (note that we have also included radiative damping):

$$f(t) = |\lambda|^2 e^{-i\omega_0 t - g(2N+1)} e^{-\frac{\gamma t}{2}} [N e^{i\Omega_0 t} + (N + 1) e^{-i\Omega_0 t}]$$

(2.29)
Using the generating function for the Bessel functions we can rewrite Eq(2.29) as:

\[ f(t) = |\lambda|^2 e^{-i\omega_0 t - g(2N + 1)} \]
\[ \times \sum_{m=-\infty}^{\infty} I_m(g e^{-\frac{2l}{\tau} \sqrt{N(N+1)}} e^{-im\Omega_0 t + m\frac{\beta_N}{\tau}} \quad (2.30) \]

A further simplification results by using a series representation of the modified Bessel function \( I_m(x) \):

\[ I_m(x) = \sum_{l=0}^{\infty} \frac{(x^2)^l}{l! \Gamma(m + l + 1)} \quad (2.31) \]

Using Eq(2.31) in Eq(2.30) we finally obtain:

\[ f(t) = |\lambda|^2 e^{-i\omega_0 t - g(2N + 1)} \]
\[ \times \sum_{m=-\infty}^{\infty} \sum_{l=0}^{\infty} \frac{g \sqrt{N(N+1)}}{2^{m+2l} l! \Gamma(m + l + 1)} e^{m\frac{\beta_N}{\tau}} e^{-im\Omega_0 t - (m+2l)\frac{\beta_N}{\tau}} \quad (2.32) \]

The lineshape function now decays due to phonon damping. However, the theory that we have described above is quite rudimentary. The introduction of \( e^{-\frac{2l}{\tau}} \) in the lineshape function \( f(t) \) in Eq(2.29) is arbitrary and cannot be justified on microscopic grounds. Also, it does not take into account the effect of anharmonic phonon processes on the radiative dynamics of the quantum dot and a consistent description in terms of the underlying microscopic processes is lacking.

In the next subsection we write down a full quantum mechanical Hamiltonian of the quantum dot-photon-phonon system. This quantum mechanical Hamiltonian serves as a starting point to describe the interaction of two-level systems with electromagnetic and undamped lattice degrees of freedom. In the following chapters we derive expressions for the lineshape function for a general photonic reservoir. The lineshape function is then evaluated for the unstructured reservoir of free space and the structured reservoir of a photonic crystal. Later on we generalize the model Hamiltonian to include the role of anharmonic phonon processes and phonon-assisted electronic transitions. This allows us to capture the essential physics pertaining to polaron decay and obtain a complete description of quantum dot physics in a photonic crystal.
2.3 Model Hamiltonian for a full quantum mechanical treatment

We now present a model Hamiltonian which describes the dynamics of a quantum dot exciton interacting with a general photonic reservoir and also coupled to bath of phonons. The model Hamiltonian that we write down takes into account relevant physical processes which are central to quantum dot dynamics. It is well known that nonradiative exciton decay in quantum dots caused by phonons can be suppressed due to quantization effects\cite{132, 133}. For instance, in a bulk medium with nearly dispersionless longitudinal-optical(LO) phonons, purely nonradiative decay requires the exciton recombination energy to be an integer multiple of LO phonon energies. This resonance condition leads to the Phonon – Bottleneck effect. In a photonic crystal environment, in which quantum dots are embedded in periodic array of sub-micron scale dielectric rods, such bottlenecks may apply to acoustic phonons as well. Quantum confinement in the quantum dot increases the coupling of electrons to short-wavelength phonons. This strengthens multiple phonon-assisted absorption and emission processes. Phonon sidebands exist for both acoustic and optical phonons. Optical phonons give rise to discrete sidebands separated by the optical phonon energy. Acoustic phonons in a bulk medium occupy a broad continuum of energies and give rise to significant line broadening.

We consider a quantum dot (two-level system) with level separation much larger than all phonon energies so that phonon induced mixing of different electronic levels can be neglected. The full Hamiltonian of our dot exciton, phonon and photon system can be written as $H = H_0 + H_I$ where

$$H_0 = \hbar \omega_0 \hat{\sigma}_{ee} + \sum_k \hbar \omega_k \hat{a}_k^\dagger \hat{a}_k + \sum_q \hbar \Omega_q \hat{c}_q^\dagger \hat{c}_q$$

$$H_I = \sum_k (\lambda_k \hat{\sigma}_{eg} \hat{a}_k + \lambda_k^* \hat{c}_k^\dagger \hat{\sigma}_{ge}) + \hat{\sigma}_{ee} \sum_q \eta_q (\hat{c}_q + \hat{c}_q^\dagger)$$

where $\hat{\sigma}_{eg}(\hat{\sigma}_{ge})$ is the raising(lowering) operator for the two-level atom, $\hat{c}_q^\dagger(\hat{c}_q)$ is the creation(annihilation) operator for the phonon with wavenumber $q$, and $\hat{a}_k^\dagger(\hat{a}_k)$ is the creation(annihilation) operator for the photon. $\lambda_k$ and $\eta_q$ are exciton-phonon and exciton-phonon coupling parameters, respectively. $\omega_0$ is the resonant frequency of the two-level atom and $\Omega_q$ and $\omega_k$ are the phonon and photon dispersion relations respectively. The
two-level atom operators obey the commutation relation:

$$[\hat{\sigma}_{ij}, \hat{\sigma}_{kl}] = \delta_{jk}\hat{\sigma}_{il} - \delta_{il}\hat{\sigma}_{kj}$$  \hspace{1cm} (2.34)

The photon and phonon operators obey bosonic commutation relations: $$[\hat{a}_k, \hat{a}^\dagger_{k’}] = \delta(k - k’)$$ and $$[\hat{c}_q, \hat{c}^\dagger_{q’}] = \delta(q - q’)$$. In the absence of dot-photon coupling ($\lambda_k = 0$), the Hamiltonian Eq(2.33) reduces to the “Independent – Boson” model used in the literature [52] to study pure dephasing of two-level systems coupled to a phonon bath (see chapter 1.3.2). Optical interaction with quantum dots in a smooth, featureless electromagnetic reservoir can be treated in the standard Fermi-golden rule framework. However, in the “colored vacuum” of a photonic crystal, abrupt changes in the photon density of states (with frequency) introduce non-Markovian memory effects [134] in the quantized electromagnetic field. The resonant interaction of the two-level system with the quantized electromagnetic field is written in a rotating wave approximation, where the dot-photon coupling constant is given by Eq(1.7).

The interaction of the electron with phonons both shifts the exciton recombination energy and leads to scattering effects. In order to isolate the overall energy shift, we use the (canonical) polaron – transformation [130]:

$$\tilde{H} = \exp(S)H\exp(-S)$$  \hspace{1cm} (2.35a)

where

$$S = \hat{\sigma}_{ee} \sum_q \frac{\eta_q}{\Omega_q} (\hat{c}^\dagger_q - \hat{c}_q)$$  \hspace{1cm} (2.35b)

For convenience we choose units such that $\hbar = 1$. The canonical transformation diagonalizes the Hamiltonian Eq(2.33) to the form $\tilde{H} = \tilde{H}_0 + H_p$ where

$$\tilde{H}_0 = (\omega_0 - \Delta)\hat{\sigma}_{ee} + \sum_k \omega_k \hat{a}_k^\dagger \hat{a}_k + \sum_k (\lambda_k \hat{C}_+ \hat{\sigma}_{eg} \hat{a}_k + \lambda^*_k \hat{a}_k^\dagger \hat{C}_- \hat{\sigma}_{qe})$$  \hspace{1cm} (2.36a)

$$H_p = \sum_q \Omega_q \hat{c}^\dagger_q \hat{c}_q$$  \hspace{1cm} (2.36b)

Here we have introduced a lattice displacement operator:

$$\hat{C}_\pm = \exp(\pm \sum_q \frac{\eta_q}{\Omega_q} (\hat{c}^\dagger_q - \hat{c}_q))$$  \hspace{1cm} (2.37a)
and

$$\Delta = \sum_q \frac{\eta_q^2}{\Omega_q} \quad (2.37b)$$

\(\Delta\) is called the polaron shift and accounts for the renormalization of the resonant frequency of the two-level atom due to phonon emission and reabsorption.

A mean-field theory of the electron-thermal lattice vibration interaction is obtained by replacing \(\hat{C}_+\) and \(\hat{C}_-\) by their thermal expectation values \(\langle \hat{C}_+ \rangle\) and \(\langle \hat{C}_- \rangle\) evaluated with respect to the pure phonon Hamiltonian \(H_p[130]\)

$$\langle \hat{C}_\pm \rangle = \frac{Tr(\exp(-\beta H_p)\hat{C}_\pm)}{Tr(\exp(-\beta H_p))} = \exp\left(-\frac{1}{2} \sum_q \frac{\eta_q^2}{\Omega_q} (1 + 2\langle \hat{c}_q^\dagger \hat{c}_q \rangle)\right) \quad (2.38)$$

As we show, this approximation can be used to recapture important features of the exciton lineshape caused by multi-phonon absorption and emission sidebands. The thermal averages of the phonon operators are evaluated assuming that the phonon bath is at thermal equilibrium at temperature \(T\) and \(\beta = 1/k_B T\). The Hamiltonian Eq(2.36) is simplified to

$$H_{MF} = (\omega_0 - \Delta)\hat{\sigma}_{ee} + \sum_k \omega_k \hat{a}_k^\dagger \hat{a}_k + \sum_k \left(\tilde{\lambda}_k \hat{\sigma}_{eg} \hat{a}_k + \tilde{\lambda}_k^* \hat{a}_k^\dagger \hat{\sigma}_{ge}\right) \quad (2.39)$$

where \(\tilde{\lambda}_k = \lambda_k \langle \hat{C}_+ \rangle\) and \(\tilde{\lambda}_k^* = \lambda_k^* \langle \hat{C}_- \rangle\). This mean-field approximation relies on the separation of time scales between optical and vibrational processes for realistic systems in which \(\lambda_k << \eta_q\). Since \(\hbar/\lambda_k >> \hbar/\eta_q\), phonon scattering occurs very rapidly compared to purely radiative processes. In other words the dot-phonon system can reach equilibrium quickly in response to any slow change arising from radiative processes.

Despite the possible non-Markovian nature of dot-photon interaction in a photonic crystal, it is still very weak compared to dot-phonon interaction. The dot-photon coupling constant, \(\lambda_k\), has an upper bound obtained by choosing \(V = a^3\), the Wigner-Seitz cell volume of a photonic crystal with lattice constant \(a\). The dipole moment \(D_{ge}\) is given by the spatial average of the exciton wavefunction when the electron and the hole are at the same position and is of the order of \(10^{-28}\text{Coul} - m\ [52]\). We consider a quantum dot with a transition near optical frequencies \(\omega_0 \simeq 10^{15}\text{s}^{-1} \simeq 1\text{ev}\) coupled to photons near a PBG centred at \(a\omega/2\pi c \simeq 0.8\). Using this we obtain a dot-photon coupling \(\lambda_k \simeq 1\mu\text{ev}\).

For the quantum dot-phonon coupling \(\eta_q\), as an example we assume the standard
Frohlich coupling \cite{121} $\eta_q = \frac{A}{V^q q}$, where

$$A^2 = \frac{\hbar \Omega q e^2}{2 \epsilon_0} \left( \frac{1}{\kappa_{\infty}} - \frac{1}{\kappa_0} \right)$$

(2.40)

$q = |q|$, $\kappa_{\infty}$ and $\kappa_0$ are high-frequency and static dielectric constants respectively. $V'$ is the volume of the electronic unit-cell of the quantum dot material. The dispersion relation for optical phonons can be approximated as

$$\Omega_q^2 = c_1 + [c_1^2 - c_2 \sin^2 \left( \frac{\pi q}{2k_{\max}} \right)]^{\frac{1}{2}}$$

(2.41)

where $c_1 = 1500 \text{ps}^{-2}$, $c_2 = 2 \times 10^6 \text{ps}^{-4}$ and for an fcc lattice $k_{\max} = \frac{4\pi}{a_e} (a_e$ is the electronic lattice constant $a_e \approx .35 \text{nm}$). Using these typical values, $\eta_q \approx 1 \text{mev}$. The timescale of dot-phonon interaction is of the order of picoseconds whereas the timescale for dot-radiation processes is nanoseconds. Therefore, eliminating the lattice degrees of freedom in this way, we introduce an effective temperature dependent optical dipole moment coupling the dot to the radiation reservoir. This enables us to recapture the semiclassical picture of multi-phonon assisted optical transitions and allows us to explore non-Markovian radiative dynamics in the photonic crystal.

Finally in this chapter, we mention that problems with similar mathematical structure have been studied in the literature in other contexts. One example is the work of Hewson and Newns on polaron effects in intermediate-valence compounds\cite{135, 136}. Similar problems arise in the context of image charge effects in chemisorption\cite{137, 138}. The mean-field decomposition we use in this thesis has also been considered by Anda and Ure\cite{137} in the context of chemisorption of Hydrogen. The theory of the Auger effect in solids where plasmonic excitations are the analogues of phonons in our model, likewise consider a similar Hamiltonian to ours\cite{139, 140}. In the next chapter we use the mean-field Hamiltonian Eq(2.39) to calculate the optical susceptibility and lineshape for the full quantum mechanical Hamiltonian Eq(2.33).
Chapter 3

Optical susceptibility and lineshape for the full quantum mechanical Hamiltonian

In this chapter we present a general formalism to calculate the absorption lineshape, polarization dynamics and lifetime decay of a quantum dot exciton coupled to a general electromagnetic reservoir in the presence of a thermal reservoir of phonons. The absorption lineshape and the polarization dynamics is computed using the method of thermodynamic Green’s functions. The population dynamics is studied using the Heisenberg equations of motion. In Appendix B we present a generalization of the method of thermodynamic Green’s functions, the method of generalized-Laplace transforms and use it to study the nonequilibrium problem of population decay of an initially excited two-level atom.

3.1 Susceptibility using the method of thermodynamic Green’s function

We begin by deriving a general formalism to calculate the absorption lineshape and the rate of decay of an exciton coupled to a general photonic and a thermal phonon reservoir. The absorption lineshape $\chi_\alpha(\omega)$ is related to the two-time dipole correlation function [77]:

$$\chi_\alpha(\omega) = \frac{i N_d}{\epsilon_0 \hbar} \int_{-\infty}^{0} dt \exp(-i \omega t) \langle \hat{d}(0), \hat{d}(t) \rangle$$  (3.1)
where the angular brackets denote thermodynamic averages and $\hat{d}(t) = \exp(iHt)\hat{d}(0) \exp(-iHt)$ where $H$ is defined in Eq(2.33) and $\hat{d}(0)$ is defined below. Also $N_d$ is the number of dipoles in a unit volume. Using the fact that the equilibrium is invariant under translations in time, Eq(3.1) can also be written as:

$$\chi_a(\omega) = \frac{iN_d}{\epsilon_0\hbar} \int_{-\infty}^{\infty} dt \Theta(t) \exp(i\omega t) \langle [\hat{d}(t), \hat{d}(0)] \rangle$$  \hspace{1cm} (3.2)

where the step function $\Theta(t)$ is defined by $\Theta(t) = 1$ for $t \geq 0$ and $\Theta(t) = 0$ otherwise.

### 3.1.1 Absorption lineshape

The absorption lineshape Eq(3.2) can, in turn, be related to the thermodynamic Green’s function defined as [141, 142]

$$G_t(\hat{A}(t); \hat{B}(t')) = -i\Theta(t - t')\langle [\hat{A}(t), \hat{B}(t')] \rangle_H$$  \hspace{1cm} (3.3)

Here $\hat{A}(t)$ and $\hat{B}(t')$ are in the Heisenberg representation and the expectation value is calculated with respect to the density operator for the full Hamiltonian Eq(2.33). Using invariance under time translation, the Fourier transform of the Green’s function Eq(3.3) is given by $G_\omega(\hat{A}; \hat{B}) = \int_{-\infty}^{\infty} dt G_t(\hat{A}(t); \hat{B}(0)) \exp(i\omega t)$. Differentiating $G_t(\hat{A}(t); \hat{B}(t'))$ with respect to $t$, it follows that $G_\omega(\hat{A}; \hat{B})$ satisfies the equation of motion[143]:

$$\omega G_\omega(\hat{A}; \hat{B}) = \langle [\hat{A}(0), \hat{B}(0)] \rangle_H + G_\omega([\hat{A}, H]; \hat{B}).$$  \hspace{1cm} (3.4)

Here, the first term on the RHS is the equal time commutator evaluated at thermal equilibrium. In practice an approximate solution for $G_\omega(\hat{A}; \hat{B})$ is obtained from its equation of motion. In general this leads to a hierarchy of equations relating $G_\omega(\hat{A}; \hat{B})$ to higher order quantum correlation functions. Truncating the hierarchy under some suitable physical assumptions, yields a mean-field approximation for the relevant susceptibilities.

The susceptibility $\chi_a(\omega)$ describes the linear response of a two-level atom placed in a normalization volume $V$ to an external field of frequency $\omega$. The dipole moment operator $\hat{d}$ of an isotropic two-level system is given by $\hat{d} = |D_{ge}|(\hat{\sigma}_{eg} + \hat{\sigma}_{ge})$ where $|D_{ge}|$ is the transition dipole moment connecting the ground($|g\rangle$) and the excited($|e\rangle$) states.
Using Eqs (3.2) and (3.3) the susceptibility is given by

\[
\chi_a(\omega) = -\frac{N_d}{\epsilon_0\hbar} |D_{ge}|^2 \lim_{\epsilon \to 0^+} [G_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) + G_{\omega+i\epsilon}(\hat{\sigma}_{eg}; \hat{\sigma}_{ge})] + G_{\omega+i\epsilon}(\hat{\sigma}_{eg}; \hat{\sigma}_{eg}) + G_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{ge})].
\] (3.5)

The infinitesimal imaginary part \(\epsilon\) is added to shift the poles of \(\chi(\omega)\) to the lower half of the complex \(\omega\)-plane. The susceptibility \(\chi_a(\omega)\) is well-defined for positive and negative frequencies. The Green’s function satisfies the symmetry property \(G_{\omega}(\hat{A}; \hat{B}) = G_{-\omega}(\hat{B}; \hat{A})\) [130] as verified from its spectral representation:

\[
G_{\omega}(\hat{A}; \hat{B}) = Z^{-1} \sum_{m,n} \exp\left(-\frac{E_m}{K_BT}\right) - \exp\left(-\frac{E_n}{K_BT}\right) \langle n|\hat{A}|m\rangle \langle m|\hat{B}|n\rangle.
\] (3.6)

Here, \(Z^{-1} = Tr(\exp(-\hat{H}/K_BT))\) and \(|n\rangle\) and \(E_n\) are the exact eigenstates and eigenvalues of the Hamiltonian \(\hat{H}\) (assumed to have no explicit time-dependence). Therefore \(G_{\omega+i\epsilon}(\hat{\sigma}_{eg}; \hat{\sigma}_{ge})\) can be calculated from \(G_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg})\) by substituting \(\omega\) with \(-\omega\). Moreover \(G_{\omega+i\epsilon}(\hat{\sigma}_{eg}; \hat{\sigma}_{eg})\) and \(G_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{ge})\) are zero as we show in Appendix A. In the following we focus on the evaluation of \(G_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg})\) in the expression for optical susceptibility Eq (3.5).

Consider the Green’s function in the time domain

\[
G_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) = -i\Theta(t) (\langle [\hat{\sigma}_{ge}(t), \hat{\sigma}_{eg}(0)] \rangle)_H
\] (3.7)

\[
= -i\Theta(t) \langle (e^{i\hat{H}t}\hat{\sigma}_{ge}e^{-i\hat{H}t}\hat{\sigma}_{eg})_H - \langle \hat{\sigma}_{eg}e^{i\hat{H}t}\hat{\sigma}_{ge}e^{-i\hat{H}t}\rangle_H \rangle
\]

where the correlation functions are evaluated in thermodynamic equilibrium with respect to the full Hamiltonian Eq (2.33). Here and throughout the rest of this paper we use units in which \(\hbar = 1\). Using the fact that the trace is invariant under unitary transformations we get

\[
G_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) = \tilde{G}_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \equiv -i\Theta(t) \langle (e^{i\tilde{H}t}\hat{\sigma}_{ge}e^{-i\tilde{H}t}\hat{\sigma}_{eg})_H - \langle \hat{\sigma}_{eg}e^{i\tilde{H}t}\hat{\sigma}_{ge}e^{-i\tilde{H}t}\rangle_H \rangle
\] (3.8)

where \(\tilde{H}\) is the polaron-transformed Hamiltonian defined in Eq (2.36), \(\hat{\sigma}_{ge} = \exp(S)\hat{\sigma}_{ge}\exp(-S) = \hat{\sigma}_{ge}\hat{C}_-\), \(\hat{\sigma}_{eg} = \exp(S)\hat{\sigma}_{eg}\exp(-S) = \hat{\sigma}_{eg}\hat{C}_+\), \(S = \hat{\sigma}_{ee} \sum_q \frac{\psi_q}{\hbar} (\hat{c}_q^+ - \hat{c}_q)\), and \(\hat{C}_\pm\) are defined in Eq (2.37). The trace can be evaluated in a closed form in a mean-field approximation
that replaces $\hat{H}$ with $H_{MF} + H_p$. With respect to the Hamiltonian Eq(2.39) we have

$$
\hat{G}_t^{MF}(\sigma_{ge}; \sigma_{eg}) = -i \Theta(t) (\langle e^{iH_{MF} t} \sigma_{ge} e^{-iH_{MF} t} \sigma_{eg} \rangle_{H_{MF}} (e^{iH_p t} \hat{C}_- e^{-iH_p t} \hat{C}_+)_{H_p} (3.9)
- \langle \sigma_{eg} e^{iH_{MF} t} \sigma_{ge} e^{-iH_{MF} t} \rangle_{H_{MF}} (\hat{C}_+ e^{iH_p t} \hat{C}_- e^{-iH_p t})_{H_p})
- \langle \sigma_{eg} e^{iH_{MF} t} \sigma_{ge} e^{-iH_{MF} t} \rangle_{H_{MF}} (\hat{C}_+ (0) \hat{C}_- (0))_{H_p})
$$

where $\hat{G}_t^{MF}(\sigma_{ge}; \sigma_{eg})$ is the Green’s function calculated with respect to the polaron transformed mean field Hamiltonian Eq(2.39) plus the phonon Hamiltonian Eq(2.36b). The thermal averages over the phonon modes are well known and are given by[130]

$$
\langle \hat{C}_\pm (t) \hat{C}_\mp (0) \rangle_{H_p} = e^{-(\Phi(0) - \Phi(t))}
$$
(3.10a)

$$
\langle \hat{C}_\pm (0) \hat{C}_\mp (t) \rangle_{H_p} = e^{-(\Phi(0) - \Phi(-t))}
$$
(3.10b)

where $\Phi(t) = \Phi_+(t) + \Phi_-(t)$ and

$$
\Phi_+(t) = \sum_q \frac{\eta_q^2}{\Omega_q^2} N_q \exp(i\Omega_q t)
$$
(3.11)

$$
\Phi_-(t) = \sum_q \frac{\eta_q^2}{\Omega_q^2} (N_q + 1) \exp(-i\Omega_q t)
$$

and $N_q = \langle \hat{c}_q^\dagger \hat{c}_q \rangle = (\exp(\beta \Omega_q) - 1)^{-1}$. From Eq(2.38) we see that( we have dropped the subscript $H_p$ on the thermal averages)

$$
\langle \hat{C}_\pm \rangle \langle \hat{C}_\pm \rangle = \exp(-\sum_q \frac{\eta_q^2}{\Omega_q^2} (1 + 2N_q)) = e^{-\Phi(0)}.
$$
(3.12)

For illustration purposes we now consider the Einstein model of dispersionless phonons[130]. In this case $\Phi(t) = \Phi_+(t) + \Phi_-(t)$ where

$$
\Phi_+(t) = gN \exp(i\Omega_0 t)
$$
(3.13)

$$
\Phi_-(t) = g(N + 1) \exp(-i\Omega_0 t)
$$

where $g = \sum_q \frac{\eta_q^2}{\Omega_q^2}$ and $N = \frac{1}{\exp(\beta \omega_0) - 1}$ is the number of thermally excited optical phonons.

We now relate the correlation functions $\langle e^{iH_{MF} t} \sigma_{ge} e^{-iH_{MF} t} \sigma_{eg} \rangle_{H_{MF}}$ and $\langle \sigma_{eg} e^{iH_{MF} t} \sigma_{ge} e^{-iH_{MF} t} \rangle_{H_{MF}}$. 

...
to the corresponding Green’s functions $G^{MF}_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \equiv -i\Theta(t)\langle[\hat{\sigma}_{ge}(t), \hat{\sigma}_{eg}(0)]\rangle_{HMF} (H_{MF} \text{ is defined in Eq(2.39})$ and $G^{MF}_\omega(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i\omega t} G^{MF}_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg})$ using the inversion formulae[142, 143]:

$$\langle e^{iH_{MF}t}\hat{\sigma}_{ge}e^{-iH_{MF}t}\hat{\sigma}_{eg} \rangle_{H_{MF}} = -\frac{1}{2\pi i} \int d\omega e^{-i\omega t} \frac{e^{\beta_s \omega}}{\epsilon_{\beta_s \omega} + 1} G^{MF}_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \quad (t > 0)$$

(3.14a)

$$\langle \hat{\sigma}_{eg}e^{iH_{MF}t}\hat{\sigma}_{ge}e^{-iH_{MF}t} \rangle_{H_{MF}} = -\frac{1}{2\pi i} \int d\omega e^{-i\omega t} \frac{1}{\epsilon_{\beta_s \omega} + 1} G^{MF}_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \quad (t > 0)$$

(3.14b)

where $\beta_s$ is the equilibrium temperature of the two-level system interacting with the photon reservoir. The contour consists of the real axis and a large semicircle in the lower-half plane. The integral over the semicircle tends to zero as the radius tends to infinity. Since the spectral weight of $G^{MF}_{\omega}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg})$ is concentrated near $\omega \simeq \omega_0$, we can evaluate the thermal weight factors at frequency $\omega_0$ and bring them out of the integrals. Since the equilibrium temperature is assumed very low compared to the two-level transition frequency ($\beta_s \omega_0 >> 1$), we conclude that:

$$\langle e^{iH_{MF}t}\hat{\sigma}_{ge}e^{-iH_{MF}t}\hat{\sigma}_{eg} \rangle_{H_{MF}} \simeq -\frac{1}{2\pi i} \int d\omega e^{-i\omega t} G^{MF}_{\omega+i\epsilon}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \quad (t > 0)$$

$$\langle \hat{\sigma}_{eg}e^{iH_{MF}t}\hat{\sigma}_{ge}e^{-iH_{MF}t} \rangle_{H_{MF}} \simeq 0$$

(3.15)

Therefore using Eq(3.15) in Eq(3.9) we obtain:

$$\tilde{G}^{MF}_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \simeq G^{MF}_t(\hat{\sigma}_{ge}; \hat{\sigma}_{eg})e^{-i(\Phi(0) - \Phi(t))} \quad (t > 0)$$

(3.16)

Expanding exp($-(\Phi(0) - \Phi(t))$) in a power series in exp($i\Omega_0 t$), it can be shown [130] that the Green’s function has the form

$$\tilde{G}^{MF}_\omega(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) = \sum_{n=\infty}^{\infty} L_n G^{MF}_{\omega-n\Omega_0}(\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \quad (t > 0)$$

(3.17)

where

$$L_n = e^{-g(2N+1)} e^{i\Omega_0 \beta_s / 2} I_n(2g \sqrt{N(N+1)})$$

(3.18)

and $I_n(z)$ is the $n^{th}$ order modified-Bessel function of the $1^{st}$ kind with complex argument $z$. Here we have made use of the relation $\sqrt{(N+1)/N} = e^{\frac{\omega_0}{2}}$ as well as the identity[131] $e^{z \cos(\theta)} = \sum_{n=-\infty}^{\infty} I_n(z)e^{in\theta}$. The $n^{th}$ term in this infinite series represents the net change
in phonon number during an optical transition. For \( n \geq 1 \), this corresponds to phonon-assisted optical transitions where \( n \) more phonons are emitted than absorbed by the quantum dot into the phonon reservoir. The detailed phonon sideband interpretation \([9, 50]\) of Eq(3.17) follows from a double power series expansion

\[
\langle \hat{C}_-(t)\hat{C}_+(0) \rangle = e^{-\Phi(0)} \sum_{n_+,n_-=0}^{\infty} \frac{(\Phi_+(t))^{n_+}(\Phi_-(t))^{n_-}}{(n_+!)(n_-!)}. \tag{3.19}
\]

This series describes all possible phonon interactions in which \( n_+ \) phonons are emitted by the reservoir and \( n_- \) phonons are simultaneously absorbed. Comparing the Fourier transform of Eq(3.19) with Eq(3.17), we may identify \( n = n_- - n_+ \). Analogous results can be derived for dispersive phonon-models, e.g. acoustic phonons. However a simple power series expansion of the acoustic phonon Green’s function cannot be made and recourse must be made to numerical methods. In order to simplify the notation we denote \( G_{MF,\omega,n}^{\geq\leq} (\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \equiv G_{\omega,n}^{MF} (\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \). It is easy to see that \( G_{\omega,n}^{MF} (\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) \) satisfies the equation of motion:

\[
(\omega - n\Omega_0)G_{\omega,n}^{MF} (\hat{\sigma}_{ge}; \hat{\sigma}_{eg}) = \langle [\hat{\sigma}_{ge}(0), \hat{\sigma}_{eg}(0)] \rangle_{H_{MF}} + G_{\omega,n}^{MF} ([\hat{\sigma}_{ge}, H_{MF}], \hat{\sigma}_{eg}). \tag{3.20}
\]

The first term on the RHS is equal to unity in thermal equilibrium since \( \langle \hat{\sigma}_{gg} \rangle \simeq 1 \) and \( \langle \hat{\sigma}_{ee} \rangle \simeq 0 \) for \( \hbar \omega_0 >> k_B T \). The corresponding equation of motion for \( G_{\omega,n}^{MF} ([\hat{\sigma}_{ge}, H_{MF}], \hat{\sigma}_{eg}) \) leads to a hierarchy of equations involving higher order correlation functions. This hierarchy can be closed by decoupling certain photon correlation functions from atomic correlation functions. Physically, this decoupling scheme corresponds to neglecting multiphoton processes and considering only temperatures very low compared to the quantum dot transition energy scale. On the other hand, multiphonon processes are retained to all orders.
3.2 Phonon sideband contribution to mean-field thermodynamic Green’s function

Evaluating the commutator $[\hat{\sigma}_{ge}, H_{MF}]$ in Eq(3.20) using the mean-field Hamiltonian Eq(2.39), we find

$$[\hat{\sigma}_{ge}, H_{MF}] = (\omega_0 - \Delta)\hat{\sigma}_{ge} + \sum_k \lambda_k (\hat{\sigma}_{gg} - \hat{\sigma}_{ee})\hat{a}_k.$$  \hspace{1cm} (3.21)

Using Eq(3.21) we find

$$(\omega - n\Omega_0)G_{\omega,n}^{MF}([\hat{\sigma}_{ge}, H_{MF}], \hat{\sigma}_{eg}) = 1 + (\omega_0 - \Delta)G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$$

$$+ \sum_k \lambda_k G_{\omega,n}^{MF}((\hat{\sigma}_{gg} - \hat{\sigma}_{ee})\hat{a}_k, \hat{\sigma}_{eg}).$$ \hspace{1cm} (3.22)

In order to close the system of equations, we need to express $G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg})$ and $G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg})$ in terms of $G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$. The equation of motion for $G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg})$ is

$$(\omega - n\Omega_0)G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg}) = \langle [\hat{\sigma}_{ee}\hat{a}_k(0), \hat{\sigma}_{eg}(0)] \rangle_{H_{MF}} + G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, H_{MF}, \hat{\sigma}_{eg}).$$ \hspace{1cm} (3.23)

The first term on the right hand side is identically zero. The commutator $[\hat{\sigma}_{ee}\hat{a}_k, H_{MF}]$ is

$$[\hat{\sigma}_{ee}\hat{a}_k, H_{MF}] = \omega_k \hat{\sigma}_{ee}\hat{a}_k + \sum_q \lambda_q \hat{\sigma}_{eg}\hat{a}_k\hat{a}_q - \sum_q \lambda_q^* \hat{\sigma}_{ge}\hat{a}_q^\dagger\hat{a}_k.$$ \hspace{1cm} (3.24)

Using Eq(3.24) in Eq(3.23) we obtain an equation of motion for $G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg})$:

$$(\omega - n\Omega_0)G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg}) = \omega_k G_{\omega,n}^{MF}(\hat{\sigma}_{ee}\hat{a}_k, \hat{\sigma}_{eg}) + \sum_q \lambda_q G_{\omega,n}^{MF}(\hat{\sigma}_{eg}\hat{a}_k\hat{a}_q, \hat{\sigma}_{eg})$$

$$- \sum_q \lambda_q^* G_{\omega,n}^{MF}(\hat{\sigma}_{ge}\hat{a}_q^\dagger\hat{a}_k, \hat{\sigma}_{eg}).$$ \hspace{1cm} (3.25)

It is easy to see that the Green’s functions $G_{\omega,n}^{MF}(\hat{\sigma}_{eg}\hat{a}_k\hat{a}_q, \hat{\sigma}_{eg})$ and $G_{\omega,n}^{MF}(\hat{\sigma}_{ge}\hat{a}_q^\dagger\hat{a}_k, \hat{\sigma}_{eg})$ generate equations of motion involving higher powers of photon operators. In order to close this hierarchy of equations we decouple the two-level system and photon operators in the Green’s function by replacing the photon operators with their thermal expectation.
The contribution of the procedure outlined above is equivalent to considering one-photon processes and ignoring all multiphoton contributions. Note, however, that the phonon processes are included to obtain equations of motion involving higher order photon operators. We decouple the two-level system and photon operators in the Green’s function as before to obtain

\[ G_{\omega,n}^{MF}(\hat{a}_k^\dagger \hat{a}_q \hat{a}_{\sigma q} \hat{q}), \hat{\sigma}_{\sigma q}) \approx \delta_{kq} \langle \hat{a}_k^\dagger \hat{a}_q \hat{a}_{\sigma q} \hat{q} \rangle G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q}, \hat{\sigma}_{\sigma q}) \] (3.26)

\[ G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k^\dagger \hat{a}_q, \hat{\sigma}_{\sigma q}) \approx \delta_{kq} \langle \hat{a}_k^\dagger \hat{a}_q \hat{a}_{\sigma q} \hat{q} \rangle G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q}, \hat{\sigma}_{\sigma q}) \]

We observe that in thermal equilibrium \( \langle \hat{a}_k \hat{\sigma}_{\sigma q} \rangle = \langle \hat{a}_k^\dagger \hat{\sigma}_{\sigma q} \rangle \approx 0 \). Hence we conclude that the contribution of \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) \) to \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q}, \hat{\sigma}_{\sigma q}) \) can be ignored. The decoupling procedure outlined above is equivalent to considering one-photon processes and ignoring all multiphoton contributions. Note, however, that the phonon processes are included to all orders because of the polaron transformation.

We now consider the equation of motion for \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) \). This is given by

\[ (\omega - n\Omega_0)G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) = \langle [\hat{\sigma}_{\sigma q} \hat{a}_k(0), \hat{\sigma}_{\sigma q}(0)] \rangle_{H_{MF}} + G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, H_{MF}, \hat{\sigma}_{\sigma q}). \] (3.27)

The first term on the right hand side is identically zero. The commutator \([\hat{\sigma}_{\sigma q} \hat{a}_k, H_{MF}]\) is now evaluated to obtain

\[ [\hat{\sigma}_{\sigma q} \hat{a}_k, H_{MF}] = \omega_k \hat{\sigma}_{\sigma q} \hat{a}_k - \sum_q \lambda_q \hat{\sigma}_{\sigma q} \hat{a}_q \hat{a}_k + \sum_q \lambda_q^* \hat{\sigma}_{\sigma q} \hat{a}_k \hat{a}_q^\dagger. \] (3.28)

Using Eq(3.28) in Eq(3.27) we obtain an equation of motion for \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) \):

\[ (\omega - n\Omega_0)G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) = \omega_k G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) - \sum_q \lambda_q G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_q \hat{a}_k, \hat{\sigma}_{\sigma q}) \]

\[ + \sum_q \lambda_q^* G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k \hat{a}_q^\dagger, \hat{\sigma}_{\sigma q}). \] (3.29)

We note that the Green’s functions \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) \) and \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k \hat{a}_q^\dagger, \hat{\sigma}_{\sigma q}) \) generate equations of motion involving higher order photon operators. We decouple the two-level system and photon operators in the Green’s function as before to obtain

\[ G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k, \hat{\sigma}_{\sigma q}) \approx \delta_{kq} \langle \hat{a}_k \hat{a}_k \rangle G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q}, \hat{\sigma}_{\sigma q}) \] (3.30)

\[ G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k \hat{a}_q^\dagger, \hat{\sigma}_{\sigma q}) \approx \delta_{kq} (1 + \langle \hat{a}_k^\dagger \hat{a}_k \rangle) G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q}, \hat{\sigma}_{\sigma q}) \]

Since in thermal equilibrium \( \langle \hat{a}_k \hat{a}_k \rangle = \langle \hat{a}_k^\dagger \hat{a}_k \rangle \approx 0 \), we conclude that \( G_{\omega,n}^{MF}(\hat{\sigma}_{\sigma q} \hat{a}_k \hat{a}_k, \hat{\sigma}_{\sigma q}) \approx \)
0 and $G_{\omega,n}^{MF}(\hat{\sigma}_{gg} \hat{a}_k, \hat{\sigma}_{eg})$ can be expressed in terms of $G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$ by using Eq(3.30) in Eq(3.29) as

$$G_{\omega,n}^{MF}(\hat{\sigma}_{gg} \hat{a}_k, \hat{\sigma}_{eg}) \approx \lambda_k^* G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) \frac{1}{\omega - n\Omega_0 - \omega_k}.$$  \hfill (3.31)

Eliminating $G_{\omega,n}^{MF}(\hat{\sigma}_{gg} \hat{a}_k, \hat{\sigma}_{eg})$ between Eqs(3.22) and (3.31) and using $G_{\omega,n}^{MF}(\hat{\sigma}_{eg} \hat{a}_k, \hat{\sigma}_{eg}) \approx 0$, we obtain for $G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$ using Eq(3.20):

$$G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) = 1 + (\omega_0 - \Delta)G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) + \sum_k \frac{|\lambda_k|^2}{\omega - n\Omega_0 - \omega_k} G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}).$$ \hfill (3.32)

Solving for $G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$ we obtain Eq(3.33)

$$G_{\omega,n}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) = -\frac{1}{\omega_0 - \Delta - (\omega - n\Omega_0) - \sum_k \frac{|\lambda_k|^2}{\omega_k - (\omega - n\Omega_0)}} \hfill (3.33)$$

The same procedure can be used to calculate $G_{\omega}(\hat{\sigma}_{eg}, \hat{\sigma}_{ge})$ by using the mean-field approximation to write $G_{\omega}(\hat{\sigma}_{eg}, \hat{\sigma}_{ge}) \approx e^{-(\Phi(0) - \Phi(t))} G_{\omega}^{MF}(\hat{\sigma}_{eg}, \hat{\sigma}_{ge})$. The Fourier transform of $G_{\omega}^{MF}(\hat{\sigma}_{eg}, \hat{\sigma}_{ge})$, $G_{\omega}^{MF}(\hat{\sigma}_{eg}, \hat{\sigma}_{ge})$, can be obtained from the expression for $G_{\omega}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$ using the symmetry properties of the Green’s functions. The absorption spectrum $\chi_a^n(\omega)$ is defined as the imaginary part of the susceptibility Eq(3.5) and using $G_{\omega}(\hat{\sigma}_{ge}, \hat{\sigma}_{ge}) = G_{\omega}(\hat{\sigma}_{eg}, \hat{\sigma}_{eg}) = 0$ (see Appendix A), we obtain:

$$\chi_a^n(\omega) = \frac{N_d}{\epsilon_0 \hbar} |D_{ge}|^2 \text{Im} \left[ \lim_{\epsilon \to 0} \sum_n L_n \frac{1}{\omega_0 - \Delta - (\omega - n\Omega_0)} - \sum_k \frac{|\lambda_k|^2}{\omega_k - (\omega - n\Omega_0) - i\epsilon} \right] \hfill (3.34)$$

Equation(3.34) is one of the central formal results of this chapter. The sideband structure is clearly seen in this expression. The self-energy terms $\sum_k \frac{|\lambda_k|^2}{\omega_k - (\omega - n\Omega_0) - i\epsilon}$ contain the details of the electromagnetic reservoir. We restrict our attention to those values of $n$ and $\Omega_0$ such that $\omega - n\Omega_0$ is close to the quantum dot transition frequency $\omega_0$. Terms with large values of $n$ have vanishingly small spectral weights due to the rapid decrease of $L_n$ with $n$ and the contribution of these sidebands to the lineshape is negligible. Since $\omega_k$ is positive-definite, the second term in the expression for the optical
susceptibility has a negligible imaginary part and can be dropped. We finally have
\[
\chi^n_a(\omega) = \frac{N_d}{\epsilon_0 \hbar} |D_{ge}|^2 \text{Im} \lim_{\epsilon \rightarrow 0} \sum_n L_n \frac{1}{\omega_0 - \Delta - (\omega - n\Omega_0) - \sum_k \frac{\lambda_k^2}{\omega_k - (\omega - n\Omega_0) - i\epsilon}} \tag{3.35}
\]
In this analysis, phonon sidebands are centered at frequencies \(\omega_0 + n\Omega_0\) and have widths determined by the parameter \(\tilde{\Delta}_k^2 = |\langle \tilde{C}_+ \rangle|^2|\lambda_k|^2\). Clearly the coupling of the dot to the phonons in mean-field theory reduces the width of all spectral features by the Franck-Condon factor \(\langle \tilde{C}_+ \rangle \langle \tilde{C}_- \rangle = e^{-\Phi(0)}\) (see Eq(3.12)) when compared to the dot with no phonon coupling. The excited quantum dot is in a displaced lattice state compared to the phonon state when it is de-excited. The displaced states arise from a coherent superposition of phonons that surround the exciton and give rise to the polaron shift in its energy [144]. The displacement of the many-body wavefunction grows with exciton-phonon coupling, \(g\), and the number of thermally excited phonons. The greater the displacement of the phonon-dressed exciton, the weaker the effective exciton dipole transition rate.

This modification of the radiative decay rate by phonon dressing of the excited quantum dot is a distinguishing feature of our quantum mechanical mean-field description, absent in other semiclassical treatments[50]. As we show in section IV.(C), the physical linewidth of the phonon sidebands is also heavily influenced by the lifetime of the phonons in the polaronic cloud. The damping of phonons (arising for instance from the breakup of phonons into lower energy phonons) limits the influence of the Franck-Condon effect and provides a more dominant channel for energy dissipation from the quantum dot excited state.

In the absence of phonon damping effects, it is easy to verify that the susceptibility Eq(3.35) satisfies the spectral sum rule:
\[
\int_{-\infty}^{\infty} d\omega \chi^n_a(\omega) = \frac{\pi N_d}{\epsilon_0 \hbar} |D_{ge}|^2 \tag{3.36}
\]
providing that all of the sideband energies \(\omega_0 + n\Omega_0\) coincide with some radiative mode of frequency \(\omega_k\) (outside of any gap in the electromagnetic spectrum). In this case the self-energies can be evaluated in the pole approximation \(\omega = \omega_0[70]\). The spectral sum rule then follows from the fact that \(\sum_n L_n = 1\).
3.3 Polarization autocorrelation and Excited State Lifetime

In this section we derive general expressions for the polarization autocorrelation function and the lifetime of the excited state of a two-level system. In the absence of dot-photon coupling, $\hat{\sigma}_{ee}$ commutes with the rest of the Hamiltonian and there is no broadening of the zero-phonon line. The role of phonons is to induce pure dephasing without relaxation. In the presence of dot-photon coupling there is line broadening due to radiative decay and the total rate of dephasing is due to both photon and phonon contributions. A simple two-level atom coupled to a free space electromagnetic vacuum has a purely exponential decay. However, a two-level atom placed near a photonic bandgap, exhibits non-Markovian radiative dynamics[11].

The polarization is defined as the temporal dipolar autocorrelation function:

$$P(t) = -i\Theta(t)\langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(0) \rangle_H.$$  (3.37)

It describes the overlap between the initial system state (say the atom fully excited) and the state at a later time $t$. This captures the effects of dephasing due to the coupling of the two-level atom to the electromagnetic modes and the phonon reservoir. Using the mean-field decomposition outlined previously we rewrite Eq(3.37) as

$$P_{MF}(t) \simeq -i\Theta(t)\langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(0) \rangle_{H_{MF}} e^{-(\Phi(0)-\Phi(t))}.$$  (3.38)

Since the polarization is defined only for $t > 0$, we make use of the inversion formulae Eq(3.14) relating the correlation function to the corresponding thermodynamic Green’s function. The discussion following Eq(3.14) is pertinent here and using Eq(3.15) we have $\langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(0) \rangle_{H_{MF}} \simeq iG_{t}^{MF}(\hat{\sigma}_{ge},\hat{\sigma}_{eg})$ for $\beta \omega_0 \gg 1$. Physically this follows from the fact that $\langle \hat{\sigma}_{eg}(0)\hat{\sigma}_{ge}(t) \rangle_{H_{MF}} \simeq 0$ for $\beta \omega_0 \gg 1$ since this term has a finite contribution only for the excited state. In thermal equilibrium and temperatures low compared to the quantum dot optical transition energy, the atom is primarily in the ground state. Therefore we have $P_{MF}(t) = \Theta(t)G_{t}^{MF}(\hat{\sigma}_{ge},\hat{\sigma}_{eg})e^{-(\Phi(0)-\Phi(t))}$. $G_{t}^{MF}(\hat{\sigma}_{ge},\hat{\sigma}_{eg})$ can be evaluated by extending $G_{\omega}^{MF}(\hat{\sigma}_{ge},\hat{\sigma}_{eg})$ on the whole complex plane and using the inverse-Laplace transform:

$$G_{t}^{MF}(\hat{\sigma}_{ge},\hat{\sigma}_{eg}) = \frac{1}{2\pi i} \int_{\epsilon - i\infty}^{\epsilon + i\infty} \frac{1/i}{s + i(\omega_0 - \Delta) + \Sigma(s)} ds.$$  (3.39)
where $\Sigma(s)$ is the self-energy function

$$\Sigma(s) = e^{-\Phi(0)} \sum_k \frac{|\lambda_k|^2}{s + i\omega_k}. \quad (3.40)$$

and $\epsilon$ is chosen such that all the poles lie to the left of the contour of integration. We evaluate the self-energy function for specific photon density of states in later sections and compare their temporal polarization autocorrelation functions.

Until now we have focused solely on the equilibrium dynamics of the two-level atom interacting with a photon and a phonon reservoir. We now consider an important non-equilibrium problem of population dynamics of the two-level atom. The atom, at $t = 0$, is assumed to be in the excited state and hence its density matrix ceases to be of the canonical form ($\exp(-\beta H)/Z$). This invalidates the use of thermodynamic Green's function developed in the previous sections. The decay dynamics of the two-level atom can be obtained by defining a formal non-equilibrium correlation function $L(\hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t')) = \langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle$ for a general density matrix, not necessarily of the canonical form, and considering the equal-time limit $t \to t'$ (see Appendix B). The steady state (non-equilibrium) population is given by the limit $t, t' \to \infty$ of $L(\hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t'))$.

In this section we derive an expression for the lifetime of the excited state by studying the temporal evolution of $\langle \hat{\sigma}_{ee}(t) \rangle$ using the Heisenberg’s equation of motion. In Appendix B we rederive the same result using the method of generalized Laplace-transform following the work of Pike and Swain[142]. The results obtained using the two methods are the same under an equivalent set of approximations. However the method of generalized Laplace-transform is formally elegant and substantially easier to apply to more diverse models involving multiple and multilevel atoms. The method of generalized-Laplace transforms is an extension of the thermodynamic Green’s functions to nonequilibrium problems and shares mathematical similarities with its equilibrium version. Furthermore this method provides insight into the approximations made to incorporate phonon effects on the lifetime of the two-level atom using the Heisenberg equations of motion.

We now proceed with the Heisenberg equations of motion. Since $\hat{\sigma}_{ee}$ commutes with $\exp(-S)$, the equation of motion for the operator $\hat{\sigma}_{ee}$ can be written using either Hamiltonian Eq(2.33) or the polaron-transformed Hamiltonian Eq(2.36). An approximate solution is obtained by solving the problem in the Born-approximation in which the equation of motion for $\hat{\sigma}_{ee}(t)$ is considered to only second-order in the dot-photon coupling. The dot-phonon coupling, on the other hand, is treated exactly to all orders and is subse-
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Subsequently approximated by thermal averages of the phonon displacement operators. The equation of motion for $\hat{\sigma}_{ee}(t)$ is given by

$$\frac{\partial \hat{\sigma}_{ee}(t)}{\partial t} = i[\hat{H}, \hat{\sigma}_{ee}].$$

(3.41)

Using Eq(2.36) this becomes

$$\frac{\partial \hat{\sigma}_{ee}(t)}{\partial t} = -i \sum_k \lambda_k \hat{C}_+ \hat{\sigma}_{eg} \hat{a}_k + i \sum_k \lambda_k^* \hat{a}_k^\dagger \hat{C}_- \hat{\sigma}_{ge}$$

(3.42)

The equation of motion for $\hat{a}_k$ is

$$\frac{\partial \hat{a}_k(t)}{\partial t} = -i \omega_k \hat{a}_k - i \lambda_k^* \hat{C}_- \hat{\sigma}_{ge}$$

Integrating Eq(3.43) we have

$$\hat{a}_k(t) = \hat{a}_k(0)e^{-i\omega_k t} - i \lambda_k^* \int_0^t dt' \hat{C}_-(t') \hat{\sigma}_{ge}(t') e^{-i\omega_k (t-t')}$$

Substituting Eq(3.43) in Eq(3.42) we obtain

$$\frac{\partial \hat{\sigma}_{ee}(t)}{\partial t} = -i \sum_k |\lambda_k|^2 \int_0^t dt' \hat{C}_+(t') \hat{\sigma}_{eg}(t') \hat{C}_-(t') \hat{\sigma}_{ge}(t') e^{-i\omega_k (t-t')} + h.c.$$  (3.43)

$$+ \hat{N}oise(t)$$

where

$$\hat{N}oise(t) = -i \sum_k \lambda_k e^{-i\omega_k t} \hat{C}_+(t) \hat{\sigma}_{eg}(t) \hat{a}_k(0) + h.c.$$  

and $h.c$ is the Hermitian conjugate. We now evaluate the quantum expectation value of this operator equation to calculate $\langle \hat{\sigma}_{ee}(t) \rangle = Tr(\rho_A \otimes \rho_{phonon} \otimes \{0\} \{\hat{\sigma}_{ee}(t)\} \{0\})$ where the atom is described by an arbitrary density matrix $\rho_A$, $\rho_{phonon} = \frac{e^{-\beta H_p}}{Tr(e^{-\beta H_p})}$ and $\{0\}$ represents the photon vacuum. The phonon bath is assumed to be in thermal equilibrium at a temperature $\beta$.

We now need to evaluate $\langle \hat{C}_+(t) \hat{\sigma}_{eg}(t) \hat{\sigma}_{ge}(t') \rangle$. In what follows, we perform the mean-field factorization $\langle \hat{C}_+(t) \hat{\sigma}_{eg}(t) \hat{C}_-(t') \hat{\sigma}_{ge}(t') \rangle \simeq \langle \hat{C}_+(t) \hat{C}_-(t') \rangle \langle \hat{\sigma}_{eg}(t) \hat{\sigma}_{ge}(t') \rangle$ and we provide a formal justification for relating the atomic polarization autocorrelation function
to the atomic population according to the relation

\[ \langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle \simeq e^{i\omega'_0(t-t')}\langle \hat{\sigma}_{eg}(t')\hat{\sigma}_{ge}(t') \rangle = e^{i\omega'_0(t-t')}\langle \hat{\sigma}_{ee}(t') \rangle \]

where \( \omega'_0 = \omega_0 - \Delta \) is the dressed atomic transition frequency.

We introduce the Liouvillian \( \mathcal{L} = \mathcal{L}_0 + \mathcal{L}_p + \mathcal{L}_I \) where \( \mathcal{L}_0\hat{\mathcal{O}}(t) = [\hat{\mathcal{O}}(t), \hat{H}_0], \mathcal{L}_p\hat{\mathcal{O}}(t) = [\hat{\mathcal{O}}(t), \hat{H}_p] \) and \( \mathcal{L}_I\hat{\mathcal{O}}(t) = [\hat{\mathcal{O}}(t), H_I] \) for any general operator \( \hat{\mathcal{O}}(t) \) with \( \hat{H}_0 = (\omega_0 - \Delta)\hat{\sigma}_{ee} + \sum_k \omega_k \hat{a}_k^\dagger \hat{a}_k, \quad \hat{H}_I = \sum_k (\lambda_k \hat{C}_+ \hat{\sigma}_{eg} \hat{a}_k + \lambda_k^* \hat{a}_k^\dagger \hat{C}_- \hat{\sigma}_{ge}) \) and \( \hat{H}_p = \sum_q \Omega_q \hat{c}_q^\dagger \hat{c}_q \). In terms of the Liouvillian \( \hat{\sigma}_{eg}(t) = e^{-i(\mathcal{L}_0+\mathcal{L}_p+\mathcal{L}_I)(t-t')}\hat{\sigma}_{eg}(t') \). Since Eq(3.43) is already second order in \( \lambda_k \), Born approximation implies that

\[ \hat{\sigma}_{eg}(t) \simeq e^{-i\mathcal{L}_p(t-t')} e^{-i\mathcal{L}_0(t-t')} \hat{\sigma}_{eg}(t') \]

(344)

\[ \approx e^{-i\mathcal{L}_0(t-t')} \hat{\sigma}_{eg}(t') \]

where we have used \( \mathcal{L}_p\hat{\sigma}_{eg} = 0 \) Evaluating Eq(344), it is easy to see that \( \hat{\sigma}_{eg}(t) \simeq e^{i(\omega_0-\Delta)(t-t')}\hat{\sigma}_{eg}(t') \). Using the Born approximation, defined above, the right hand side of Eq(3.43) can be simplified to:

\[ \frac{\partial \langle \hat{\sigma}_{ee}(t) \rangle}{\partial t} = -\left( \sum_k |\lambda_k|^2 \right) \int_0^t dt' \langle \hat{C}_+(t)\hat{C}_-(t') \rangle \langle \hat{\sigma}_{eg}(t')\hat{\sigma}_{ge}(t') \rangle e^{i(\omega'_0-\omega_k)(t-t')} + h.c. \]

(3.45)

\[ + N\hat{\text{Noise}}(t) \]

where \( \langle \hat{C}_+(t)\hat{C}_-(t') \rangle = Tr(\rho_{\text{phonon}} \hat{C}_+(t)\hat{C}_-(t')) \), \( \langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t) \rangle = Tr(\rho_A \{ \{ 0 \} | \hat{\sigma}_{ee}(t) \{ 0 \} \}) \), \( \omega'_0 = \omega_0 - \Delta \) and

\[ N\hat{\text{Noise}}(t) = -i \sum_k \lambda_k e^{-i\omega_k t} \langle \hat{C}_+(t) \rangle \langle \hat{\sigma}_{eg}(t) \rangle \langle \hat{a}_k(0) \rangle + h.c. \]

Using the thermal averages evaluated over the phonon reservoir \( \langle \hat{C}_+(t)\hat{C}_-(t') \rangle \) given by Eq(3.10) and the fact that the electromagnetic vacuum expectation values \( \langle \hat{a}_k(0) \rangle = \langle \hat{a}_k^\dagger(0) \rangle = 0 \), we rewrite Eq(3.45) as

\[ \frac{\partial \ell(t)}{\partial t} = -\left( \sum_k |\lambda_k|^2 \right) \int_0^t dt' e^{-(\Phi(0)-\Phi(t-t'))} \ell(t') e^{i(\omega'_0-\omega_k)(t-t')} + c.c. \]

(3.46)

where \( \ell(t) = \langle \hat{\sigma}_{ee}(t) \rangle \) and c.c. denotes complex conjugate. As an illustration, we specialize to a bath of optical phonons. Using Eq(3.13) and the expansion \( e^{-(\Phi(0)-\Phi(t-t'))} = \)
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\[ \sum_n L_n e^{-i\Omega_0 (t-t')} \text{ we obtain:} \]

\[ \frac{\partial \ell(t)}{\partial t} = - \sum_n L_n \sum_k |\lambda_k|^2 \int_0^t dt' e^{-i\Omega_0 (t-t')} \ell(t') e^{i(\omega_{n}-\omega_k)(t-t')} + c.c. \quad (3.47) \]

We solve this integro-differential equation for different photonic reservoirs in later chapters.

### 3.4 Discussion

In this chapter we presented a formalism to describe the role of phonons in modifying the optical properties of two level systems. Using the method of thermodynamic Green’s function we calculated the absorption spectrum in the Born approximation without making the Markov approximation. The Green’s functions were evaluated using a mean-field decomposition of the radiative and lattice degrees of freedom. The atomic state was found to be dressed with various number of phonons and the optical absorption spectrum consisted of transitions between the atomic ground state and various dressed excited states (phonon sidebands). The mean-field theory involved replacing the lattice displacement operator by its equilibrium thermal average over the phonon reservoir. This resulted in an effective temperature-dependent radiative coupling constant describing the (Franck-Condon) overlap between the displaced excited state and the ground state. The effective radiative coupling was found to decrease with increasing temperature and quantum dot-phonon coupling producing a subnatural linewidth for the various sidebands.

We then presented a general expression for the polarization dynamics of the quantum dot by computing the temporal dipolar autocorrelation function. The autocorrelation function was evaluated using the mean-field factorization described above. We then studied the population dynamics of the excited quantum dot using the Heisenberg equations of motion for the relevant atomic operators. The hierarchy of equations of motion was closed using a Born approximation that collapses the two-time nonequilibrium atomic dipole correlation function to the atomic population at single time. In Appendix B we present an alternate treatment of the non-equilibrium population dynamics using a generalization of the the thermodynamic Green’s functions to a more general non-equilibrium description in terms of generalized-Laplace transforms. The method of generalized-Laplace transforms allows us to produce results for nonequilibrium using an
equivalent set of approximations as used in the case of equilibrium description.

In the next chapter we evaluate the general expressions for the optical susceptibility, polarization and population in specific electromagnetic reservoirs. We first consider the unstructured electromagnetic reservoir of free space. We then consider the structured electromagnetic reservoir of a photonic crystal which is modelled using an approximate isotropic dispersion in the vicinity of the photonic band edge. We discuss the role of phonons using a dispersionless Einstein model where all the phonons are assumed to have identical frequencies. Later on in this thesis we consider acoustic phonon models. We also include additional processes such as phonon-assisted electronic decay and finite phonon lifetimes which introduce qualitative changes to the general results presented in this chapter.
Chapter 4

Optical susceptibility, polarization and population dynamics for different electromagnetic reservoirs

In the previous chapter we derived a formalism to calculate the linear susceptibility, polarization and population dynamics of the quantum dot in a general electromagnetic reservoir and coupled to a thermal reservoir of phonons. In this chapter we evaluate these general expression to obtain explicit expressions for the optical lineshape, polarization and population dynamics of a two-level atom in the unstructured reservoir of free space and the structured reservoir of a photonic crystal.

4.1 Free space

In this section we calculate the absorption spectrum and study the polarization and population dynamics of a two-level atom coupled to photons and phonons for a smooth featureless photonic density of states. We assume the photonic density of states to be that of free space where the Wigner-Weisskopf approximation is valid[70]. This provides a valuable diagnostic for our formalism. In the absence of phonon coupling, we recapture the standard two-level atom susceptibility in ordinary vacuum. In the presence of phonon coupling, we recapture the semiclassical picture of multiple-phonon-assisted absorption and emission of light[50]. This provides a clear physical picture of the content of our formalism and sets the stage for describing non-Markovian radiative dynamics of a quantum dot in the "colored" electromagnetic vacuum of a PBG material with phonon-mediated
radiative relaxation processes.

4.1.1 Vanishing coupling to phonons

We first consider the limiting case of vanishing dot-phonon coupling. In this limit we expect the standard Lorentzian lineshape to emerge from our theory. In our theory this corresponds to taking the limit $g \to 0$. From the expression for the absorption spectrum Eq(3.35), the only contribution is from the $n = 0$ term. Clearly $L_n \to 0 (n \neq 0)$ and $e^{-\Phi(0)} \to 1$ (see discussion below Eq(3.13)) as $g \to 0$. The absorption spectrum reduces to

$$
\chi''_a(\omega) = \frac{N_d}{\epsilon_0 \hbar} |D_{ge}|^2 \times \operatorname{Im} \lim_{\epsilon \to 0} \frac{1}{\omega_0 - \Delta - \omega - \sum_k \frac{|\lambda_k|^2}{\omega_k - \omega_0 - i\epsilon}}
$$

(4.1)

In the standard Wigner-Weisskopf theory[70], the radiative self-energy correction to the excited state energy is evaluated in the pole approximation $\omega = \omega_0$ and:

$$
\sum_k \frac{|\lambda_k|^2}{\omega_k - \omega_0 - i\epsilon} = \Delta_0(\omega_0) + \frac{1}{2}i\Gamma_0(\omega_0)
$$

(4.2)

where $\Delta_0$ is the Lamb-shift, $\Gamma_0(\omega_0)$ is the rate of spontaneous emission and $\Gamma_0(\omega_0)/2$ is the rate of dipolar dephasing. In order to make contact with the physical decay rates, we re-interpret the summation over $\vec{k}$ to be an integral over wavevectors and a sum over two equivalent polarization channels. Using the coupling Eq(1.7) we obtain $\Delta_0(\omega_0) = \frac{\omega_0 |D_{ge}|^2}{3\epsilon_0 \omega_0 \hbar c}$. This well-known result provides a first consistency check of our general formalism. The formally divergent Lamb shift is reinterpreted by the excitonic mass renormalization and is implicitly included in the observable transition frequency $\omega_0[70]$. 

4.1.2 Finite coupling to phonon bath

We now consider the optical susceptibility Eq(3.35) when the dot-phonon coupling is finite. In our mean-field theory, using the Wigner-Weisskopf approximation as before, and including two equivalent polarization channels in the wavevector summation and Eq(3.12), the $n$ – phonon sideband self-energy becomes:

$$
\sum_k \frac{|\tilde{\lambda}_k|^2}{\omega_k - (\omega_0 - n\Omega_0) - i\epsilon} = e^{-\Phi(0)}(\Delta_0(\omega_0 - n\Omega_0) + \frac{1}{2}i\Gamma_0(\omega_0 - n\Omega_0))
$$

(4.3)
where $\Gamma_0(\omega_0 - n\Omega_0) = \frac{(\omega_0 - n\Omega_0)^3|D_{ge}|^2}{3\pi\epsilon_0\hbar}$ and the factor $e^{-\Phi(0)}$ is obtained from Eq(3.12). Clearly the broadening of the absorption lineshape function for a given sideband $n$ is determined by the spontaneous emission rate at $\omega_0 - n\Omega_0$. As before, we include the Lamb shift into the observable transition frequency. The absorption lineshape Eq(3.34) is now given by

$$\chi_a(\omega) = \frac{N_d}{\epsilon_0\hbar} |D_{ge}|^2 \times \left[ \sum_n L_n \frac{1}{\omega_0 - \Delta - (\omega - n\Omega_0) - e^{-\Phi(0)} \frac{\Gamma_0(\omega_0 - n\Omega_0)}{2}} \right]$$

(4.4)

where $L_n = e^{-g(2N+1)} e^{n\Omega_0\beta/2} I_n(2g\sqrt{N(N+1)})$. The sideband structure due to phonon-assisted processes is clearly evident from Eq(4.4). The $n^{th}$ sideband is centered at frequency $\omega_0 - \Delta + n\Omega_0$ with a spectral weight $L_n$, the width of which is given by $e^{-\Phi(0)} \frac{\Gamma_0(\omega_0 - n\Omega_0)}{2}$. The contribution of higher phonon processes is progressively smaller due to correspondingly smaller weights attached to these processes.

In Fig(4.1) and Fig(4.2) we plot the absorption spectra of a quantum dot in an unstructured electromagnetic reservoir for various temperatures and dot-phonon coupling strength. We assume a quantum dot transition frequency of $1eV$ and an optical phonon of frequency $0.01eV$. The spectral weight in the sidebands also increases with the dot-phonon coupling strength and can have substantial spectral weight even at low temperatures. At high temperatures and strong dot-phonon coupling strengths a substantial fraction of the spectral weight is contained in the sidebands.

### 4.1.3 Population and polarization decay of the excited state in free-space

We now calculate the time evolution of the polarization of an initially excited state in free space. The polarization of the excited state at any time $t$ is given by $P_{MF}(t) = \Theta(t) G_{\text{MF}}^t(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) e^{-\Phi(0) - \Phi(t)}$ where $G_{\text{MF}}^t(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$ is given by Eq(3.39). The self energy function $\Sigma(s)$ is given by Eq(3.40) and in the (Markovian) Wigner-Weisskopf approximation reduces to

$$\Sigma(s) = \frac{1}{2} \Gamma_0(\omega_0) - i\Delta_0(\omega_0)$$

(4.5)

where $\Gamma_0(\omega_0) = e^{-\Phi(0)} \frac{\omega_0^2|D_{ge}|^2}{3\pi\epsilon_0\hbar}$ and $\Delta_0(\omega_0)$ is the Lamb shift which is absorbed in $\omega_0$. Inserting these relations into Eq(3.39) and performing the integrations, we obtain $G_{\text{MF}}^t(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) = -ie^{-(\omega_0-\Delta)t - \frac{1}{2} \Gamma_0(\omega_0)t}$. For a bath of optical phonons, we expand $e^{-\Phi(0) - \Phi(t)}$ in a power
series in $e^{in\Omega_0 t}$. Using Eq(3.38), the absolute value of polarization can be written as

$$|P_{MF}(t)| = \left| \sum_n L_n e^{i(\omega_0 - \Delta + n\Omega_0)t - \frac{1}{2} \Gamma_0(\omega_0)t} \right|$$

This provides a measure of the residual coherence at any time $t$. The presence of phonon coupling introduces an infinite series of dressed states at frequencies $\omega_0 - \Delta + n\Omega_0$. The dressed state at frequency $\omega_0 - \Delta + n\Omega_0$ is weighted by a factor $L_n$. Interference between these dressed states determines the polarization dynamics at short times (on the scale of inverse phonon oscillator frequencies). At low temperatures, high $\beta$, and low $g$, the spectral weight is concentrated in a few dressed states about $n = 0$ as compared to high temperatures and strong dot-phonon coupling where the spectral weight is spread over a large number of dressed states. Interference between these dressed states gives rise to oscillatory behaviour (on the $\text{ps}$ timescale) in the polarization decay. If the dot-phonon coupling weak, the majority of the spectral weight resides in the zero-phonon line at frequency $\omega_0 - \Delta$ and the oscillatory behaviour is less pronounced. In the absence of phonon damping (sub $\text{ns}$ phonon lifetime) the longer time($\text{ns}$) polarization dynamics is determined by radiative dephasing. In our mean-field approximation, phonon dephasing is manifest in the reduced and temperature-dependent optical dipole-moment. In the absence of phonon damping, these dephasing effects lead to a longer lifetime of the excited quantum dot. In section VI we consider the dephasing of polarization due to acoustic phonons where the above discussion is extended further. The continuum of energies in the case of acoustic phonons lead to pronounced pure dephasing as compared to the case of optical phonons where polarization oscillations are seen at inverse phonon frequency timescales.

We now study the time evolution of the population of an initially excited state. Using Wigner-Weisskopf theory it is easy to see that:

$$\sum_k |\lambda_k|^2 \int_0^t dt' \ell(t') \left( e^{i(\omega_0 - \omega_k - n\Omega_0)(t-t')} + e^{-i(\omega_0 - \omega_k - n\Omega_0)(t-t')} \right) = \Gamma_0(\omega_0 - n\Omega_0) \ell(t)$$

where $\Gamma_0(\omega_0 - n\Omega_0) = \frac{(\omega_0 - n\Omega_0)^3 |D_{\text{exc}}|^2}{3\pi\epsilon_0\hbar c^3}$. Using Eq(4.7) in Eq(3.47) we find

$$\frac{\partial \ell(t)}{\partial t} = -\sum_n L_n \Gamma_0(\omega_0 - n\Omega_0) \ell(t)$$
Integrating Eq(4.8) we have

\[ \ell(t) = e^{-\sum_n L_n \Gamma_0(\omega_0 - n \Omega_0)t} \] (4.9)

It is easy to see from this expression the contribution of the different sidebands to the decay of the excited state. In the absence of phonons we obtain the usual result

\[ \ell(t) = e^{-\Gamma_0(\omega_0)t} \] (4.10)

Also, the polarization has a simple exponential form

\[ P(t) = -i\Theta(t)e^{-i\omega_0t - \frac{\Gamma_0(\omega_0)t}{2}} \] (4.11)

where \( \Gamma_0(\omega_0) = \frac{\omega_0^2 |D_{\text{ph}}|^2}{4\epsilon_0 \hbar c} \). By comparing with Eq(4.10) we recapture the standard result that (in the absence of phonon coupling) the dephasing rate is half the relaxation rate in free space.
Figure 4.1: Plots of absorption spectra of the quantum dot in an unstructured electromagnetic reservoir Eq(4.4) for the dot-phonon coupling strength $g = \sum \frac{\Delta^2}{\Omega_p^2} = 0.5$ and two different temperatures of the optical phonon bath $k_B T = 0.001\text{eV}$ and $k_B T = 0.025\text{eV}$. The absorption spectra of the various phonons sidebands are separated by optical phonon frequencies with a width determined by the electromagnetic density states at that frequency. For comparison we have squeezed the spacing between the sidebands and shifted them close to each other and we have scaled the frequency as $\Delta = \omega / \Gamma$. The sideband with index $n$ has a width $\Gamma = e^{-\phi(0)} \frac{\Gamma_0(\omega - \phi(0))}{2}$ where $\Gamma_0(\omega_0) = \frac{\omega_0^3 |D_{p}^2|}{3\pi\epsilon_0 \hbar c^3}$. The quantum dot transition is assumed to be of the order of $1\text{eV}$. The optical phonon frequency is assumed to be of the order of $0.01\text{eV}$. Temperature is expressed in units of $\text{eV}$. In (a) the thermal phonon reservoir is at a temperature of $k_B T = 0.001\text{eV}$. In (b) the phonon reservoir is at a temperature of $k_B T = 0.025\text{eV}$. At low temperatures and at low dot-phonon coupling strengths, the maximum spectral weight is contained in the $n = 0$ zero-phonon line and a few adjacent sidebands (also see Fig(4.2)).
Figure 4.2: Plots of absorption spectra of the quantum dot in an unstructured electromagnetic reservoir Eq(4.4) for $g = \sum \frac{n^2}{n^2} = 1$ and two different temperatures of the optical phonon bath. The absorption spectra of the various phonons sidebands are separated by optical phonon frequencies with a width determined by the electromagnetic density states at that frequency. For comparison we have squeezed the spacing between the sidebands and shifted them close to each other and we have scaled the frequency as $\omega/\Gamma$. The sideband with index $n$ has a width $\Gamma = e^{-\Phi(0)} \frac{\Gamma_0(\omega_0-n\Omega_0)}{2}$ where $\Gamma_0(\omega_0) = \frac{\omega_0^3|D_0|^2}{3\pi\epsilon_0\hbar^2}$. The quantum dot transition is assumed to be of the order of 1 eV. The optical phonon frequency is assumed to be of the order of 0.01 eV. Temperature is expressed in units of eV. In (a) the thermal phonon reservoir is at a temperature of $k_B T = 0.001 eV$. In (b) the phonon reservoir is at a temperature of $k_B T = 0.025 eV$. At these strong dot-phonon coupling strengths, significant dephasing due to phonons is to be expected. At very high temperatures and $g$'s, the spectral weight shifts to the sidebands (compare Fig(4.1)).
4.2 Optical susceptibility in a photonic crystal

In this section we calculate the optical susceptibility for a two-level atom inside a photonic crystal coupled to a phonon bath. Since the density of states changes rapidly in the vicinity of the photonic band edge, the Wigner-Weisskopf approximation is inadequate. In order to capture the non-Markovian nature of the atom-photon interaction we must evaluate the self-energy more precisely[145, 146].

4.2.1 Absorption spectra

We consider a simplified isotropic dispersion relation for the PBG material, obtained by expanding the photonic dispersion to leading order about the band edge wave vector \( k_0 \). In the effective mass approximation it is given by \( \omega_k = \omega_e + A(k - k_0)^2 \) (see discussion preceding Eq(1.34)). \( \omega_e \) is the band-edge frequency and \( A \) is a constant which depends on the photonic crystal parameters (see Eq(1.35)). For convenience of illustration we choose \( A = \frac{\omega_e}{k_0^2} \). A physical 3D photonic crystal is highly anisotropic and the isotropic dispersion model Eq(1.34) is an oversimplification. On the other hand the dispersion relation Eq(1.34) can be realized in a 3D PBG material with 1D waveguide mode that has a cutoff inside the PBG[147, 148].

For the structured electromagnetic vacuum defined by Eq(1.34), the self-energy term in Eq(3.35) can be easily evaluated[11, 134] to yield the (non-Lorentzian) absorption line:

\[
\chi_a(\omega) = -\frac{N_d}{\epsilon_0 \hbar} |D_{ge}|^2 \sum_n L_n \left( \frac{1}{(\omega - n\Omega_0) - \omega_0 + \Delta + e^{-\Phi(0)}} \right) \sqrt{(\omega - n\Omega_0) - \omega_e} \quad (4.12)
\]

with \( C = \frac{\omega_e^2 |D_{ge}|^2 k_0^3}{12 \pi \epsilon_0 \hbar \omega_e^{3/2}} L_n = e^{-g(2N+1)} e^{n\Omega_0 \beta/2} I_n(2g \sqrt{N(N+1)}) \) and we have absorbed the Lamb shift into the resonant frequency of the quantum dot. In the limit of vanishing coupling to phonons, the expression for susceptibility reduces to

\[
\chi_a(\omega) = -\frac{N_d}{\epsilon_0 \hbar} |D_{ge}|^2 \frac{1}{\omega - \omega_0 + \frac{\omega_e}{\sqrt{\omega - \omega_e}}} \quad (4.13)
\]

Clearly, this susceptibility has no imaginary part for \( \omega < \omega_e \) and there is no absorption inside the PBG. However when the coupling to phonons is included, the atom can absorb at frequencies inside the band gap by phonon-assisted processes. For example, for \( \omega_0 < \omega_e \) and \( \Omega_0 > \omega_e - \omega_0 \), the \( n = 1 \) term describes the absorption of a photon whose
energy is outside the gap assisted by an emission of a phonon into the thermal reservoir. The contribution of higher order phonon processes becomes progressively smaller due to smaller spectral weights attached to them, but the probability of a phonon-mediated process increases with rise in temperature.

In Fig(4.3) and Fig(4.4) we plot the absorption spectra of a quantum dot in the structured reservoir of a photonic crystal. The quantum dot transition frequency is $1\text{eV}$ and the optical phonon frequency is $0.01\text{eV}$. Temperature is measured in units of $\text{eV}^{-1}$. Absorption is seen for frequencies inside the photonic band gap at strong dot-phonon coupling strengths and high temperatures. At low cryogenic temperatures and weak dot-phonon coupling the absorption spectrum is dominated by zero-phonon line.

### 4.2.2 Polarization of the two-level atom in a photonic crystal

In this section we consider the effect of phonons on the polarization (coherence) of the two-level system inside a photonic crystal. For an excited atom whose resonant frequency is near the band edge, the atom is dressed by its own localized radiation field. This leads to Rabi splitting of the excited state into an Autler-Townes doublet\[11\]. If the dressing is strong enough, the lower frequency component of the doublet is pushed inside the photonic band gap and this component is immune from spontaneous radiative decay. This photon-atom bound state results in a non-zero steady state population and finite coherence for large times. In the model Hamiltonian of Eq(2.33) and our subsequent mean-field theory, the role of phonons is to dephase the optical dipole of the quantum dot and to displace the excited state wavefunction from that of the ground state. This leads to diminished polarization and coherence but enhanced excited state population for the photon-atom bound state. In this model, the lifetime of the photon-atom bound state is limited by damping of the phonons themselves due to anharmonic or dissipative processes. The important role of phonon lifetime in modifying the photon-atom bound state is discussed in Section VI(C).

We now study the temporal evolution of the polarization of an initially excited quantum dot coupled to the structured electromagnetic reservoir of the photonic crystal and a bath of thermalized optical phonons. The coherence of the two-level atom coupled to phonons is $|P_{MF}(t)|$ as defined by Eq(3.38). The Green’s function $G_{t}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$ defined by Eq(3.39) has a Laplace-transform $G_{s}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) = \frac{1/\text{i}}{s \pm (\omega_{0} - \Delta) + \sum_{k} \frac{2 \omega_{k}}{\omega_{k}^{2} - s^{2}}}$, It is convenient to perform a shift in the complex-$s$ plane and evaluate the function $G_{s-\text{i}(\omega_{0} - \Delta)}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})$. 
For the isotropic effective mass dispersion relation, the photonic band edge density of
states has a square root singularity. It can be shown that \[11\]:

\[
G_{s-i(\omega_0-\Delta)}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) = \frac{1}{i(s-i\delta)^{1/2}} \frac{(s-i\delta)^{1/2}}{s(s-i\delta)^{1/2} - (i\alpha)^{1/2}}
\]

(4.14)

where

\[
\alpha^2 = \frac{e^{-\Phi(0)\omega_0^2|D_{ge}|^2}}{6\pi\epsilon_0\hbar c^3}
\]

(4.15)

and \(\delta = \omega_0 - \Delta - \omega_e\) is the detuning of the zero phonon line from the band edge.

The function \(G_{t}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg})\) can be computed from the inverse-Laplace transform using Eq(3.39) to yield:

\[
iG_{t}^{MF}(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) = 2a_{1}x_{1}e^{\alpha x_{1}^{2}t-i\omega_{0}t} + a_{2}(x_{2}+y_{2})e^{\alpha x_{2}^{2}t-i\omega_{0}t}
- \sum_{j=1}^{3} a_{j}y_{j}Erfc(\sqrt{\alpha x_{j}^{2}t})e^{\alpha x_{j}^{2}t-i\omega_{0}t}
\]

(4.16)

where

\[
x_{1} = (A_{+} + A_{-}) \exp(i\pi/4)
\]

(4.17a)

\[
x_{2} = (A_{+} \exp(-i\pi/6) - A_{-} \exp(i\pi/6)) \exp(-i\pi/4)
\]

(4.17b)

\[
x_{3} = (A_{+} \exp(i\pi/6) - A_{-} \exp(-i\pi/6)) \exp(i3\pi/4)
\]

(4.17c)

\[
A_{\pm} = \left[\frac{1}{2} \pm \frac{1}{2}[1 + \frac{4}{27} \frac{\delta^{3}}{\alpha^{3}}]^{1/2}\right]
\]

(4.17d)

\[
a_{j} = \frac{x_{j}}{(x_{j} - x_{i})(x_{j} - x_{k})} \quad (j \neq k \neq i; j, i, k = 1, 2, 3)
\]

(4.17e)

\[
y_{j} = \sqrt{x_{j}^{2}} \quad (j = 1, 2, 3)
\]

(4.17f)

and \(Erfc(x)\) is the complementary error function. The polarization can now be obtained
from Eq(3.38). Using Eq(3.11) for \(e^{-(\Phi(0) - \Phi(t))}\) we find:

\[
P_{MF}(t) = -i\Theta(t) \sum_{n} L_{n}e^{-i(n\Omega_0+\omega_{e})t}[2a_{1}x_{1}e^{\alpha x_{1}^{2}t} + a_{2}(x_{2}+y_{2})e^{\alpha x_{2}^{2}t} - \sum_{j=1}^{3} a_{j}y_{j}Erfc(\sqrt{\alpha x_{j}^{2}t})e^{\alpha x_{j}^{2}t}]
\]

(4.18)

In the absence of any coupling to phonons \((L_{0} = 1 \text{ and } L_{n} = 0 \text{ for all } n \neq 0)\) it has
been shown[11] that for \(\omega_0\) near the band edge, strong interaction between the atom and
its localized radiation splits the atomic level into dressed states. These dressed states are formed at frequencies \( \omega_e - \alpha \text{Im}(x_1^2) \) and \( \omega_e - \alpha \text{Im}(x_2^2) \). The first dressed state is pushed inside the gap and this bound state leads to the fractionalized steady-state atomic population in the excited state. However in the presence of phonon coupling, we see using Eq(4.18) that there exists an infinite number of dressed states formed at frequencies \( \omega_e - n\Omega_0 - \alpha \text{Im}(x_1^2) \) and \( \omega_e - n\Omega_0 - \alpha \text{Im}(x_2^2) \). The relative spectral weights of these dressed states are determined by \( L_n \). For low \( \beta \) and large \( g \), the spectral weight is distributed over a large number of phonon sidebands. However for low temperatures (cryogenic temperatures, high \( \beta \)) and \( g << 1 \) the spectral weight is contained in a few sidebands about the zero-phonon line. The short time \( \sim ps \) temporal evolution of polarization of the two-level atom is determined by the interference of these sidebands. However undamped optical phonons do not lead to complete dephasing at long time scales. Moreover the coupling of the two-level system to the phonon bath results in a Franck-Condon shift that reduces the optical dipole-moment resulting in slower electromagnetic dephasing. The ultimate cause of decoherence and decay of the photon-atom bound state occurs from damping (finite lifetime) of the phonons themselves or other non-radiative decay channels (See section VI).

4.2.3 Residual population on the fractionalized steady state

We now calculate the residual population on the fractionalized steady state. We start from Eq(3.47) which we solve using the method of Laplace transforms. The Laplace transform of \( \ell(t) \) is given by:

\[
\hat{\ell}(s) = \frac{\ell(0)}{s + \sum_n L_n \tilde{Z}(s - in\Omega_0) + \sum_n L_n \tilde{Z}^*(s - in\Omega_0)} \tag{4.19}
\]

where

\[
\tilde{Z}(s - in\Omega_0) = \sum_k \frac{|\lambda_k|^2}{s + \ell(\omega_k + n\Omega_0 - \omega_0 + \Delta)} \tag{4.20}
\]

and \( \ell(0) \) is the population at \( t = 0 \). \( \ell(t) \) is then obtained from the inverse-Laplace transform of Eq(4.19). The self energy is easily evaluated\[17, 134\] for the isotropic dispersion relation Eq(1.34):

\[
\tilde{Z}(s - in\Omega_0) = \frac{\alpha^2 \hat{\epsilon} \mp \sqrt{s - i\Delta + in\Omega_0}}{s - i\Delta + in\Omega_0} \tag{4.21}
\]
where $\alpha^2 = \frac{\sqrt{D_0}}{\theta_{\text{opt}}}$ The resulting inverse-Laplace transform cannot be evaluated analytically and the inversion is done numerically. The population on the fractionalized steady state in the absence of any coupling to phonons is given by

$$\tilde{\ell}(s) = \frac{\ell(0)}{s + \tilde{Z}(s) + \tilde{Z}^*(s)}$$

(4.22)

with $\tilde{Z}(s) = \frac{\alpha^2 e^{-\alpha s}}{\sqrt{s+\alpha^2}}$ and $\alpha$ as defined above.

At zero temperature, $L_n = 0$ for $n < 0$ ("hot" sidebands inactive) since there are no phonons available to absorb from the thermal phonon reservoir. Only the cold bands ($n > 0$), involving phonon emission contribute to the spectrum. At finite temperatures all sidebands are active, and the weight of $L_n$'s, for $n < 0$, increases with temperature. With increasing temperatures, there is a marginal shift in the spectral weight to the $n < 0$ sidebands. The dressing of the photon-atom bound state by the phonons leads to (i) a shift of the spectral weight further inside the photonic band gap due to the polaron shift, (ii) a reduction of the effective transition dipole moment due to dephasing and (iii) a diminished overlap between the excited state many-body (atom + phonon cloud) wavefunction from the ground state. In the absence of phonon damping, this leads to the peculiar result that optical phonons suppress radiative decay. In the next section, we consider additional physical effects that need to be included for a realistic picture of quantum dot phonon interactions.
Figure 4.3: Plots of absorption spectra Eq(4.12) of the quantum dot in a photonic crystal. The quantum dot transition is assumed to be 1eV and the optical phonon frequency is 0.01eV. Temperature is chosen 40eV for all the plots. The frequency scale is $\alpha = C \frac{2}{k_B T}$ where $C = \frac{\omega_0^2 D_{2\omega_0}^2 k_B^3}{12 \pi \epsilon_0 \hbar \omega_0^2}$. The sidebands (separated by $\Omega_0$ in frequency) have been shifted for comparison in the same plot. The band edge and the resonant frequency of the atom are shown with respect to the zero-phonon line. The dot-phonon coupling $g = \sum g_{n,n+1}$ is 0.5 in (a) and $g = 1.0$ in (b). The detuning is $\delta = -3\alpha$. The effect of phonon sidebands is clearly seen at room temperatures ($k_B T = 40$eV). Absorption inside the band gap is noticeable due to enhanced phonon-mediated absorption due to strong dot-phonon coupling and an increase in temperature of the phonon bath. Phonon-mediated absorption also increases with the dot-phonon coupling strength (also see Fig(4.4)).
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Figure 4.4: Plots of absorption spectra Eq(4.12) of the quantum dot in a photonic crystal. The quantum dot transition is assumed to be 1eV and the optical phonon frequency is 0.01 eV. Temperature is chosen 40 eV for all the plots. The frequency scale is \( \alpha = C \frac{\omega_0^2 |D_{\omega_0}|^2}{\omega_0^2} \) where \( C = \frac{\omega_0^2 |D_{\omega_0}|^2}{12\pi^2 \hbar} \frac{k_B^2}{\omega_0^2} \). The sidebands (separated by \( \Omega_0 \) in frequency) have been shifted for comparison in the same plot. The band edge and the resonant frequency of the atom are shown with respect to the zero-phonon line. The dot-phonon coupling \( g = \sum \frac{n^2}{n^2} \) is 0.5 in (a) and \( g = 1.0 \) in (b). The detuning is \( \delta = \alpha \). As the dot is detuned away from the band edge, the absorption spectra closely follows the spectrum (Lorentzian) in free space. For low temperatures \( (k_B T \approx 10^{-3} eV) \), we expect no phonon assisted absorption. The spectral weight in the sidebands is also smaller at low temperatures. In the absence of phonon damping, the width of all the peaks is purely radiative in nature (also see Fig(4.3)).
4.3 Discussion

In this chapter we have presented explicit results for the optical lineshape and the polarization and population dynamics of a two-level atom in the unstructured reservoir of free space and the structured reservoir of a photonic crystal. We first considered the linear susceptibility of a two-level atom in a conventional Markovian reservoir of free space and considered the limiting case of no coupling to phonons. The Lorentzian spectrum associated with Markovian dynamics was obtained. We then introduced coupling of the two-level atom to a bath of phonons which was modelled as dispersionless Einstein phonons. The phonon sideband structure emerged in the optical lineshape with sidebands centered at frequencies $\omega_0 + n\Omega_0$ with spectral weights $L_n$. The dressed state at frequency $\omega_0 - \Delta + n\Omega_0$ is weighted by a factor $L_n$. Interference between these dressed states determines the polarization dynamics at short times (on the scale of inverse phonon oscillator frequencies). At low temperatures, high $\beta$, and low $g$, the spectral weight is concentrated in a few dressed states about $n = 0$ as compared to high temperatures and strong dot-phonon coupling where the spectral weight is spread over a large number of dressed states. Interference between these dressed states gives rise to oscillatory behaviour (on the $ps$ timescale) in the polarization decay. If the dot-phonon coupling is weak, the majority of the spectral weight resides in the zero-phonon line at frequency $\omega_0 - \Delta$ and the oscillatory behaviour is less pronounced. In the absence of phonon damping (sub $ns$ phonon lifetime) the longer time($ns$) polarization dynamics is determined by radiative dephasing. In our mean-field approximation, phonon dephasing is manifest in the reduced and temperature-dependent optical dipole-moment. In the absence of phonon damping, these dephasing effects lead to a longer lifetime of the excited quantum dot. The width of the $n^{th}$ sideband was found to be $e^{-\frac{\Phi(0)}{2} + \frac{g^2 n\Omega_0}{2}}$. The mean-field theory employed in the computation of the optical lineshape reduced the width of all spectral features by the Franck-Condon factor $\langle \hat{C}_+ \rangle \langle \hat{C}_- \rangle = e^{-\Phi(0)}$ as compared to the dot with no phonon coupling. This modification of the radiative decay rate by phonon dressing of the excited quantum dot is a a crucial result obtained in our quantum mechanical mean-field description which is absent in other semiclassical treatments. The subnatural physical linewidth of the phonon sidebands can lead to the erroneous conclusion that phonon coupling can suppress radiative decay. This artifact results from the incompleteness of the Spin – Boson model which only includes phonon processes leading to pure dephasing. In other words there is no mechanism for the polaronic cloud to decay in the
absence of radiative processes.

We then studied the optical lineshape of the quantum dot placed inside the structured reservoir of a photonic crystal. In the absence of any coupling to phonons, we found that the susceptibility had no imaginary part for $\omega < \omega_c$ and there was no absorption inside the PBG. When the coupling to phonons was introduced, the atom was found to absorb at frequencies inside the band gap by phonon-assisted processes. The contribution of higher order phonon processes was found to decrease with increasing $n$ due to smaller spectral weights attached to them. However, the probability of a phonon-mediated process was found to increase with temperature. We then studied the polarization and population dynamics of the quantum dot. In the absence of any coupling to phonons and for atomic transition frequency near the photonic band edge, we found strong interaction between the atom and its localized radiation which resulted in the splitting of the atomic level into two dressed states. The first dressed state has a transition frequency inside the gap and forms a bound state. This photon-atom bound state which does not decay can trap the atomic coherence. In the presence of phonon coupling we found an infinite number of dressed states at a discrete set of frequencies separated by the optical phonon frequency. The relative spectral weights of these dressed states was determined by the dot-phonon coupling strength and the temperature of the phonon bath. At high temperatures and strong dot-phonon couplings, the spectral weight is distributed over a large number of phonon sidebands. However for low temperatures (cryogenic temperatures) and weak dot-phonon couplings, the spectral weight is contained in a few sidebands about the zero-phonon line. The short time $\sim ps$ temporal evolution of polarization of the two-level atom is determined by the interference of these sidebands. Undamped optical phonons do not cause dephasing at long time scales which is solely due to electromagnetic dephasing. The coupling of the two-level system to the phonon bath also resulted in a Franck-Condon shift of the excited state which reduced the optical dipole-moment resulting in slower electromagnetic dephasing. The population dynamics of the excited atom was likewise determined by the relative frequencies of the sidebands and the detuning of the atomic transition frequency from the photonic band gap. The dressing of the photon-atom bound state by the phonons was found to shift the spectral weight further inside the photonic band gap due to the polaron shift. In the absence of phonon damping, we obtained a peculiar result that optical phonons suppress radiative decay.

In the next chapter we consider dephasing and population dynamics due to acoustic phonons where the above discussion is extended further. The continuum of energies in
the case of acoustic phonons leads to pronounced pure dephasing as compared to the case of optical phonons where undamped polarization oscillations are seen at inverse phonon frequency timescales. We also introduce additional processes such as phonon assisted electronic transitions and finite lifetimes of phonons (arising for instance from the breakup of phonons into lower energy phonons) which provide a sub-\textit{ns} relaxation channel for the polaronic cloud especially at high temperatures. We also show that the diminished and subnatural linewidth due to the Franck-Condon effect is now modified in the presence of these processes.
Chapter 5

Role of acoustic phonons, nonradiative relaxation and phonon lifetime

In the previous chapter we studied the polarization and population dynamics of a quantum dot using a simple model of dispersionless model of Einstein phonons. In this chapter we study the polarization and population dynamics of the quantum dot with coupling to a bath of undamped acoustic phonons. However undamped optical and acoustic phonons do not lead to relaxation of the polaron cloud associated with the quantum dot. This can be traced back to the nature of dot-phonon coupling in the "Independent – Boson" model. This results in a subnatural linewidth resulting from the dephasing of the optical dipole moment due to phonons. This can be rectified by including nonradiative processes which are not contained in the "Independent – Boson" model. This is accomplished by introducing a purely nonradiative decay channel in our model Hamiltonian. We also consider phonon-phonon scattering and anharmonic phonon processes using a phenomenological model which can also enable complete decay of the photon-atom bound state.

5.1 Undamped acoustic phonons

The optical phonon model used in the previous sections produced well-separated phonon sidebands. Interference between these sidebands gives rise to oscillations in the polarization at optical phonon timescales. However, the polarization of a photon-atom bound
state does not decay to zero at long times. For coupling to a continuum of acoustic phonons, the distinct sidebands likewise merge into a continuum. Quantum interference within this continuum of modes lead to polarization decay to a new steady state without any subsequent revivals.

For acoustic phonons we adopt a model used in earlier literature[52, 91] to describe GaAs-based quantum dots (see Chapter 1.3.2). We assume a spherical dot model with acoustic deformation potential interactions with the dot-phonon coupling given by Eq(1.13). For small phonon wavevectors \( q \) an effective linear dispersion relation \( \omega = c_l q \) is assumed and Eq(3.11) for \( \Phi(t) \) is now given by Eq(1.14). The dimensionless dot-phonon coupling (Huang-Rhys factor) \( \alpha_p \) is defined in Eq(1.15). An analytic expression for \( \Phi(t) \) can be derived by approximating \( \cosh(\frac{\hbar \omega}{2k_B T}) \simeq \frac{2\pi}{\hbar \omega} \) which is certainly valid for \( \frac{\hbar \omega}{2k_B T} \ll 1 \) (\( T \gg 7.8K \)). The integrals are straightforward to perform analytically and we obtain:

\[
\Phi(t) = \alpha_p(t) e^{-\frac{t^2}{2}}
\]  

(5.1)

where \( \alpha_p(t) = \alpha_p^{(1)} + i \alpha_p^{(2)} \) and \( \alpha_p^{(1)} = \sqrt{\frac{2\pi \alpha_p}{\hbar^3}} \) and \( \alpha_p^{(2)} = \sqrt{\frac{\pi}{2}} \alpha_p \). The spectral weight of the zero-phonon line, \( e^{-\Phi(0)} \), is now given by \( e^{-\alpha_p^{(1)}} \).

We now consider the population decay dynamics Eq(3.46) in the presence of an acoustic phonon reservoir. Unlike the case of optical phonons a straightforward expansion in terms of the phonon sidebands cannot be made. Using Eq(5.1) in Eq(3.46) for \( \ell(t) \), we obtain:

\[
\frac{\partial \ell(t)}{\partial t} \simeq - e^{-\Phi(0)} \sum_k |\lambda_k|^2 \int_0^t dt' e^{\alpha_p(t-t')i} e^{-\frac{(t-t')^2}{2}} \ell(t') e^{i(\omega_k - \omega_k)(t-t')} + c.c.
\]  

(5.2)

where \( \alpha_p(t-t') = \alpha_p^{(1)} + i \frac{t-t'}{\tau} \alpha_p^{(2)} \). A straightforward scaling analysis shows that for \( t \ll \alpha^{-1}, \frac{\partial \ell(t)}{\partial t} \simeq 0 \). The RHS of Eq(5.2) is non-zero only for \( t \geq \alpha^{-1} \) when population decay kicks in due to radiative processes which happen on timescales of \( \alpha^{-1} \). This is due to the fact that the role of phonons is confined to pure dephasing (as modelled using the Independent – Boson model for the dot-phonon dynamics in Eq(2.33)) wherein phonons do not cause any population decay (since \( \hat{\sigma}_{ee} \) commutes with the phonon part of the interaction Hamiltonian in Eq(2.33)) but are only associated with polarization decay.

The radiative memory kernel \( \Sigma(t-t') \equiv \sum_k |\lambda_k|^2 e^{i(\omega_k - \omega_k)(t-t')} \) describes non-Markovian effects in the vicinity of a photonic band edge. When the radiative transition is detuned by a small amount, \( \delta \), from a \( 1 - D \) band edge singularity in the electromagnetic den-
sity of states, this memory kernel decays[17] as a power law \( \Sigma(t - t') \simeq \frac{\alpha \Phi}{\sqrt{t - t'}} e^{i \Phi(t - t')} \).

Phonon dephasing effects tend to suppress these memory effects. The more phonons that participate in the decay process, the more rapidly memory is lost. This can be seen by expanding the function \( e^{\Phi(t)} \) in a Taylor series:

\[
\frac{\partial \ell(t)}{\partial t} \simeq - e^{-\Phi(0)} \sum_{n=0}^{\infty} \int_0^t dt' \frac{\alpha n(t-t')}{n!} e^{-n(t-t')^2} \Sigma(t - t') \ell(t') + c.c. \tag{5.3}
\]

Clearly, only the zero-phonon \((n = 0)\) term experiences the power law decay of the radiative memory kernel, whereas processes involving \(n > 0\) phonons lose memory of any light localization effects at rates (defined by the Gaussian factors in Eq(5.3)) that increase with the number of phonons. Nevertheless, in the absence of damping of the phonons themselves, a fractionalized steady state population of the quantum dot is possible as a result of the \(n = 0\) component of the overall memory kernel.

In Fig(5.1) we plot \(|e^{-(\Phi(0)-\Phi(t))}|\) as a function of time \(\frac{t}{\tau}\) for dot-phonon coupling strength \(\alpha_p = 0.033\) (\(\Phi(t)\) defined in Eq(5.1)). Pure dephasing due to undamped acoustic phonons is clearly seen as a rapid loss of coherence at ps time scales. The coherence, however, does not decay to zero and approaches a constant value, implying that dephasing is only partial. The residual coherence decreases with temperature and dot-phonon coupling strength. We also plot the spectral profile which is the imaginary part of the Fourier transform of the correlation function \(e^{-(\Phi(0)-\Phi(t))}\) where \(\Phi(t)\) is given by Eq(5.1)). The sharp unbrodened zero-phonon line is accompanied by a broad sideband continuum. At low temperatures the spectrum is found to be asymmetric about the zero-phonon line. The asymmetry decreases with increasing temperature due to transfer of spectral weight from the ”cold” part of the spectrum to the ”hot” band. However the zero-phonon line is unbroadened even at high temperatures.

In Fig(5.2) and Fig(5.3) we plot mean-field polarization \(|P_{MF}(t)|\) in the structured reservoir of a photonic crystal as a function of time \(t\) (in units of \(\alpha\)) for different temperatures of the undamped acoustic phonon reservoir for an isotropic dispersion relation Eq(1.34). The mean-field polarization \(|P_{MF}(t)|\) is given by:

\[
P_{MF}(t) = -i \Theta(t)e^{-(\Phi(0)-\Phi(t))} e^{-i\omega_t t}[2a_1 x_1 e^{\alpha x_2 t} + a_2 (x_2 + y_2) e^{\alpha y_2 t} - \sum_{j=1}^{3} a_j y_j Er f c(\sqrt{\alpha x_j^2 t}) e^{\alpha x_j^2 t}] \tag{5.4}
\]

where \(\alpha^2 = \frac{e^{-\Phi(0)} e_{\phi}^2 |D_{\phi}|^2}{\hbar \omega_0 \hbar c^3}\). \(x_j, y_j\)‘s and \(a_j\)’s are defined in Eq(4.17) and \(\Phi(t)\) is defined
in Eq(1.14). Note that for $t \simeq \alpha^{-1}$, $e^{-(\Phi(0) - \Phi(t))} \rightarrow e^{-\Phi(0)}$. Pure dephasing is clearly seen at very short time scales, leading to a reduced steady state coherence. The residual coherence of the photon-atom bound state decreases with increasing temperature.

The primary effect of undamped acoustic phonons in our model is a very rapid decay of polarization at \textit{picosecond} timescales followed by radiative decay at long timescale but with a reduction in strength of the effective radiative coupling $|\lambda_k|^2$ due to the Franck-Condon shift of the excited state wave function. In a PBG material the excited-state population in the photon-atom bound state is enhanced. Despite the continuum of acoustic phonon modes, the model Hamiltonian does not allow for decay of the system to its ground state by pure phonon effects. This picture is valid at very low temperatures. In ordinary vacuum this corresponds to the situation where the dynamics of quantum dots and their polarization decay is "radiatively limited"[95, 98]. In a PBG material, where radiative processes can be suspended on time scales much longer than in ordinary vacuum, it is important to consider additional channels for phonon-mediated decay of the photon-atom bound state. This is the subject for the next two subsections.
Figure 5.1: (a) Plot of $|\exp(-(\Phi(0) - \Phi(t)))| (\Phi(t)$ defined in Eq(5.1) for coupling of a quantum dot to undamped acoustic phonons as a function of $t$ for $\alpha_p = 0.033$ ($\alpha_p$ defined in Eq(1.15)) and for temperatures ranging from $T = 15.6K$ (upper curve) to $T = 78K$ (lower curve). The timescale is measured in units of $\tau \equiv L/c_l \simeq 1ps$ where $L$ and $c_l$ are defined in Eq(1.13). We see a rapid decay at early times which approaches a constant but reduced value at later times due to rapid transfer of coherence from the quantum dot to the associated phonon reservoirs. The long time residual value $e^{-\Phi(0)}$ indicates incomplete dephasing of the quantum dot polarization due to undamped phonons in the Spin – Boson model. (b) Spectral profile corresponding to (a). The zero of frequency coincides with the polaron shifted resonant frequency $\omega_0 - \Delta$. The $\delta$-function peak in the middle corresponds to the unbroadened zero-phonon line characteristic of pure dephasing. This $\delta$-function may be broadened by including either radiative or nonradiative decay as well as phonon damping. The remainder of the spectrum is a slightly asymmetric (visible at low temperatures) Gaussian corresponding to the phonon sidebands. The asymmetry in the spectrum is due to the uneven spectral weights for positive and negative frequencies (relative to the zero-phonon line). The spectrum becomes more symmetric with increasing temperatures due to transfer of spectral weight from the "cold" bands to the "hot" bands. The width of the Gaussian sideband spectrum is $D = \sqrt{\alpha_p^2 / \tau}$ and increases with temperature ($\propto \sqrt{T}$).
Figure 5.2: Plots of quantum dot polarization (dipole autocorrelation function) Eq(5.4) for an initially excited quantum dot as a function of time for coupling to an undamped acoustic phonon reservoir with $\alpha_p = 0.033$ in a photonic crystal. Here we consider the case of (a) no quantum dot-phonon coupling and (b) the phonon bath is at a temperature $T = 15.6K$ for various detunings $\delta$ of the quantum dot transition frequency from the photonic band edge. The timescale is measured in units of $\alpha^{-1}$ where $\alpha = 10^{10} s^{-1}$ (See Eq(4.15)). For an initially excited quantum dot, rapid decay at early times ($t \sim ps$) is a signature of phonon dephasing. The rapid drop in polarization from its initial value of unity appears as a nearly discontinuous jump at $t = 0$ in (b) on the timescale depicted. The long time decay rate ($t \sim ns$) is determined by electromagnetic dephasing. The long time residual coherence for detunings near and inside the photonic band edge is due to the fractionalized steady state of the photon-atom bound state. The coherence trapped in the photon atom bound state increases as the atom is detuned further inside the gap. Compare Fig(5.3)
Figure 5.3: Plots of quantum dot polarization (dipole autocorrelation function) Eq(5.4) for an initially excited quantum dot as a function of time for coupling to an undamped acoustic phonon reservoir with $\alpha_p = 0.033$ in a photonic crystal. The undamped phonon reservoir is at a temperature (a) $T = 26K$ and (b) $T = 78K$ for various detunings $\delta$ of the quantum dot transition frequency from the photonic band edge. The timescale is measured in units of $\alpha^{-1}$ where $\alpha = 10^{10} s^{-1}$ (See Eq(4.15)). The residual coherence decreases with temperature due to enhanced phonon dephasing. Also see Fig(5.2)
5.2 Addition of purely nonradiative decay channel

Our model Hamiltonian Eq(2.33) recaptures certain features of multiple-phonon-assisted radiative transitions. In this picture, the quantum dot excited state is dressed by various numbers of phonons, leading to a series of side-bands with energies determined by the number and energy of phonons involved. However, transitions from these dressed states to the quantum dot ground state are purely radiative in nature. The spectral width of these sidebands is determined by the optical density of states at the sideband transition frequency and the modified optical transition matrix element. The optical transition dipole, in this simple model, is diminished due to phonon dephasing effects and the Franck-Condon displacement of the dressed excited states from the ground state. This leads to an artificially small linewidth to the phonon sidebands. In a more realistic picture, we must incorporate phonon-mediated decay mechanisms in addition to the phonon-mediated dressing of the excited state. As we show below, these additional decay channels limit the extent to which the optical dipole transition can be suppressed by the Franck-Condon effect and provide a more realistic picture of the spectral linewidths of the phonon sidebands.

In this section we introduce pure nonradiative decay due to phonons through an additional term in the interaction Hamiltonian:

$$H_{\text{decay}} = \sum_p (\zeta_p \hat{d}_p \sigma_{eg} + \zeta_p^* \hat{d}_p^\dagger \sigma_{ge})$$

(5.5)

where $\hat{d}_p^\dagger (\hat{d}_p)$ is the creation (annihilation) operator for the decay phonon with wavenumber $p$. We assume that these decay phonons form a continuum with smooth and featureless (vibrational) density of states. The operator $\hat{d}_p^\dagger$ could, for instance, correspond to the simultaneous creation of many phonons of the type described earlier with energies $\{\hbar \Omega_k\}$ such that $p = \sum_i k_i$ and $\hbar \omega_0 \approx \sum_i \hbar \Omega_k$. Rather than solving the full Hamiltonian we capture the role of this decay term by introducing a phenomenological decay rate, $\gamma_{\text{nonrad}}$, for the excited state amplitude. In the limiting case of $H_I = 0$ (see Eq(2.33b)), this corresponds to the replacement $\omega_0 \rightarrow \omega_0 - i \gamma_{\text{nonrad}}$ and the decay of the two-level atom correlation function is given by $-i \Theta(t) \langle \hat{\sigma}_{ge}(t) \hat{\sigma}_{eg}(0) \rangle = -i \Theta(t) e^{-i \omega_0 t} \exp(-\gamma_{\text{nonrad}} t)$.

We start by considering the effect of $H_{\text{decay}}$ on the "Spin-Boson part" of the Hamil-
tonian $H$ in Eq(2.33). The modified Spin-Boson Hamiltonian becomes:

$$H_0 = \hbar \omega_0 \hat{\sigma}_{ee} + \sum_q \hbar \Omega_q \hat{c}_q^\dagger \hat{c}_q + \sum_p \hbar \xi_p \hat{d}_p^\dagger \hat{d}_p$$ (5.6a)

$$H_{I}^{\text{decay}} = \hat{\sigma}_{ee} \sum_q \eta_q (\hat{c}_q + \hat{c}_q^\dagger) + H_{\text{decay}}$$ (5.6b)

The correlation function $g(t) = -i \Theta(t) \langle \hat{\sigma}_{ge}(t) \hat{\sigma}_{eg}(0) \rangle$ for the unperturbed Spin-Boson Hamiltonian is well-known[52, 130]. Using the linked-cluster theorem[130] the correlation function can be written as

$$g(t) = g_0(t) e^{W(t)}$$

Here $g_0(t) = -i \Theta(t) e^{-\Delta \omega t}$ is the two-level atom propagator in the absence of dot-phonon interaction and $W(t)$ is given by[130]

$$W(t) = \frac{(-i)^2}{2!} e^{i \omega_0 t} \int_0^t dt_1 \int_0^t dt_2 \langle \hat{\sigma}_{ge}(t) H_{I}^{\text{decay}}(t_1) H_{I}^{\text{decay}}(t_2) \hat{\sigma}_{eg}(0) \rangle$$ (5.7)

and $H_{I}^{\text{decay}}(t)$ is the interacting part of the Hamiltonian in the interaction picture. This simple form for $W(t)$ is due to the fact that there is only one distinct "linked" diagram for the phonons[139] in the linked-cluster expansion. In the absence of decay phonons ($H_{\text{decay}} = 0$), the interaction Hamiltonian is given by:

$$H_{I}(t) = \hat{\sigma}_{eg}(t) \hat{\sigma}_{ge}(t) \hat{V}(t)$$ (5.8a)

$$\hat{V}(t) = \sum_q \eta_q (\hat{c}_q e^{-i \Omega_q t} + \hat{c}_q^\dagger e^{i \Omega_q t})$$ (5.8b)

The term $\langle \hat{\sigma}_{ge}(t) H_{I}(t_1) H_{I}(t_2) \hat{\sigma}_{eg}(0) \rangle$ can now be evaluated using Wick’s theorem[130] as:

$$\langle \hat{\sigma}_{ge}(t) H_{I}(t_1) H_{I}(t_2) \hat{\sigma}_{eg}(0) \rangle = 2 g^0(t - t_1) g^0(t_1 - t_2) g^0(t_2) D(t_1 - t_2)$$ (5.9)

and $D(t_1 - t_2)$ is the phonon correlation function[130] defined as:

$$\langle \hat{V}(t_1) \hat{V}(t_2) \rangle = i D(t_1 - t_2) = \sum_q \eta_q^2 (N_q + 1) e^{-i \Omega_q |t_1 - t_2|} + N_q e^{i \Omega_q |t_1 - t_2|}$$ (5.10)

We now write:

$$W(t) = -e^{i \omega_0 t} \int_0^t dt_1 \int_0^t dt_2 D(t_1 - t_2) g^0(t - t_1) g^0(t_1 - t_2) g^0(t_2)$$ (5.11)

It is now straightforward to show[139] that $g(t) = -i \Theta(t) e^{-i (\omega_0 - \Delta) t} e^{-\Phi(0)}$. This is
the well known result[130] for the Spin-Boson model with \( \Phi(t) \) and \( \Delta \) defined in Eq(3.11) and Eq(2.37) respectively.

We now consider the effect of the decay term, \( H_{\text{decay}} \), on the correlation function of the Spin – Boson Hamiltonian. This decay term interferes with the pure phonon dephasing process. If the timescale of the decay-phonon processes is substantially faster than the pure dephasing phonons, the atom will undergo nonradiative decay even before it can transfer energy and emit or absorb pure dephasing phonons. If the timescales of these two phonon processes are comparable, the spectral structure of the phonon sidebands due to pure dephasing phonons should be modified by the nonradiative decay processes. The contribution of this decay term to the Spin – Boson correlation function can be included in the lowest order by the ansatz[149]:

\[
D(t_1 - t_2) \rightarrow D(t_1 - t_2)e^{-\gamma_{\text{nonrad}}|t_1 - t_2|}
\]  

(5.12)

A microscopic derivation of this replacement follows from the interaction Hamiltonian Eq(5.5). Our ansatz corresponds to the leading correction to the irreducible self-energy of the temporal atomic dipole propagator, arising from additional interaction Eq(5.5). The decay term acts only during the times when the dot interacts with the purely dephasing phonons and leads to the renormalization of the dot-pure dephasing phonon coupling due to direct nonradiative processes. This simultaneous interaction of the nonradiative decay processes and purely dephasing processes results in broadening of the phonon sidebands. A microscopic expression for \( \gamma_{\text{nonrad}} \) can be obtained from \( H_{\text{decay}} \). To the lowest order in the dot-nonradiative decay coupling, we obtain \( \gamma_{\text{nonrad}} = \pi \sum_p (n_p + \frac{1}{2})|\xi_p|^2\delta(\omega - \xi_p) \) where \( n_p = \frac{1}{e^{\xi_p} - 1} \). The excited state probability then decays as \( e^{-2\gamma_{\text{nonrad}}t} \). Assuming a smooth density of states in the vicinity of \( \omega_0 \), the sum can be converted into an integral and evaluated. For \( \beta\xi_p << 1 \), it is easy to see that \( \gamma_{\text{nonrad}} \) is linear in \( T \). Therefore, in this simple model, we find that the non-radiative decay rate increases linearly with temperature (in the high-temperature limit \( \beta\xi_p << 1 \)).

Using the definition of the unperturbed correlation function, the evaluation of \( W(t) \) now reduces to:

\[
W(t) = -\int_0^t dt_1 \int_0^{t_1} dt_2 \sum_q \eta_q^2[(N_q + 1)e^{-\Omega_q - \gamma_{\text{nonrad}}}|t_1 - t_2| + N_q e^{\Omega_q - \gamma_{\text{nonrad}}}|t_1 - t_2|] \quad (5.13)
\]

Straightforward evaluation of the time integrals yields[127] \( W(t) \) in terms of the modified
\( \Phi(t) \) given by:

\[
W(t) = -t \Phi'(0) - (\Phi(0) - \Phi(t))
\]

\[
\Phi(t) = \sum_q \eta_q^2 \left[ \frac{N_q}{(\Omega_q + i\gamma_{\text{nonrad}})^2} e^{(i\Omega_q - \gamma_{\text{nonrad}})t} + \frac{N_q + 1}{(\Omega_q - i\gamma_{\text{nonrad}})^2} e^{(-i\Omega_q - \gamma_{\text{nonrad}})t} \right]
\]

We rewrite \( \Phi(t) = \Phi_1(t) + i\Phi_2(t) \) where:

\[
\Phi_1(t) = \sum_q \frac{\eta_q^2(1 + 2N_q)}{(\Omega_q^2 + \gamma_{\text{nonrad}}^2)^2} [(\Omega_q^2 - \gamma_{\text{nonrad}}^2) \cos(\Omega_q t) + 2\Omega_q \gamma_{\text{nonrad}} \sin(\Omega_q t)] e^{-\gamma_{\text{nonrad}} t}
\]

\[
\Phi_2(t) = \sum_q \frac{\eta_q^2}{(\Omega_q^2 + \gamma_{\text{nonrad}}^2)^2} [2\Omega_q \gamma_{\text{nonrad}} \cos(\Omega_q t) - (\Omega_q^2 - \gamma_{\text{nonrad}}^2) \sin(\Omega_q t)] e^{-\gamma_{\text{nonrad}} t}
\]

Also \( \Phi(0) \) can be decomposed as \( \Phi(0) = \Phi_1(0) + i\Phi_2(0) \) where \( e^{-\Phi_1(0)} \) is the spectral weight of the zero-phonon line[127] and \( \Phi_2(0) \) is a phase. Also \( \Phi'(0) = \frac{d\Phi(t)}{dt} \bigg|_{t=0} = \gamma_{\text{nonrad}} - i\Delta \) where \( \gamma_{\text{nonrad}} \) is an additional decay term contributing to polarization decay due to modified pure dephasing caused by \( H_{\text{decay}} \) and \( \Delta \) is the new polaron shift. They are given by

\[
\gamma_{\text{nonrad}} = \sum_q \frac{\eta_q^2 \gamma_{\text{nonrad}}}{\Omega_q^2 + \gamma_{\text{nonrad}}^2} (1 + 2N_q)
\]

\[
\Delta = \sum_q \frac{\eta_q^2}{\Omega_q^2 + \gamma_{\text{nonrad}}^2}
\]

We now study an analytical and instructive example of the modification of the phonon correlation function due to nonradiative decay \( \gamma_{\text{nonrad}} \) using the acoustic phonon model described in the previous subsection (Eq(1.13)- Eq(1.15)). The modified function \( W(t) \) (see Eq(5.12) and Eq(5.13)) now takes the form:

\[
W(t) = \alpha_p \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^\infty dx \exp\left(-\frac{x^2}{2}\right) \exp(i x \tilde{t}_{12}) - \cos(x \tilde{t}_{12}) \coth\left(\frac{x \beta x}{2\tau}\right) e^{-\tau_{nr} \tilde{t}_{12}}
\]

where \( \tau_{nr} = \gamma_{\text{nonrad}} \tau \) and \( \tilde{t}_{12} = (t_1 - t_2)/\tau \) We now approximate \( \coth\left(\frac{x \beta x}{2\tau}\right) \approx \frac{2\tau}{x \beta x} \) as in
Eq(5.1). Eq(5.17) is then straightforward to compute and we obtain:

\[
W(t) = \alpha_p^{(1)} e^{-\frac{t}{2}(\gamma_{nr} + \tilde{T})} [(1 + \frac{\gamma_{nr}^2}{\gamma_{nr}}) - e^{\frac{t}{2}(2\gamma_{nr} + \tilde{T})}\{(1 + \gamma_{nr}(\tilde{T} + \gamma_{nr}))}\] (5.18)

\[
+ \sqrt{\pi} \gamma_{nr} e^{\frac{1}{2}(\gamma_{nr} + \tilde{T})^2} (2 + \gamma_{nr}^2 + 2\gamma_{nr}\tilde{T}) (Erf\left(\frac{\gamma_{nr} + \tilde{T}}{\sqrt{2}}\right) - Erf\left(\frac{\gamma_{nr}}{\sqrt{2}}\right))
\]

\[
- i\alpha_p^{(2)} e^{-\frac{t}{2}(2\gamma_{nr} + \tilde{T})} [(2\gamma_{nr} + \tilde{T}) - e^{\frac{t}{2}(2\gamma_{nr} + \tilde{T})}(2\gamma_{nr} + \tilde{T}(-1 + \gamma_{nr}^2))]
\]

\[
+ \gamma_{nr}^2 e^{\frac{1}{2}(\gamma_{nr} + \tilde{T})^2} \sqrt{\frac{\pi}{2}} (3 + \gamma_{nr}^2 + 2\gamma_{nr}\tilde{T}) (Erf\left(\frac{\gamma_{nr} + \tilde{T}}{\sqrt{2}}\right) - Erf\left(\frac{\gamma_{nr}}{\sqrt{2}}\right))
\]

The modified phonon correlation function can now be obtained directly from Eq(5.18) and Eq(5.14a) without making any further approximations and has the form:

\[
\tilde{\Phi}_1(t) = \alpha_p^{(1)} e^{-\frac{t}{2} (2\gamma_{nr} + \tilde{T})} [(1 + \gamma_{nr}^2) + \sqrt{\pi} \gamma_{nr} e^{\frac{1}{2}(\gamma_{nr} + \tilde{T})^2} (2 + \gamma_{nr}^2 + 2\gamma_{nr}\tilde{T}) Erf\left(\frac{\gamma_{nr} + \tilde{T}}{\sqrt{2}}\right) - Erf\left(\frac{\gamma_{nr}}{\sqrt{2}}\right)] (5.19a)
\]

\[
\tilde{\Phi}_2(t) = -\alpha_p^{(2)} e^{-\frac{t}{2} (2\gamma_{nr} + \tilde{T})} [(2\gamma_{nr} + \tilde{T}) + \gamma_{nr}^2 e^{\frac{1}{2}(\gamma_{nr} + \tilde{T})^2} \sqrt{\frac{\pi}{2}} (3 + \gamma_{nr}^2 + \gamma_{nr}\tilde{T}) Erf\left(\frac{\gamma_{nr} + \tilde{T}}{\sqrt{2}}\right) - Erf\left(\frac{\gamma_{nr}}{\sqrt{2}}\right)] (5.19b)
\]

The zero-phonon line intensity \(e^{-\tilde{\Phi}_1(0)}\) which is determined by \(\tilde{\Phi}_1(0)\) and the phase \(\tilde{\Phi}_2(0)\) are easily extracted from the above. It is instructive to consider the situation in which direct nonradiative decay is weak perturbation compared to the dot-acoustic phonon coupling \((\gamma_{nr} \equiv \gamma_{\text{nonrad}} \tau \ll 1)\). In this case we make a power series expansion in \(\gamma_{nr}\) to obtain:

\[
\tilde{\Phi}_1(0) = \alpha_p^{(1)} [1 + 3\gamma_{nr}^2 + O(\gamma_{nr}^4)] (5.20a)
\]

\[
\tilde{\Phi}_2(0) = -2\alpha_p^{(2)} [\gamma_{nr} + 2\gamma_{nr}^3 + O(\gamma_{nr}^5)] (5.20b)
\]

Note that the spectral weight of the zero-phonon line \(e^{-\tilde{\Phi}_1(0)}\) decreases with increasing nonradiative decay. The decay term in the modified phonon correlation function, \(\gamma_{\text{nonrad}}\) (which causes exponential decay at long times \(t \gg \tau\)), and the modified polaron shift \(\tilde{\Delta}\) are now obtained using \(\tilde{\Phi}_1(0)\) and \(\tilde{\Phi}_2(0)\) respectively:

\[
\gamma_{\text{nonrad}} = \alpha_p^{(1)} \gamma_{\text{nonrad}}(1 + \sqrt{\pi} \gamma_{nr} e^{\frac{-\gamma_{nr}}{2}\sqrt{2} Erf\left(\frac{\gamma_{nr}}{\sqrt{2}}\right)}) (5.21a)
\]

\[
\tilde{\Delta} = \alpha_p^{(2)} ((1 - \gamma_{nr}^2) - \sqrt{\pi} \gamma_{nr}^2 e^{\frac{-\gamma_{nr}}{2}\sqrt{2} Erf\left(\frac{\gamma_{nr}}{\sqrt{2}}\right)}) (5.21b)
\]
A Taylor expansion of $\tilde{\gamma}_{\text{nonrad}}$ and $\tilde{\Delta}$ lead to:

\begin{align}
\tilde{\gamma}_{\text{nonrad}} &= \alpha_p^{(1)} \tau_{\text{nr}} [1 + \gamma_{\text{nr}}^3 + O(\gamma_{\text{nr}}^5)] \\
\tilde{\Delta} &= \alpha_p^{(2)} [1 - \gamma_{\text{nr}}^2 + O(\gamma_{\text{nr}}^4)]
\end{align}

(5.22a, 5.22b)

Note that $\tilde{\gamma}_{\text{nonrad}} \to 0$ as $\gamma_{\text{nonrad}} \to 0$ as one would expect. Moreover the modified polaron shift $\tilde{\Delta}$ decreases with increasing $\gamma_{\text{nonrad}}$ reflecting the fact that polaronic effects are suppressed by direct nonradiative effects.

The short time phonon dynamics $t \ll \tau$ of $e^{-(\Phi(0)-\Phi(t)+t\Phi'(0))}$ can be obtained by expanding $\tilde{\Phi}(0) - \tilde{\Phi}(t) + t\tilde{\Phi}'(0)$ as a power series in $\tilde{t}$ which is found to be given by:

\begin{align}
\tilde{\Phi}(0) - \tilde{\Phi}(t) + t\tilde{\Phi}'(0) &= \frac{\alpha_p^{(1)}}{2} \tilde{t}^2 + O(\tilde{t}^3)
\end{align}

(5.23)

Neglecting terms $\tilde{t}^3$ and higher, we find that the absorption spectrum which is the Fourier transform of the dipolar autocorrelation function is:

\begin{equation}
\chi''(\omega) = \frac{1}{\sqrt{2\pi}D} e^{-\left(\frac{\omega - \omega_0}{D}\right)^2}
\end{equation}

(5.24)

where $\omega_0$ is the bare quantum dot transition frequency and $D^2 = \frac{\alpha_p^{(1)}}{\tau}$ is the width of the Gaussian spectrum. Note that $D \propto \sqrt{T}$ and the width of the sidebands increases with temperature.

The long time phonon dynamics ($t \gg \tau$) is easy to obtain by making an asymptotic expansion of $\tilde{\Phi}(t) = \tilde{\Phi}_1(t) + i\tilde{\Phi}_2(t)$ in Eq(5.19) as follows:

\begin{align}
\tilde{\Phi}_1(t) &= \alpha_p^{(1)} \left[ e^{\frac{\tau}{2}} \sqrt{\frac{\pi}{2}} \frac{\tau_{\text{nr}}^3 \tau + \tau_{\text{nr}} (2 + \tau_{\text{nr}}^2)}{\tau_{\text{nr}}^3} \right] \\
&+ e^{-\frac{\tau}{2}(2\tau_{\text{nr}} + \tau)} \left[ 1 - \frac{2\tau_{\text{nr}}}{\tau} + \frac{3\tau_{\text{nr}}^2}{\tau^2} + O\left(\frac{1}{\tau^2}\right) \right]
\end{align}

(5.25a)

\begin{align}
\tilde{\Phi}_2(t) &= -\alpha_p^{(2)} \left[ e^{\frac{\tau}{2}} \sqrt{\frac{\pi}{2}} \frac{\tau_{\text{nr}}^3 \tau + \tau_{\text{nr}} (3 + \tau_{\text{nr}}^2)}{\tau_{\text{nr}}^3} \right] \\
&- e^{-\frac{\tau}{2}(2\tau_{\text{nr}} + \tau)} \left[ \tau - 2\tau_{\text{nr}} + \frac{3\tau_{\text{nr}}^2}{\tau} + O\left(\frac{1}{\tau^2}\right) \right]
\end{align}

(5.25b)
For large $t$ we consider Eq(5.25) to leading order and obtain:

$$
\tilde{\Phi}_1(t) \simeq \alpha_p^{(1)} e^{\frac{\tilde{\gamma}_{nr} t}{2}} \sqrt{\frac{\pi}{2}} \left( \tilde{\gamma}_{nr} t + \tilde{\gamma}_{nr} (2 + \tilde{\gamma}_{nr}) \right) \tag{5.26a}
$$

$$
\tilde{\Phi}_2(t) \simeq -\alpha_p^{(2)} e^{\frac{\tilde{\gamma}_{nr} t}{2}} \sqrt{\frac{\pi}{2}} \left( \tilde{\gamma}_{nr} t + \tilde{\gamma}_{nr} (3 + \tilde{\gamma}_{nr}) \right) \tag{5.26b}
$$

For simplicity denote $\tilde{\Phi}_1(t) = \tilde{\Phi}_1^{(1)}(0) + \tilde{\Phi}_1^{(2)} t$ and similarly we denote $\tilde{\Phi}_2(t) = \tilde{\Phi}_2^{(1)}(0) + \tilde{\Phi}_2^{(2)} t$. The dipole autocorrelation function now has the following asymptotic form for $t \gg \tau$:

$$
e^{-i(\omega_0 - \Delta)t} e^{-(\tilde{\Phi}(0) - \tilde{\Phi}(t)) - \tilde{\gamma}_{nonrad} t} \simeq e^{-i(\omega_0 - \Delta)t} e^{-(\tilde{\Phi}_1^{(1)}(0) - \tilde{\Phi}_1^{(1)}(t))} e^{-i(\tilde{\Phi}_2^{(1)}(0) - \tilde{\Phi}_2^{(1)}(t))} e^{-i(\tilde{\Phi}_2^{(2)} t)} \tag{5.27}
$$

The oscillating part $e^{i\tilde{\Phi}_2^{(2)} t}$ is absorbed into the term oscillating at the atomic resonant frequency $e^{-i(\omega_0 - \Delta)t}$ and provides correction to the polaron shift $\tilde{\Delta}$. The absorption spectrum is now easy to obtain as the imaginary part of the Fourier transform of the dipole autocorrelation function Eq(5.27) and is a Lorentzian with width $\left( \frac{\tilde{\gamma}_{nonrad} - \tilde{\Phi}_1^{(2)}(t)}{\tau} \right)$. Combining the short time and long time dynamics of the phonon correlation function, we conclude that the overall lineshape exhibits Lorentzian behaviour near the peak of the spectrum and decays more rapidly as a Gaussian in the wings.

We now consider population decay dynamics of the two-level atom coupled to the radiation reservoir and in the presence of direct non-radiative decay term $H_{\text{decay}}$. The nonequilibrium correlation function $\langle \hat{C}_+(t)\hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle$ in Eq(3.43) (see the discussion following Eq(3.44)), in the presence of nonradiative decay, can be expressed as (see Appendix C for proof):

$$
\langle \hat{C}_+(t)\hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle \simeq e^{-(\tilde{\Phi}(0) - \tilde{\Phi}(t-t'))} e^{-\tilde{\gamma}_{nonrad}(t-t')} \langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle \tag{5.28}
$$

where we have used $\langle \hat{C}_+(t)\hat{\sigma}_{ge}(t') \rangle = e^{-(\tilde{\Phi}(0) - \tilde{\Phi}(t-t'))} e^{-\tilde{\gamma}_{nonrad}(t-t')}$. Using the Born approximation described in Eq(3.43)-(3.45) and replacing the atomic transition frequency $\omega_0' \rightarrow \omega_0' + i\tilde{\gamma}_{nonrad}$ (see discussion after Eq(5.5)) we obtain:

$$
\langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle \simeq e^{i\omega_0'(t-t')} e^{-\tilde{\gamma}_{nonrad}(t-t')} \langle \hat{\sigma}_{ee}(t') \rangle \tag{5.29}
$$
where \( \omega'_0 = \omega_0 - \Delta \). Using Eq(5.29) and Eq(5.28), Eq(3.46) for \( \ell(t) \) now has the form:

\[
\frac{d\ell(t)}{dt} = -2\gamma_{\text{nonrad}}\ell(t) - \sum_k |\lambda_k|^2 \int_0^t dt' e^{-(\gamma_{\text{nonrad}} + \gamma_{\text{nonrad}})(t-t') - (\Phi(0) - \Phi(t))} \ell(t') e^{i(\omega' - \omega)(t-t')} + c.c. 
\]

In the presence of nonradiative decay, Eq(3.46) now has an additional contribution on the right hand side given by \(- \sum_p (1 + 2n_p) |\zeta_p|^2 \int_0^t dt' \ell(t') e^{i(\omega'_0 - \omega_p)(t-t')} \) where \( \zeta_p \) is the dispersion of this additional decay channel (see Eq(5.5)). In the Markovian approximation we write \( \sum_p (1 + 2n_p) |\zeta_p|^2 \int_0^t dt' \ell(t') e^{i(\omega'_0 - \omega_p)(t-t')} \simeq 2\gamma_{\text{nonrad}}\ell(t) \) and obtain Eq(5.30). Note that in the limit when \( \lambda_k \to 0 \ \forall k \) (no radiative coupling), the population decays due to the first term on the RHS which arises due to the additional decay Hamiltonian Eq(5.5).

Using a simple scaling analysis it is easy to show that, for \( t \ll \alpha^{-1} \), Eq(5.30) simplifies to:

\[
\frac{d\ell(t)}{dt} = -2\gamma_{\text{nonrad}}\ell(t) \quad (5.31)
\]

and the only decay mechanism for population is through the direct nonradiative decay term since the second term on the RHS does not contribute at these timescales. For \( t \simeq \alpha^{-1} \) we now use the asymptotic expansion of \( \Phi(t) \) (valid for \( t \gg \tau \)) outlined in Eq(5.25) and Eq(5.26). Eq(5.30) can then be rewritten as:

\[
\frac{d\ell(t)}{dt} \simeq -2\gamma_{\text{nonrad}}\ell(t) - e^{-\Phi_1(0)} \sum_k |\lambda_k|^2 \int_0^t dt' e^{-\Phi_2(0)} e^{-\tau_{\text{nonrad}}(t-t')} \ell(t') e^{i(\omega'_0 - \omega)(t-t')} + c.c. 
\]

where \( \tau_{\text{nonrad}} = \gamma_{\text{nonrad}} + \gamma_{\text{nonrad}} - \Phi^{(2)}(0) = \Phi_1(0) - \Phi_1^{(1)}(0) \) and \( \Phi_2(0) = \Phi_2(0) - \Phi_2^{(1)}(0) \) and we have absorbed the term \( e^{i\Phi^{(2)}(t-t')} \) in \( e^{i(\omega'_0 - \omega)(t-t')} \). Note that the dot-photon coupling constant is now modified in the presence of direct nonradiative decay as \( |\lambda_k|^2 \to e^{-\Phi_1(0)} |\lambda_k|^2 \). We denote \( e^{-\Phi_1(0)} \sum_k |\lambda_k|^2 e^{i(\omega'_0 - \omega)(t-t')} \equiv \Sigma_R(t-t') \) as the phonon renormalized radiative memory kernel where \( e^{-\Phi_1(0)} \) is the effective Franck-Condon factor which renormalizes the dot-photon coupling. Eq(5.32) is now rewritten as (for \( t \geq \alpha^{-1} \)):

\[
\frac{d\ell(t)}{dt} = -2\gamma_{\text{nonrad}}\ell(t) - e^{-\Phi_2(0)} \int_0^t dt' e^{-\tau_{\text{nonrad}}(t-t')} \Sigma_R(t-t') \ell(t') + c.c. 
\]

We note that the non-radiative decay channel leads to weakening of the photon memory because of the exponential damping term \( e^{-\tau_{\text{nonrad}}(t-t')} \). Using the isotropic dispersion relation Eq(1.34) for the structured reservoir of a photonic crystal, Eq(5.33) can be solved...
by Laplace transformation \[134\] to yield:

\[
\hat{\ell}(s) = \frac{\ell(0)}{s + 2\gamma_{\text{nonrad}} + 2\text{Re } \tilde{Z}(s)} \tag{5.34}
\]

where \[\tilde{Z}(s) = e^{-\overline{\Phi}_1(0)} \frac{a_{\frac{3}{2}}}{\sqrt{s + i\delta + \gamma_{\text{nonrad}}}}e^{-\overline{\Phi}_2(0)}}\). The photonic memory of the structured reservoir is now damped and the two-level system does not support a steady state population. The atom decays nonradiatively at a rate determined by \[\overline{\gamma}_{\text{nonrad}}\] and \[\gamma_{\text{nonrad}}\] in spite of the presence of a photonic band gap.

The polarization dynamics of a quantum dot in a PBG material and coupled to an acoustic phonon reservoir with nonradiative decay is given by (note that unlike the case of population decay we need not make the asymptotic expansion Eq(5.25) and Eq(5.26) for \(t \gg \tau\) and the following discussion is valid for all times):

\[
-i\Theta(t)\langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(0) \rangle \simeq -i\Theta(t)e^{-i\omega_c t - \gamma_{\text{nonrad}} t - \tilde{\gamma}_{\text{nonrad}} t}e^{-(\overline{\Phi}(0) - \overline{\Phi}(t))} \tag{5.35}
\]

\[
[2a_1 x_1 e^{ax_1^2 t} + a_2 (x_2 + y_2)e^{ax_2^2 t} - \sum_{j=1}^{3} a_j y_j \overline{\text{Erfc}}(\sqrt{ax_j^2 t})e^{ax_j^2 t}] \]

and

\[
a_{\frac{3}{2}} = \frac{e^{-\overline{\Phi}_1(0)} \omega_0^2 |D_{ge}|^2}{6\epsilon_0 \hbar c^3} \tag{5.36}
\]

\(x_j\)'s, \(y_j\)'s and \(a_j\)'s are defined in Eq(4.17), \(\overline{\Phi}(t)\) is given by Eq(5.19) and \(\delta = \omega_0 - \tilde{\Delta} - \omega_c\). There is no long term coherence trapped in the photon-atom bound state which, at \(t \gg \tau\), decays at a rate \(\gamma_{\text{nonrad}} + \tilde{\gamma}_{\text{nonrad}} - \frac{\tilde{\Phi}^{(2)}}{\tau}\).

In Fig(5.4) we plot important features of the modified phonon correlation function \(e^{-(\overline{\Phi}(0) - \overline{\Phi}(t) + t\tilde{\gamma}_{\text{nonrad}})}\) in the presence of direct nonradiative decay where \(\overline{\Phi}(t)\) and \(\tilde{\gamma}_{\text{nonrad}}\) are defined in Eq(5.19) and Eq(5.21) respectively. In (a) we plot the modified phonon correlation as a function of \(\frac{t}{\tau}\). Note that the correlation function now decays at long times. In (b) we plot the absorption spectrum which is the Fourier transform of the modified phonon correlation function \(e^{-(\overline{\Phi}(0) - \overline{\Phi}(t) + t\tilde{\gamma}_{\text{nonrad}})}\). The zero-phonon line is now broadened due to nonradiative decay and the spectrum acquires a finite linewidth. For large \(\gamma_{\text{nonrad}}\) the spectrum is almost Lorentzian for \(\omega\) (or large \(t\) in the vicinity of the resonant frequency. In Fig(5.5a) we plot the spectral weight of the zero-phonon line given by \(e^{-\overline{\Phi}(0)}\) as a function of inverse-temperature \(\frac{\hbar \delta}{k_B}\). The zero-phonon line intensity decreases with increasing temperature due to transfer of spectral weight to the sidebands.
and the decay of the phonon correlation function. The Franck-Condon displacement of the excited state wavefunction, $e^{-\mathcal{F}(0)}$ (see discussion following Eq(5.32)), is also studied as a function of direct nonradiative decay in Fig(5.5b). Increasing $\gamma_{\text{nonrad}} \tau$ reduces the Franck-Condon shift and the artificial subnatural linewidth of the radiative transition approaches the natural width.

In Fig(5.6) and Fig(5.7) we study the influence of nonradiative decay on quantum dot polarization $|P_{MF}(t)|$ described by Eq(5.35) and excited state population $l(t)$ described by Eq(5.34) in the structured reservoir of a photonic crystal for various detunings of the atomic transition frequency from the photonic band edge. There is no long time coherence trapped in the photon-atom bound state and no fractionalized steady state population in the presence of direct nonradiative decay for all values of detuning $\delta$. 
Figure 5.4: Salient features of the modified undamped acoustic phonon correlation function in the presence of direct nonradiative decay $e^{-\langle \Phi(0) - \Phi(t) + \gamma_{\text{nonrad}} \rangle}$ where $\Phi(t)$ and $\gamma_{\text{nonrad}}$ are defined in Eq(5.19) and Eq(5.21) respectively. In (a) we plot the modified phonon correlation as a function of $t$, which now decays at long times due to direct nonradiative decay. In (b) we plot the spectral profile corresponding to (a). The $\delta$-function peak in the middle acquires a width for finite nonradiative decay rate. The zero-phonon line resembles a Lorentzian for large $\gamma_{\text{nonrad}}$. The width of the Lorentzian also increases with $\gamma_{\text{nonrad}}$. The large $\omega$ (small $t$) Gaussian spectrum persists in the presence of direct nonradiative decay.
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Figure 5.5: Salient features of the modified undamped acoustic phonon correlation function in the presence of direct nonradiative decay $e^{-\Phi(0)-\Phi(t)+\gamma_{\text{nonrad}}t}$ where $\Phi(t)$ and $\gamma_{\text{nonrad}}$ are defined in Eq(5.19) and Eq(5.21) respectively. In (a) we plot the spectral weight of the zero-phonon line given by $e^{-\Phi(0)}$ as a function of inverse-temperature. The spectral weight decreases with increasing temperature due to excitation of the sidebands. In (b) we study the Franck-Condon shift $e^{-\Phi(0)}$ (see discussion following Eq(5.32)) of the excited state as a function of direct nonradiative decay. Increasing $\gamma_{\text{nonrad}}t$ reduces the Franck-Condon shift. The artificial subnatural linewidth of the radiative transition is thereby removed.
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Figure 5.6: Influence of nonradiative decay on quantum dot dipole autocorrelation function (polarization) $|P_{MF}(t)|$ (Eq(5.35)) in a photonic crystal for detunings from a photonic band edge $\delta/\alpha = -1$ and $\delta/\alpha = -3$. The timescale is measured in units of $\alpha^{-1}$ where $\alpha = 10^{10}\text{s}^{-1}$ for $\omega_0 \approx 10^{15}\text{s}^{-1}$. The rapid loss of coherence at picosecond timescales appears as a nearly discontinuous drop near $t = 0$ on the timescale depicted. There is no long time coherence trapped in the photon-atom bound state in the presence of direct nonradiative decay even though the quantum dot transition is inside the photonic band gap.
Figure 5.7: Influence of nonradiative decay on quantum dot excited state population $l(t)$ (Eq(5.34)) in a photonic crystal for detunings from a photonic band edge $\delta/\alpha = -1$ and $\delta/\alpha = -3$. The timescale is measured in units of $\alpha^{-1}$ where $\alpha = 10^{10} s^{-1}$ for $\omega_0 \simeq 10^{15} s^{-1}$. There is no fractionalized steady state population in the presence of direct nonradiative decay even though the quantum dot transition is inside the photonic band gap.
5.3 Lineshape and Population decay for Damped acoustic phonons

The model Hamiltonian Eq(2.33) describes purely radiative decay from each of the dressed states (phonon sidebands). In ordinary vacuum, this radiative decay is possible from all the dressed states. The linewidth of any given sideband is artificially found to be subnatural. The Franck-Condon displacement of the excited state wavefunction (with a phonon cloud surrounding the excited state quantum dot) diminishes the transition rate from each dressed state. For a quantum dot in ordinary vacuum, coupled to a continuum of undamped acoustic phonons, the subnatural linewidth of individual sidebands leads to a reduced decay rate. In a PBG material, where radiative decay can be completely suppressed over certain spectral ranges, the model Hamiltonian Eq(2.33) leads to untenable result that the lifetime of the photon-atom bound state is increased by phonon interactions. In the previous section, we introduced an independent nonradiative decay mechanism to correct this picture. In this subsection, we demonstrate how the inclusion of finite phonon lifetime leads to a more realistic picture of phonon sideband linewidth and decay of the photon-atom bound state. Our phenomenological phonon damping parameter captures processes like inelastic scattering of phonons and other anharmonic processes that allow the phonons in the polaronic cloud to decay into lower energy phonons and for energy to be dissipated within the vibrational degrees of freedom into a thermal heat bath.

We introduce a phenomenological phonon decay rate $\gamma_q$ into the phonon propagator as follows[127]:

$$iD(t) = \sum_q \eta_q^2 \left[ N_q e^{i(\Omega_q - \gamma_q)t} + (N_q + 1) e^{-(\Omega_q - \gamma_q)t} \right]$$ (5.37)

The self-energy contribution can again be evaluated using the linked cluster theorem and we find a modified $\tilde{\Phi}(t)$ now given by:

$$\tilde{\Phi}(t) = \sum_q \eta_q^2 \left[ \frac{N_q}{(\Omega_q + i\gamma_q)^2} e^{i(\Omega_q - \gamma_q)t} + \frac{N_q + 1}{(\Omega_q - i\gamma_q)^2} e^{-(\Omega_q - \gamma_q)t} \right]$$ (5.38)

This is identical to Eq(5.14b) except with the single nonradiative decay rate $\gamma_{nonrad}$ replaced with a set of phonon damping coefficients $\{\gamma_q\}$. As before we write $\tilde{\Phi}(t) = $
\( \hat{\Phi}_1(t) + i\hat{\Phi}_2(t) \). Also \( \hat{\Phi}(0) \) can be decomposed as \( \hat{\Phi}(0) = \hat{\Phi}_1(0) + i\hat{\Phi}_2(0) \) where \( e^{-\hat{\Phi}_1(0)} \) is the intensity of the zero-phonon line (Huang-Rhys factor) and \( \hat{\Phi}_2(0) \) is an additional phase:

\[
\hat{\Phi}_1(0) = \sum_q \eta_q^2 \frac{\Omega_q^2 - \gamma_q^2}{(\Omega_q^2 + \gamma_q^2)^2} (1 + 2N_q) \quad (5.39a)
\]

\[
\hat{\Phi}_2(0) = \sum_q \eta_q^2 \frac{2\Omega_q\gamma_q}{(\Omega_q^2 + \gamma_q^2)^2} \quad (5.39b)
\]

Also \( \hat{\Phi}'(0) = \Gamma_{damp} - i\tilde{\Delta} \) where \( \Gamma_{damp} \) is an additional decay term contributing to the polarization decay due to phonon damping and \( \tilde{\Delta} \) is the modified polaron shift. They are given by

\[
\Gamma_{damp} = \sum_q \frac{\eta_q^2\gamma_q}{\Omega_q^2 + \gamma_q^2} (1 + 2N_q) \quad (5.40)
\]

\[
\tilde{\Delta} = \sum_q \eta_q^2 \frac{\Omega_q}{\Omega_q^2 + \gamma_q^2}
\]

For analytical simplicity we consider a solvable model and assume that the phonon damping is wave-vector dependent and has the form \( \gamma_q = \gamma \Omega_q \) where \( \gamma \) is a real number between zero and one. The most efficient of anharmonic three-phonon processes, the Landau-Rumer process of relaxation of a transverse acoustic mode into thermal longitudinal modes, has a dependence of this form[150, 151]:

\[
\gamma_q = \frac{6G^2(k_BT)^4(1 + \frac{2c_t}{3c_l})}{\pi \rho c_T c_l^3 \hbar^3 \Omega_q} \quad (5.41)
\]

where \( c_t \) (\( c_l \)) is the transverse (longitudinal) sound velocity and \( G \) is the Gruneissen constant (taken to be 2).

The modified Phonon correlation function due to phonon damping can be easily evaluated using Eq(5.37) and Eq(5.13) for \( W(t) \) and is given by (in the approximation \( \coth(\frac{\hbar \omega}{2T}) \approx \frac{2T}{\hbar \omega} \)):

\[
W(t) = \alpha_p \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^\infty dxx^3 \exp\left( -\frac{x^2}{2} \right) (i \sin(x\tilde{t}_{12}) - \frac{2T}{\hbar \beta x} \cos(x\tilde{t}_{12})) e^{-\gamma x\tilde{t}_{12}} \quad (5.42)
\]

where \( \tilde{t}_{12} = \frac{t_1 - t_2}{\tau} \). This differs from the case of direct nonradiative decay Eq(5.17) through the appearance of the dimensionless wave vector of the damped acoustic phonons in the
final temporal exponential. The function $W(t)$ is easily evaluated and is given by:

$$W(t) = -\alpha^{(1)}_p \frac{(1 - \gamma^2) + \sqrt{\frac{27}{\pi}} \gamma (1 + \gamma^2) + (1 + \gamma^2)^2 \Re \left[ \frac{e^{z_2 E r f c[z]}}{-i + \gamma} \right]}{(1 + \gamma^2)^2}$$

$$+ \ i \alpha^{(2)}_p \frac{-1 + (1 + \gamma^2) \Im \left[ \frac{e^{z_2 E r f c[z]}}{-i + \gamma} \right]}{(1 + \gamma^2)}$$

where $z = \frac{(-i + \gamma \tau)}{\sqrt{2}}$, $\alpha^{(1)}_p = \sqrt{\frac{2}{\pi}} \alpha_p \tau$ and $\alpha^{(2)}_p = \sqrt{\frac{2}{\pi}} \alpha_p$. The modified phonon correlation function is straightforward to obtain from Eq(5.43) we obtain:

$$\tilde{\Phi}^{(1)}_1(t) = -\alpha^{(1)}_p \Re \left[ \frac{e^{z_2 E r f c[z]}}{-i + \gamma} \right]$$

$$\tilde{\Phi}^{(2)}_2(t) = \alpha^{(2)}_p \frac{1}{1 + \gamma^2}$$

The zero-phonon line spectral weight $e^{-\tilde{\Phi}(0)}$ which is determined by $\tilde{\Phi}^{(1)}_1(0) = \alpha^{(1)}_p \frac{1 + \chi^2}{(1 + \gamma^2)}$ and the phase $\tilde{\Phi}^{(2)}_2(0) = 0$. The linear decay contribution $\Gamma_{damp}$ and the modified polaron shift $\tilde{\Delta}$ are now given by:

$$\Gamma_{damp} \tau = \alpha^{(1)}_p \sqrt{\frac{2}{\pi}} \frac{\gamma}{1 + \gamma^2}$$

$$\tilde{\Delta} \tau = \alpha^{(2)}_p \frac{1}{1 + \gamma^2}$$

The short time $t \ll \tau$ phonon dynamics contained in $e^{-\tilde{\Phi}(0) - \tilde{\Phi}(t) + i \tilde{\Phi}^{(0)}}$ is identical to the case of direct nonradiative decay obtained previously in Eq(5.23). The wings of the spectrum are Gaussian with width $D^2 = \alpha^{(2)}_p \frac{1}{1 + \gamma^2}$. It is instructive to note that the short time phonon dynamics is independent of the nature of phonon damping or direct nonradiative decay. In fact it is identical to the case of undamped phonons. To see this we observe that: $e^{-\tilde{\Phi}(0) - \tilde{\Phi}(t) + i \tilde{\Phi}^{(0)}} \simeq e^{\frac{\gamma^2}{\tau} \tilde{\Phi}^{(0)}}$. Using the general expressions for $\tilde{\Phi}(t)$ obtained in Eq(1.14) for undamped phonons, Eq(5.14b) for direct nonradiative decay and Eq(5.38) for damped phonons we find:

$$\tilde{\Phi}^{(0)}(0) = -\sum_q \gamma^2_q (1 + 2N_q)$$

$$= -\frac{2\alpha_p}{\hbar \beta \tau} \int_0^\infty dx x^2 e^{-\frac{x^2}{2}} = -\frac{\alpha^{(1)}_p}{\tau^2}$$
This can also be verified using explicit expressions for $\tilde{\Phi}(t)$ obtained in Eq(5.19) for direct nonradiative decay and Eq(5.44) for damped phonons.

In the long time $t \gg \tau$ regime, the Error functions in Eq(5.44) for $\tilde{\Phi}_1(t)$ and $\tilde{\Phi}_2(t)$ can be replaced, to a very good approximation, by:

$$Er f c[z] \simeq \frac{e^{-z^2}}{\sqrt{\pi z}} \quad (|z| \gg 1) \quad (5.47)$$

Eq(5.44) for $\tilde{\Phi}_1(t)$ and $\tilde{\Phi}_2(t)$ then has a simple form:

$$\tilde{\Phi}_1(t) \simeq \alpha_{p}^{(1)} \frac{\sqrt{2 \pi \gamma (3 - \gamma^2)}}{t(1 + \gamma^2)^3} \quad (T \gg 1) \quad (5.48a)$$

$$\tilde{\Phi}_2(t) = -\alpha_{p}^{(2)} \frac{2\sqrt{2 \pi \gamma}}{(1+\gamma^2)^2} \quad (T \gg 1) \quad (5.48b)$$

Using the fact that $\tilde{\Phi}_1(t) \to 0$ for $T \gg 1$ and neglecting the time-independent factor $\tilde{\Phi}_2(t)$, we approximate $e^{-\tilde{\Phi}(0)-\tilde{\Phi}(t)+t\tilde{\Phi}(0)}$ (using $\tilde{\Phi}_2(0) = 0$, see Eq(5.44)) by $e^{-\tilde{\Phi}_1(0)-t\tilde{\Phi}(0)}$. It follows that for $T \gg 1$ (using $\Phi(0) = \Gamma_{damp} - i\bar{\Delta}$ and Eq(5.45) for $\Gamma_{damp}$):

$$e^{-(\tilde{\Phi}(0)-\tilde{\Phi}(t)+t\tilde{\Phi}(0))} \simeq e^{-\tilde{\Phi}_1(0)+i\bar{\Delta}t} e^{-\frac{2\gamma^2}{h_0(1+\gamma^2)}T} \quad (5.49)$$

The spectrum (Fourier transform of the dipole autocorrelation function) clearly exhibits a Lorentzian peak with linewidth $\frac{2\gamma^2}{(1+\gamma^2)(\hbar/\gamma)}$. As $\gamma \to 0$ the Lorentzian peak transforms to a delta-function centered at the atomic transition frequency with a broad Gaussian background as in the case of undamped acoustic phonons (see Section VI.A). The zero-phonon line is unbroadened in the absence of phonon damping, which is characteristic of pure dephasing.

The atomic dipole autocorrelation function in the structured reservoir of a photonic crystal is now given by (valid for all times):

$$-i\tilde{\Theta}(t)\langle \hat{\sigma}_{ge}(t)\hat{\sigma}_{eg}(0) \rangle \simeq -i\tilde{\Theta}(t)e^{-i\omega_c t - \Gamma_{damp} t}e^{-\tilde{\Phi}(0)-\tilde{\Phi}(t)} \quad (5.50)$$

$$[2a_1x_1e^{\alpha x^2 t} + a_2(x_2 + y_2)e^{\alpha x_2^2 t} - \sum_{j=1}^{3} a_j y_j Erf c(\sqrt{\alpha x_j^2 t}) e^{\alpha x_j^2 t}]$$
where

$$\alpha^2 = \frac{e^{-\tilde{\Phi}_1(0)}|\omega|^2}{6\pi\epsilon_0\hbar c^3}$$  \hspace{1cm} (5.51)$$

$x_j$'s, $y_j$'s and $a_j$'s are defined in Eq(4.17) and $\delta = \omega_0 - \tilde{\Delta} - \omega_e$. $\Gamma_{\text{damp}}$ and $\tilde{\Delta}$ are defined in Eq(5.45) and $\tilde{\Phi}(t)$ is defined in Eq(5.44). Note that the photon-atom bound state loses all polarization on the time scale $\Gamma_{\text{damp}}^{-1}$.

We now consider the population decay dynamics of the excited state in the presence of acoustic phonon damping. The nonequilibrium correlation function $\langle \hat{C}_+(t)\hat{\sigma}_{eg}(t)\hat{C}_-(t')\hat{\sigma}_{ge}(t') \rangle$ in Eq(3.43) (see the discussion following Eq(3.44)), in the presence of phonon damping, can be expressed as:

$$\langle \hat{C}_+(t)\hat{\sigma}_{eg}(t)\hat{C}_-(t')\hat{\sigma}_{ge}(t') \rangle \simeq e^{-\langle \tilde{\Phi}(0) - \tilde{\Phi}(t-t') \rangle} e^{-\Gamma_{\text{damp}}(t-t')} \langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle$$  \hspace{1cm} (5.52)$$

where we have used $\langle \hat{C}_+(t)\hat{C}_-(t') \rangle = e^{-\langle \tilde{\Phi}(0) - \tilde{\Phi}(t-t') \rangle} e^{-\Gamma_{\text{damp}}(t-t')}$ (see Appendix C for proof). In the Born approximation as in Eq(3.45), Eq(5.52) can be simplified as:

$$\langle \hat{C}_+(t)\hat{\sigma}_{eg}(t)\hat{C}_-(t')\hat{\sigma}_{ge}(t') \rangle \simeq e^{-\langle \tilde{\Phi}(0) - \tilde{\Phi}(t-t') \rangle} e^{i\omega_0(t-t') - \Gamma_{\text{damp}}(t-t')} \langle \hat{\sigma}_{ee}(t') \rangle$$  \hspace{1cm} (5.53)$$

Using Eq(5.48) in Eq(5.53) and noting that $\tilde{\Phi}_2(0) = 0$, $\tilde{\Phi}_1(t) \rightarrow 0$ for $t \gg \tau$ and $e^{-i\tilde{\Phi}_2(t)} \simeq 1$, Eq(3.46) for $\ell(t)$ for long time scales is governed by the integro-differential equation:

$$\frac{\partial \ell(t)}{\partial t} \simeq - \frac{e^{-\tilde{\Phi}_1(0)}}{\sum |\lambda_k|^2} \int_0^t \! dt' e^{-\Gamma_{\text{damp}}(t-t')} e^{i\omega_0(t-t') - \Gamma_{\text{damp}}(t-t')} + \text{c.c.}$$  \hspace{1cm} (5.54)$$

Solving Eq(5.54) for the structured reservoir of a photonic crystal using the isotropic dispersion model of Eq(1.34) we find $\tilde{\ell}(s) = \frac{i\ell(0)}{s + 2\Re Z(s)}$ where:

$$\tilde{Z}(s) = \frac{\alpha^2 e^{i\tilde{\Phi}_1(0)}}{\sqrt{s + i\tilde{\delta} + \Gamma_{\text{damp}}}}$$  \hspace{1cm} (5.55)$$

We note that in spite of the absence of any direct non-radiative decay channel, the atom does not have a steady state population even if the atomic transition frequency is inside the photonic band gap and decays at long times at a rate determined by $\Gamma_{\text{damp}}$. For example, if we consider the regime $\Gamma_{\text{damp}} \gg \delta$, then $\tilde{\ell}(s) \simeq \ell(0)/(s + \gamma_{\text{ph}})$ where $\gamma_{\text{ph}} \simeq e^{-\tilde{\Phi}_1(0)}\sqrt{2\alpha^3 \Gamma_{\text{damp}}}$. It follows that $\ell(t) \simeq \ell(0)e^{-\gamma_{\text{ph}}t}$ for large times. Hence there is
no fractionalized steady state irrespective of the sign of $\delta$. Unlike the case of purely nonradiative decay (where decay occurs independently of radiative coupling) our phonon damping scales with the strength of the radiative coupling constant. Phonon damping may be regarded as a vital correction to our mean-field description Eq(2.39) of the quantum dot dressed by phonons. In this mean-field theory, we neglected decay of the atomic polarization by the simultaneous emission of photons and phonons. The various phonon-sidebands were only allowed to decay by purely radiative emission (with a Franck-Condon displaced radiative coupling coefficient). Our phenomenological phonon damping process now describes the phonon-assisted decay of the atomic polarization. Phonon damping also provides a cutoff to the extent of Franck-Condon displacement of the excited state (atom plus acoustic phonons) wavefunction.

In Fig(5.8) and Fig(5.9) we study characteristic features of the modified phonon correlation function $e^{-(\tilde{\Phi}(0)-\tilde{\Phi}(t)+i\Gamma_{damp}t)}$ in the presence of phonon damping where $\tilde{\Phi}(t)$ and $\Gamma_{damp}$ are defined in Eq(5.44) and Eq(5.45) respectively. The modified phonon correlation function decays at long times due to finite lifetime of phonons. The zero-phonon line now acquires a finite width $\frac{2\gamma\gamma_{p}K_{B}T}{(1+\gamma^{2})} (\hbar/\gamma)$ due to phonon damping. The spectral weight of the zero-phonon line given by $e^{-\tilde{\Phi}(0)}$ (defined in Eq(5.45)) is found to decrease with increasing temperature as expected. The Franck-Condon shift of the excited state $e^{-\tilde{\Phi}(0)}$ (also given by Eq(5.45)) is now limited by phonon lifetime. Increasing $\gamma$ reduces the Franck-Condon shift and the artificial subnatural linewidths are removed.

In Fig(5.10) and Fig(5.11) we study the influence of finite phonon lifetimes on quantum dot polarization $|P_{MF}(t)|$ (Eq(5.50)) and excited state population $l(t)$ (Eq(5.55)) in the structured reservoir of a photonic crystal for various detunings of the atomic transition frequency from the photonic band edge. No long time coherence is trapped in the photon-atom bound state and the population decays to zero at a rate determined by the lifetime of the phonons surrounding the quantum dot.
Figure 5.8: Salient features of the modified acoustic phonon correlation function in the presence of phonon damping $e^{-(\Phi(0) - \Phi(t) + i\Gamma_{damp})}$ where $\Phi(t)$ and $\Gamma_{damp}$ are defined in Eq(5.44) and Eq(5.45) respectively. In (a) we plot the modified phonon correlation as a function of $t$. The correlation function decays at long times due to finite lifetime of phonons. In (b) we plot the spectral profile corresponding to (a). The $\delta$-function peak in the middle acquires a width for finite phonon damping. The zero-phonon line resembles a Lorentzian for large phonon damping $\gamma$ and its width increases with increasing $\gamma$. The large $\omega$ (small $t$) Gaussian spectrum remains as in the case of undamped acoustic phonons.
Figure 5.9: Salient features of the modified acoustic phonon correlation function in the presence of phonon damping $e^{-(\Phi(0) - \Phi(t) + i\Gamma_{damp})}$ where $\Phi(t)$ and $\Gamma_{damp}$ are defined in Eq(5.44) and Eq(5.45) respectively. In (a) we plot the spectral weight of the zero-phonon line given by $e^{-\Phi(0)}$ (defined in Eq(5.44)) as a function of inverse-temperature. It is found to decrease with increasing temperature. In (b) we depict the Franck-Condon shift $e^{-\Phi(0)}$ (also given by Eq(5.44)) of the excited state as a function of phonon lifetime. Increasing $\gamma$ reduces the Franck-Condon shift.
Figure 5.10: Influence of finite acoustic phonon lifetimes on quantum dot dipole autocorrelation function (polarization) $|P_{MF}(t)|$ in a photonic crystal for detunings from a photonic band edge $\delta/\alpha = -1$ and $\delta/\alpha = -3$. The timescale is measured in units of $\alpha^{-1}$ where $\alpha = 10^{10}\text{s}^{-1}$ for $\omega_0 \simeq 10^{15}\text{s}^{-1}$. The polarization dynamics is governed by Eq(5.50). No long time coherence is trapped in the photon-atom bound state with finite phonon lifetimes even though the quantum dot transition is inside the photonic band gap.
Figure 5.11: Influence of finite acoustic phonon lifetimes on quantum dot excited state population $l(t)$ in a photonic crystal for detunings from a photonic band edge $\delta/\alpha = -1$ and $\delta/\alpha = -3$. The timescale is measured in units of $\alpha^{-1}$ where $\alpha = 10^{10}\text{s}^{-1}$ for $\omega_0 \simeq 10^{13}\text{s}^{-1}$. The excited state population dynamics given by Eq(5.55). No long time fractionalized steady state population is found with finite phonon lifetimes even though the quantum dot transition is inside the photonic band gap.
5.4 Damped optical phonons

Finally we consider the modifications in the polarization and population dynamics of the quantum dot coupled to a bath of damped optical phonons and placed inside the structured reservoir of a photonic crystal. We associate a finite lifetime $\gamma_0$ to the optical phonons. The modified phonon correlation function $\tilde{\Phi}(t)$ (see Eq(5.38)) is now given by:

$$\tilde{\Phi}(t) = [\tilde{g}Ne^{i\Omega_0t} + \tilde{g}^* (N + 1)e^{-i\Omega_0t}]e^{-\gamma_0 t}$$  \hspace{1cm} (5.56)

where $\tilde{g} = g \frac{\Omega_0^2}{(1 + \frac{\Omega_0^2}{\tilde{\Omega}_0^2})^2}$, $g = \sum_q \frac{\tilde{\Omega}_0^2}{\tilde{\Omega}_0^2}$ (see Eq(3.13)) and $N = \frac{1}{e^{\frac{\tilde{\Omega}_0}{\Omega_0} - 1}}$. The modified dimensionless dot-phonon coupling (Huang-Rhys factor) $\tilde{\Phi}_1(0)$ and the phase $\tilde{\Phi}_2(0)$ are now given by:

$$\tilde{\Phi}_1(0) = \frac{g\Omega_0^2(\Omega_0^2 - \tilde{\gamma}_0^2)}{(\Omega_0^2 + \tilde{\gamma}_0^2)^2}(1 + 2N)$$  \hspace{1cm} (5.57a)

$$\tilde{\Phi}_2(0) = \frac{2g\Omega_0^3\gamma_0}{(\Omega_0^2 + \tilde{\gamma}_0^2)^2}$$  \hspace{1cm} (5.57b)

The Franck-Condon shift of the excited state is also given by $e^{-\tilde{\Phi}_1(0)}$. Note that as $\gamma_0 \to \Omega_0$, $\tilde{\Phi}_1(0) \to 0$ and the Frank-Condon shift is cutoff. Also the additional decay term $\Gamma_{damp}$ resulting in polarization decay and the modified polaron shift $\tilde{\Delta}$ are given by:

$$\Gamma_{damp} = \frac{g\gamma_0\Omega_0^2}{\Omega_0^2 + \gamma_0^2}(1 + 2N)$$  \hspace{1cm} (5.58)

$$\tilde{\Delta} = \frac{g\Omega_0^3}{\Omega_0^2 + \gamma_0^2}$$

It is useful to expand the phonon correlation function $e^{-(\tilde{\Phi}(0) - \tilde{\Phi}(t) + i\Gamma_{damp}t)}$ as a power series in $e^{i\Omega_0t}$ as was performed in Eq(3.18). It is convenient to rewrite Eq(5.56) as:

$$\tilde{\Phi}(t) = |\tilde{g}|[Ne^{-i\phi_0}e^{i\Omega_0t} + (N + 1)e^{i\phi_0}e^{-i\Omega_0t}]e^{-\gamma_0 t}$$  \hspace{1cm} (5.59)

where $|\tilde{g}| = g\frac{\Omega_0^2}{\Omega_0^2 + \gamma_0^2}$ and $\phi_0 = \cos^{-1}\frac{\Omega_0^2 - \gamma_0^2}{\Omega_0^2 + \gamma_0^2}$. The series expansion of the phonon correlation function is now straightforward to perform and we find:

$$e^{-(\tilde{\Phi}(0) - \tilde{\Phi}(t) + i\Gamma_{damp}t)} = \sum_{n=-\infty}^{\infty} L_n(t)e^{-in\Omega_0t - \Gamma_{damp}t}$$  \hspace{1cm} (5.60)
where

$$L_n(t) = e^{-|g|(2N+1)\phi} e^{in\tilde{\phi}_0} e^{n\Omega_0\beta/2} I_n(2|g|\sqrt{N(N+1)}e^{-\gamma_0 t})$$

(5.61)

The exponential time dependence of the argument in the modified-Bessel function renders analytical Fourier transformation problematic. Substantial simplification is achieved by approximating the Bessel’s functions as:

$$I_n(x) \approx \left(\frac{x}{2}\right)^n \frac{\Gamma(n+1)}{n!} \quad 0 < x \ll \sqrt{n+1}$$

(5.62)

This is valid for small Huang-Rhys factor (very weak dot-phonon coupling) and low temperatures. The expansion coefficients $L_n$ in Eq(5.60) are now simplified as:

$$L_n(t) = e^{-|g|(2N+1)\phi} e^{in\tilde{\phi}_0} e^{n\Omega_0\beta/2} \frac{|g|^n \sqrt{N(N+1)}|n|}{|n|!} e^{-|n|\gamma_0 t}$$

(5.63)

Note that the previously unbroadened phonon sidebands in the case of undamped optical phonons now acquire a finite width. The $n^{th}$ phonon sideband has a width $|n|\gamma_0 + \Gamma_{\text{damp}}$. The zero phonon line is also broadened with linewidth $\Gamma_{\text{damp}}$. Hence all the sidebands acquire a Lorentzian lineshape but with different linewidths.

The atomic dipole autocorrelation function in the structured reservoir of a photonic crystal is given by Eq(5.50) with $\alpha^\dagger$ defined in Eq(5.51). $x_j$’s, $y_j$’s and $a_j$’s are defined in Eq(4.17), $\tilde{\Phi}(t)$ is defined in Eq(5.56), $\Gamma_{\text{damp}}$ is defined in Eq(5.40) and $\delta = \omega_0 - \tilde{\Delta} - \omega_e$. The $n^{th}$-phonon dressed state decays at a rate $n|\gamma_0 + \Gamma_{\text{damp}}$. The decay of the zero-phonon line is the slowest and the photon-atom bound state loses all polarization on the time scale $\Gamma_{\text{damp}}^{-1}$.

The population of the excited state $\ell(t)$ is straightforward to study using Eq(5.60) in Eq(3.46). We now obtain for $\ell(t)$:

$$\frac{\partial \ell(t)}{\partial t} = -\sum_{n=-\infty}^{\infty} L_n(0) \int_0^t dt' e^{-in\tilde{\phi}_0(t-t')} e^{-|n|\gamma_0(t-t')} e^{-\Gamma_{\text{damp}}(t-t')} \Sigma(t-t') \ell(t') + c.c.$$

(5.64)

Eq(5.64) is easily solved by Laplace transformation to yield:

$$\tilde{\ell}(s) = \frac{\ell(0)}{s + 2 \text{Re} Z(s)}$$

(5.65)
where

\[ Z(s) = \sum_{n=-\infty}^{\infty} L_n(0) \frac{\alpha^2 e^{i\pi s}}{\sqrt{s + i\delta_n + |n|^2\gamma_0 + \Gamma_{\text{damp}}}} \]  

(5.66)

and \( \delta_n = \delta - n\Omega_0 \) with \( L_n(0) \) is obtained by setting \( t = 0 \) in Eq(5.63). The excited atom now decays even if it is detuned deep inside the photonic band gap. Compared to the continuum of damped acoustic phonons the population decay due to damped optical phonons is attributed to the simultaneous decay of all the discrete dressed states with different decay rates.

### 5.5 Discussion

In this chapter we have presented a detailed discussion of the effects of undamped acoustic phonons, phonon-assisted electronic decay processes and damped acoustic and optical phonons on the polarization and population dynamics of a quantum dot placed inside the structured reservoir of a photonic crystal. As was discussed in the introduction, the polaronic state formed by the two-level system and a bath of undamped phonons provides a correct picture of dephasing at low temperatures. Coherence loss at picosecond-timescales in this scenario is not accompanied with any population decay and the long time polarization decay is radiatively limited. However at higher temperatures, additional processes become relevant and the Independent – Boson model is insufficient due to the absence of any long time decay mechanism for the coherence of the polaronic state (except due to electromagnetic dephasing). Phonon-assisted decay processes, inelastic phonon scattering and anharmonic phonon processes can relax the polaron state at sub-nanosecond timescales and provide an additional contribution to the linewidth along with radiative decay. These additional nonradiative processes are important at higher temperatures \( (T > 50K) \) and it is important to include these processes in the Independent – Boson model. Since a fully quantum mechanical and microscopic description of all these processes is cumbersome and probably of little value, we include them at a phenomenological level by a suitable modification of the phonon correlation function. Nonetheless this phenomenological description allows us to express nonradiative decay rates in terms of microscopic parameters.

We first considered an undamped acoustic phonon reservoir coupled bilinearly to the quantum dot population operator \( (\text{Independent} – \text{Boson} \text{ model}) \). The role of phonons was limited to pure dephasing which manifested as a rapid but partial dephasing on
picosecond timescales. This rapid dephasing with no associated population decay is attributed to the formation of the polaronic cloud. For a quantum dot placed inside the structured reservoir of a photonic crystal, the long time polarization may be non-zero because of the formation of a photon-atom bound state (for negative detunings of the atomic transition frequency from the band edge). This non-zero residual coherence can be of importance for quantum information processing applications. The long time excited state population in a photonic crystal was also found to be non-zero due to the fractionalized steady state. Interestingly the fractionalized steady state population was found to increase with increasing dot-phonon coupling strength and increasing temperatures. This was explained by the dephasing of the atomic transition dipole moment by phonons resulting in a weaker coupling to the structured radiative reservoir.

We then generalized our discussion to include non-radiative decay processes. These processes are beyond the framework of our starting microscopic Hamiltonian and corresponds to the direct coupling of phonon operators to the quantum dot dipole. A fully microscopic Hamiltonian including nonradiative processes is cumbersome to solve. We included nonradiative decay by introducing a decay rate which could be related to the microscopic parameters of the decay process. A suitable correction to the undamped phonon propagator due to nonradiative decay (during the times the dot interacted with the purely dephasing phonons) allowed us to derive a modified phonon correlation function. This simultaneous interaction of the nonradiative decay processes and purely dephasing processes resulted in a modified effective dot-pure dephasing phonon coupling. The spectral profile of the Independent – Boson model was qualitatively modified by direct nonradiative processes. The zero-phonon line now acquired a finite linewidth which increased with nonradiative decay rate. The Franck-Condon displacement of the excited state was also cutoff due to direct nonradiative decay. We found that there was no long time residual coherence even when the quantum dot was placed deep inside the photonic band gap. The overall rate of polarization decay increased with temperature and nonradiative decay coupling strength. Similarly there was no fractionalized steady state population with the long time decay rate being determined by the radiative and nonradiative decay rate.

We then considered another process not included in the Independent – Boson model which provides important corrections to the optical lineshape and polarization and population dynamics of the quantum dot. We argued that phonon-phonon scatterings and anharmonic phonon processes could lead to decay of the otherwise infinitely long-lived
phonon. Damping of acoustic and optical phonons was shown to produce a qualitative modification of the phonon correlation function quite similar to the case of direct non-radiative processes. As in the case of direct nonradiative decay, damping of acoustic phonons resulted in the broadening of the zero-phonon line. However direct nonradiative decay produced additional broadening of the purely dephasing phonon sidebands (of the Independent – Boson model) which was absent in the case of damped acoustic phonons. The Franck-Condon displacement of the excited state was also cutoff due to finite phonon lifetimes. In the case of damped optical phonons all the distinct and well-separated sidebands acquired a Lorentzian lineshape but with different linewidths. As in the case of direct non-radiative decay, there was no long time residual coherence in the presence of damped acoustic and optical phonons even when the quantum dot transition frequency is placed deep inside the photonic band gap. Similarly there was no fractionalized steady state population on the excited state in the presence of phonon damping.

We now discuss the experimental verifiability of the theoretical predictions made in this chapter. For the physically relevant acoustic phonon models (in the presence of phonon-assisted decay of the excited state and finite phonon damping), the polarization and population decay rates is obtained in terms of the material properties of the quantum dot and can be compared to experimental data. As an example consider the case of acoustic phonon damping. The dominant decay mechanism is an anharmonic three-phonon process (Landau-Rumer process) which results in the relaxation of a transverse acoustic mode into a thermal longitudinal mode. Using material values of a GaAs quantum dot ($\rho = 5.37 \text{ g/cm}$, $c_L = 5110 \text{ m/s}$ and $c_T = 3340 \text{ m/s}$), the phonon damping rate can be evaluated. For example, with $q = 0.2 \text{ nm}^{-1}$ and at a temperature of $50 \text{ K}$, the linewidth of phonon due to the Landau-Rumer process is found to be $0.5 \mu \text{ eV}$. Using a simple exponential curve fitting to experimentally observed data of polarization and population decay at different temperatures, the long time decay rates can be extracted. These values can subsequently be compared to the theoretically predicted decay rates. The temperature dependence of the polaron shift and the width of the absorption spectrum are also of experimental interest. For the case of damped acoustic phonons, the broadened spectrum (due to Landau-Rumer process) is given by $\Gamma_{\text{damp}} = 0.082 \frac{K_B T}{0.7 \text{ meV}} \left(\frac{\Delta_0}{0.04 \text{ ps}}\right)^4 \text{ ps}^{-1}$ ($\text{ps} \equiv \text{picosecond}$). The polaron shift is given by $\Delta = \frac{0.04}{1+\gamma} \text{ ps}^{-1}$ and depends on temperature through its dependence on the phonon damping parameter $\gamma$. The polaron shift can also be written as $\Delta = \frac{0.04}{1+\gamma} \left(\frac{\Delta_0}{0.04 \text{ ps}}\right)^4 \text{ ps}^{-1}$ and experimentally verified.
Chapter 6

Conclusions

In this thesis we have presented a detailed discussion of optical properties of single self-assembled semiconductor quantum dots placed in the structured reservoir of a photonic crystal. Self-assembled quantum dots have several features which distinguish them from conventional bulk semiconductors or 2-D quantum wells. These features are exemplified in the interaction of the quantum dot with the nonradiative degrees of freedom (phonons) and its effect on the optical lineshape. Unlike bulk semiconductors or other low-dimensional semiconductors, energy conservation requirements impose stringent conditions on the nature of possible quantum dot-phonon interactions. At low temperatures this interaction is well described by the Independent – Boson model wherein phonons cause pure dephasing with no associated population decay. The interaction of the quantum dot with the electromagnetic reservoir is accurately described using the standard quantum electrodynamical formulation of atom-photon interaction in the dipole approximation. The nature of the optical lineshape is characteristic of the spectral features of the electromagnetic reservoir. To the best of our knowledge, studies until now have focused primarily on the optical lineshape in the unstructured electromagnetic reservoir of free space. However, the structured electromagnetic reservoir of a photonic crystal introduces substantial modifications in atom-photon interactions which are of immense value in practical applications. A theoretical description of non-Markovian quantum optical aspects of quantum dot-photon interactions conditioned by relevant quantum dot-phonon interactions is useful to understand and stimulate experimental progress in these areas. This thesis provides a formalism and presents some salient features of quantum optical properties of zero-dimensional mesoscopic semiconductor quantum dots interacting with the structured electromagnetic reservoir of a photonic crystal.
Conventional formulations of the optical lineshape problem in unstructured electromagnetic reservoirs only provide a partial picture. In the stochastic approach to the optical lineshape problem, the interaction of the quantum dot with lattice degrees of freedom is described as a modulation of the two-level system transition frequency. The modulation is mathematically treated as stochastic fluctuations of the excited state frequency. Dephasing is now governed by the root mean square of the stochastic modulations and the correlation time of the modulations of the transition frequency of the quantum dot about the average. However, none of these parameters can be related to the microscopic parameters of the underlying phonon reservoir such as quantum dot-phonon coupling or the temperature of the phonon reservoir. Moreover such an approach ignores the effects of these stochastic fluctuations on the radiative degrees of freedom. As was shown in this thesis, phonon dynamics can modify the dot-photon coupling strength by dephasing the atomic dipole moment. This cannot be demonstrated in the stochastic approach. The complete optical lineshape in this case is then given by the product of the radiative lineshape and nonradiative dephasing described as stochastic modulations of the quantum dot transition frequency. In the semiclassical approach to the optical lineshape problem, the lattice modes are described quantum mechanically assuming a parabolic adiabatic potential for the phonons. This results in the lattice modes described as harmonic oscillators following second quantization. However, the dependence of the quantum dot dipole moment on the phonon coordinates is ignored (Condon approximation). These approaches to the optical lineshape problem, though sufficient in the homogenous reservoir of free space, are not applicable to the structured electromagnetic reservoir of a photonic crystal. A complete treatment of the optical lineshape problem is possible by starting with a full quantum mechanical description of the radiative and non-radiative degrees of freedom. Such an approach allows us to study the effects of the phonon reservoir on the electromagnetic degrees of freedom which cannot be obtained either in the stochastic formulation or the semiclassical approach. It also enables us to study the dependence of the optical lineshape on the microscopic parameters of the phonon and electromagnetic reservoirs.

We first presented a mean-field theory for the role of phonons in modifying the optical properties of two level systems. Using a thermodynamic Green’s function formalism, the combined effect of photon and phonon reservoirs was shown to provide dressing of the atomic states with various numbers of phonons and the optical absorption spectrum was shown to consist of transitions between the atomic ground state and various
dressed excited states (phonon sidebands). In our mean-field theory, the lattice displacement operator was replaced by an equilibrium thermal average over the phonon reservoir. This led to an effective temperature-dependent radiative coupling constant describing the (Franck-Condon) overlap between the displaced excited state and the ground state. In this mean-field theory, phonon assisted decay of the atomic polarization was removed from the Hamiltonian. The linewidth of each dressed state was determined entirely by the Franck-Condon displaced optical transition dipole matrix element and the electromagnetic density of states at the dressed state frequency. Phonon assisted decay processes were then introduced through a phenomenological phonon damping parameter, providing a more realistic linewidth to the phonon sidebands and regulating the extent of Franck-Condon displacement. In the case of undamped optical phonons the lineshape comprises of distinct peaks separated by an optical phonon frequency with the linewidth of a sideband determined solely by the reduced transition matrix element and the electromagnetic density of states at the corresponding sideband frequency. When phonon damping is introduced, the sidebands are broadened and a realistic picture is obtained. This additional broadening is governed by the lifetime of optical phonons. Undamped acoustic phonons produce a Gaussian spectral function with a sharp peak for the zero-phonon line. For a thermal reservoir of damped acoustic phonons the zero-phonon line is broadened, resulting in a Lorentzian structure in the vicinity of the atomic transition frequency and Gaussian wings away from the transition.

The polarization dynamics of the quantum dot was studied using the temporal dipolar autocorrelation function. The autocorrelation function was evaluated using the mean-field factorization described earlier. For a phonon reservoir consisting of undamped acoustic phonons coupled linearly to the quantum dot population operator (Independent Boson model), the role of phonons was limited to pure dephasing. In an unstructured electromagnetic reservoir, the phonons cause a rapid initial partial dephasing on picosecond timescales as the polaronic cloud forms. On longer timescales, the polarization decays exponentially to zero through electromagnetic dephasing. We then generalized the system to include non-radiative decay processes and damped phonons. Direct nonradiative decay is beyond the framework of our starting Hamiltonian and corresponds to the direct coupling of phonon operators to the quantum dot dipole. Polarization decay beyond pure dephasing was obtained using a semi-phenomenological model for non-radiative relaxation. Atomic polarization decays exponentially to zero at a faster rate involving the sum of nonradiative decay rate, the phonon damping rate and the electromagnetic de-
phasing rate. The overall rate of decay increases with temperature and the nonradiative coupling strength.

In order to describe population dynamics of an excited quantum dot, we considered Heisenberg equations of motion for the relevant atomic operators. The hierarchy of equations of motion was closed using a mean-field factorization of the atomic and lattice operators and a Born approximation that collapses the resulting two-time nonequilibrium atomic dipole correlation function to the atomic population at single time. The population decays exponentially to zero in an unstructured electromagnetic reservoir and the overall decay rate is determined by the sum of the decay rate from each phonon sideband. In the absence of nonradiative decay and phonon damping, the long time excited state population in a photonic crystal may be non-zero for negative detunings from the photonic band edge leading to the "fractionalized" steady state. In the case of undamped optical and acoustic phonons, the fractionalized steady state population is artificially increased with increasing dot-phonon coupling strength and increasing temperature because of the polaron shift between the excited and ground states of the quantum dot. For acoustic phonons there was substantial dephasing of the atomic transition dipole moment with increasing temperatures, contributing to residual population in the excited state. However, in the presence of nonradiative decay and phonon damping, there is no fractionalized steady state and the population decays to zero.

We also considered a generalization of the equilibrium description (in terms of the thermodynamic Green’s functions) to a more general non-equilibrium description in terms of generalized-Laplace transforms. Experimental and technological progress in quantum optics have brought to the forefront novel systems which cannot be treated in a simple manner using traditional formalism found in textbooks. For instance, interactions of multiple atoms or multilevel systems in structured electromagnetic reservoirs in the presence of nonradiative degrees of freedom is a formidable analytical problem. The formal elegance of this technique along with its underlying simplicity in performing robust analytical computations should make it an useful tool to study quantum optics in these complicated systems.

To summarize we have presented a detailed discussion of the role of nonradiative degrees of freedom on optical properties of a single quantum dot placed inside the structured reservoir of a photonic crystal. We have developed analytical techniques to study the optical lineshape, polarization decay and population dynamics of self-assembled quantum dots along with results specific to different phonon reservoirs. These results should be
of importance in practical applications of quantum dots in structured electromagnetic reservoirs. The formalism developed in this thesis can also be readily applied to more complicated problems involving multiple quantum dots in structured reservoirs. Interacting quantum dots that are coupled to a common electromagnetic reservoir and correlated or uncorrelated phonon reservoirs are experimental systems of interest for quantum information processing. Suitably superposed quantum states of atoms (or qubits) are central objects for quantum information processing which has been shown to be capable of exponentially speeding up certain classical algorithms. The workhorse of any quantum information processing is the entanglement due to the superposition of quantum states. Robust entanglement is essential for quantum information processing but the interactions of qubits with the environment degrees of freedom are a primary source of decoherence[71][52]. In recent years semiconductor quantum dots have emerged as novel systems for quantum information processing. The interaction of the two dots with a common electromagnetic reservoir introduces an effective coupling between the two dots. It was first pointed out by Dicke[70] that the lifetimes of the atoms are changed if they are separated by a distance which is less than the wavelength of the photon emitted in a transition from the excited state to the ground state due to strong dipole-dipole interaction induced between the two dots. The initial state of the two quantum dot system can be chosen such that they are either in a symmetric or antisymmetric superposition with respect to the interchange of the dots. The symmetric superposition demonstrates superradiant dynamics. On the other hand, the antisymmetric state exhibits subradiant behaviour which is easily explained in terms of resonant dipole-dipole interaction. Assuming one of the dots is in an initially excited state, it can subsequently de-excite and the emitted photon can be absorbed by the second atom. The subsequent emission of the photon by the second atom can re-excite the first atom leading to an effective and coherent interaction between the two dots. Two entangled quantum dots placed in close proximity inside a PBG material should be of practical interest as a qubit for quantum computation. In the structured reservoir of a photonic crystal, the simple picture presented above is conditioned by strong atom-photon interaction and the proximity of the resonant frequency of the atoms to the band edge. They also depend strongly on the phonon reservoirs coupled to the two quantum dots. These effects are easy to study by a suitable generalization of the methods presented in this thesis.

The methodology developed in this thesis can also be applied to systems involving more than two energy levels. Excitonic and biexcitonic levels in semiconductor quantum
dots are effective multilevel systems of relevance in practical applications. Another useful example of a multilevel system is an impurity atom such as $Er^{3+}$ which can be doped in an photonic band gap material using ion beam implantation techniques. $Er^{3+}$ exhibits sharp free-atom like spectrum especially at low temperatures. Intense photoluminescence at 1.54 $\mu m$ is observed at temperatures below 80$K$. These systems can be modelled as three-level atoms coupled to suitable dephasing reservoirs and interacting with the electromagnetic modes through allowed dipole transitions. Three-level atomic systems interacting with radiation modes has been exhaustively studied in the literature (see [15, 16] and references therein) and can give rise to novel physical phenomenon such as electromagnetically-induced transparency. Using the quantum coherence of the three-level atom for quantum information is also possible if practical schemes are devised to control decoherence. One possible methodology is based on coherent control where quantum interference between multiple excitation pathways is used to tailor the system dynamics. Another approach is to use externally imposed coherence (such as driving the atomic system by a CW laser) to mitigate various dephasing effects. The formalism presented in this thesis enables a detailed microscopic description of these processes.
Appendix A

Equilibrium Green’s function properties

In this appendix we show that \( G(\hat{\sigma}_e; \hat{\sigma}_g) \) and \( G(\hat{\sigma}_g; \hat{\sigma}_e) \) is zero. It is easy to see that \( G(\hat{\sigma}_e; \hat{\sigma}_g) \approx \langle \hat{C}_+(t)\hat{C}_+(0) \rangle_{HF} G_{MF}(\hat{\sigma}_e; \hat{\sigma}_g) \) in the mean-field approximation. Similarly \( G(\hat{\sigma}_g; \hat{\sigma}_e) \approx \langle \hat{C}_-(t)\hat{C}_-(0) \rangle_{HF} G_{MF}(\hat{\sigma}_g; \hat{\sigma}_e) \). The equation of motion for \( G_{MF}(\hat{\sigma}_e, \hat{\sigma}_g) \) is given by

\[
\omega G_{MF}(\hat{\sigma}_e, \hat{\sigma}_g) = G_{MF}([\hat{\sigma}_e, H_{MF}], \hat{\sigma}_g) \tag{A.1}
\]

The commutator \([\hat{\sigma}_e, H_{MF}]\) is easily evaluated to obtain

\[
[\hat{\sigma}_e, H_{MF}] = - (\omega_0 - \Delta) \hat{\sigma}_e + \sum_k \hat{\lambda}_k^* \hat{a}_k^\dagger \hat{\sigma}_{ee} - \sum_k \hat{\lambda}_k^* \hat{a}_k^\dagger \hat{\sigma}_{gg} \tag{A.2}
\]

The equation of motion for \( G_{MF}(\hat{\sigma}_e, \hat{\sigma}_g) \) reduces to

\[
\omega G_{MF}(\hat{\sigma}_e, \hat{\sigma}_g) = - (\omega_0 - \Delta) G_{MF}(\hat{\sigma}_e, \hat{\sigma}_g) + \sum_k \hat{\lambda}_k^* G_{MF}(\hat{a}_k^\dagger \hat{\sigma}_{ee}, \hat{\sigma}_g) - \sum_k \hat{\lambda}_k^* G_{MF}(\hat{a}_k^\dagger \hat{\sigma}_{gg}, \hat{\sigma}_e) \tag{A.3}
\]

We now compute the equation of motion of \( G_{MF}(\hat{a}_k^\dagger \hat{\sigma}_{ee}, \hat{\sigma}_g) \). It is given by

\[
(\omega + \omega_k) G_{MF}(\hat{a}_k^\dagger \hat{\sigma}_{ee}, \hat{\sigma}_g) = \sum_q \hat{\lambda}_q G_{MF}(\hat{a}_k^\dagger \hat{a}_q \hat{\sigma}_{ee}, \hat{\sigma}_g) - \sum_q \hat{\lambda}_q^* G_{MF}(\hat{a}_k^\dagger \hat{a}_q \hat{\sigma}_{ge}, \hat{\sigma}_g) \tag{A.4}
\]
Using the decoupling procedure, we find $G^{MF}_\omega(\hat{a}_k^\dagger \hat{a}_q \hat{\sigma}_{eg}, \hat{\sigma}_{ge}) \simeq \delta_{kq} \langle \hat{a}_k^\dagger \hat{a}_k \rangle G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{ge}) \simeq 0$ and $G^{MF}_\omega(\hat{a}_q^\dagger \hat{a}_q \hat{\sigma}_{ge}, \hat{\sigma}_{eg}) \simeq \delta_{kq} \langle \hat{a}_k^\dagger \hat{a}_k \rangle G^{MF}_\omega(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) \simeq 0$ and conclude that $G^{MF}_\omega(\hat{a}_k^\dagger \hat{\sigma}_{ee}, \hat{\sigma}_{eg}) \simeq 0$. The equation of motion for $G^{MF}_\omega(\hat{a}_k^\dagger \hat{\sigma}_{gg}, \hat{\sigma}_{eg})$ is given by

$$ (\omega + \omega_k)G^{MF}_\omega(\hat{a}_k^\dagger \hat{\sigma}_{gg}, \hat{\sigma}_{eg}) = - \sum_q \tilde{\lambda}_q G^{MF}_\omega(\hat{a}_q^\dagger \hat{a}_k^\dagger \hat{\sigma}_{eg}, \hat{\sigma}_{eg}) + \sum_q \tilde{\lambda}_q G^{MF}_\omega(\hat{a}_k^\dagger \hat{a}_q^\dagger \hat{\sigma}_{ge}, \hat{\sigma}_{eg}) \quad (A.5) $$

Decoupling the two-level system and photon operators, we find $G^{MF}_\omega(\hat{a}_q^\dagger \hat{a}_k^\dagger \hat{\sigma}_{eg}, \hat{\sigma}_{eg}) \simeq \delta_{kq}(1 + (\hat{a}_k^\dagger \hat{a}_k))G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg}) \simeq G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg})$ and $G^{MF}_\omega(\hat{a}_k^\dagger \hat{a}_q^\dagger \hat{\sigma}_{ge}, \hat{\sigma}_{eg}) \simeq \delta_{kq}(\hat{a}_k^\dagger \hat{a}_k)G^{MF}_\omega(\hat{\sigma}_{ge}, \hat{\sigma}_{eg}) \simeq 0$ and conclude that $G^{MF}_\omega(\hat{a}_k^\dagger \hat{\sigma}_{gg}, \hat{\sigma}_{eg}) \simeq \frac{\tilde{\lambda}_k}{\omega + \omega_k} G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg})$. Using this and the result following Eq(A.4), we solve for $G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{ge})$ and obtain

$$ \omega G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg}) = - (\omega_0 - \Delta) G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg}) + G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg}) \sum_k \frac{\tilde{\lambda}_k^2}{\omega + \omega_k} \quad (A.6) $$

which is valid for all $\omega$. This implies that $G^{MF}_\omega(\hat{\sigma}_{eg}, \hat{\sigma}_{eg}) = 0$. Using similar arguments one can show that $G^{MF}_\omega(\hat{\sigma}_{ge}, \hat{\sigma}_{ge}) = 0$. 


Appendix B

Method of generalized-Laplace transforms

In Chapter III we calculated the population dynamics of the two-level system coupled to
the phonon and photon reservoir using the Heisenberg equations of motion. We obtained
an integro-differential equation for the time evolution of $\langle \hat{\sigma}_{ee}(t) \rangle$ which was then solved
in the Born approximation. The purpose of this Appendix is twofold. First, following the
work of Pike and Swain[142], we discuss a generalization of the thermodynamic Green’s
function (the method of generalized Laplace-transforms) which can be used to study
non-equilibrium problems such as population decay (the density matrix at $t = 0$ is not
canonical). We then we use this technique to provide an alternate derivation of the
result obtained for population decay dynamics derived previously using the Heisenberg
equations of motion Eq(3.47). The inherent simplicity of the formalism provides clear
explanation of the various approximations made.

B.1 Introduction

The population decay of an initially excited two-level atom is an important non-equilibrium
problem. The atom, at $t = 0$, is assumed to be in the excited state and its den-
sity matrix ceases to be of the canonical form ($\hat{\rho} \neq \exp(-\beta H)/Z$) and the method
of thermodynamic Green’s function cannot be used. The decay dynamics of the two-
level atom can be obtained by defining a formal non-equilibrium correlation function
$L(\hat{\sigma}_{eg}(t), \hat{\sigma}_{ge}(t')) = \langle \hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle$ (the trace is over an arbitrary density matrix) and
considering the equal-time limit $t \rightarrow t'$ of $L(\hat{\sigma}_{eg}(t), \hat{\sigma}_{ge}(t'))$. The steady state (non-
equilibrium) population is given by the limit \( t, t' \to \infty \) of \( L(\hat{\sigma}_{eg}(t), \hat{\sigma}_{ge}(t')) \).

A general non-equilibrium correlation function, denoted \( L(\hat{A}(t), \hat{B}(0)) = \langle \hat{A}(t)\hat{B}(0) \rangle \), can be defined in Fourier representation as

\[
L_\omega(\hat{A}, \hat{B}) = -i \int_0^\infty dt L(\hat{A}(t), \hat{B}(0)) \exp(i\omega t)
\]

for an arbitrary density matrix \( \hat{\rho} \). It has a spectral representation

\[
L_\omega(\hat{A}, \hat{B}) = \sum_{n,m} \frac{\hat{A}_{nm}(\hat{B}\hat{\rho})_{mn}}{\omega + E_{nm}}
\]

and satisfies an equation of motion

\[
\omega L_\omega(\hat{A}, \hat{B}) = \langle \hat{A}\hat{B} \rangle_{NE} + L_\omega([\hat{A}, H], \hat{B})
\]

where \( \langle \hat{A}\hat{B} \rangle_{NE} \) is the non-equilibrium initial condition at \( t = t' \). This technique can also be easily applied to calculate the time evolution of a single operator \( \hat{A} \) by defining a function \( L_\omega(\hat{A}) \). The properties of \( L_\omega(\hat{A}) \) can be obtained from the above equations by replacing \( \hat{B} \) with unity. Its equation of motion is given by

\[
\omega L_\omega(\hat{A}) = \langle \hat{A} \rangle_{NE} + L_\omega([\hat{A}, H])
\]

A very clear discussion of the differences between the generalized Laplace-transform technique and the thermodynamic Green’s functions is given in Pike and Swain[142].

### B.2 Population dynamics of a single two-level atom

We now calculate the time evolution of the population of the excited two-level atom using the method of generalized Laplace-transforms. The probability of the atom being excited at time \( t \) is obtained by calculating \( L(\hat{\sigma}_{ee}(t)) \). We calculate the equation of motion of \( L(\hat{\sigma}_{ee}(t)) \) for the polaron transformed Hamiltonian Eq(2.36). The polaron transformation modifies the dot-photon coupling term by introducing the phonon-displacement operators. Consequently, any radiative transition is now accompanied by a ”shower” of phonons[136] resulting from this coherent phonon state. To lowest order in the two-level
Appendix B. Method of generalized-Laplace transforms

Atom photon coupling constant, the self-energy contributions are:

\[ \Sigma^+ (t_1 - t_2) = \sum_k \langle \hat{C}_+(t_1) \hat{C}_-(t_2) \rangle |\lambda_k|^2 e^{i(\omega_0 - \Delta - \omega_k)(t_1 - t_2)} \]  \hspace{1cm} (B.5)

\[ \Sigma^- (t_1 - t_2) = \sum_k \langle \hat{C}_+(t_2) \hat{C}_-(t_1) \rangle |\lambda_k|^2 e^{-i(\omega_0 - \Delta - \omega_k)(t_1 - t_2)} \]

Using the expansion \( \langle \hat{C}_+(t_1) \hat{C}_-(t_2) \rangle = \sum_{n=-\infty}^{\infty} L_n e^{-i\Omega_0 (t_1 - t_2)} \) and \( \langle \hat{C}_+(t_2) \hat{C}_-(t_1) \rangle = \sum_{n=-\infty}^{\infty} L_n e^{i\Omega_0 (t_1 - t_2)} \) for optical phonons, we find that in \( \omega \)-space the self energies can be written as:

\[ \Sigma^\pm (\omega) = \sum_{n=-\infty}^{\infty} L_n \sum_k \frac{|\lambda_k|^2}{\omega \pm (\omega_0 - \Delta - n\Omega_0 - \omega_k)} \]  \hspace{1cm} (B.6)

which are exactly of the same form as in the Fourier-transformed representation of Eq(3.47). We use the following prescription to calculate \( L_\omega (\hat{\sigma}_{eg} \hat{\sigma}_{ge}) \). As a first step we calculate \( L_\omega (\hat{\sigma}_{eg} \hat{\sigma}_{ge}) \) by ignoring the renormalization of the two-level photon coupling by the phonon displacement operators \( \hat{C}_\pm \). The self energy contribution is calculated to order \( \lambda_k^2 \) in the two-level photon coupling constant. The modification of the self-energy due to the phonon displacement operators \( \hat{C}_\pm \) is included by convolving the self-energy due to pure radiative coupling by the finite-temperature phonon correlation function \( \langle \hat{C}_\pm (t_1) \hat{C}_\pm (t_2) \rangle \). This is done very simply by the ansatz:

\[ \sum_k \frac{|\lambda_k|^2}{\omega \pm (\omega_0 - \Delta - \omega_k)} \rightarrow \sum_{n=-\infty}^{\infty} L_n \sum_k \frac{|\lambda_k|^2}{\omega \pm (\omega_0 - \Delta - n\Omega_0 - \omega_k)} \]  \hspace{1cm} (B.7)

in the presence of phonon coupling. For a phonon reservoir consisting of undamped acoustic phonons (see Sec VI.A) we have the ansatz:

\[ \sum_k \frac{|\lambda_k|^2}{\omega \pm (\omega_0 - \Delta - \omega_k)} \rightarrow e^{-\Phi(0)} \sum_k \frac{|\lambda_k|^2}{\omega \pm (\omega_0 - \Delta - \omega_k)} \]  \hspace{1cm} (B.8)

At the first stage we ignore the renormalization of the dot-photon coupling in the interaction Hamiltonian by the phonon displacement operators. The interaction part of Eq(2.36) is written as

\[ H_I = \sum_k (\lambda_k \hat{\sigma}_{eg} \hat{\alpha}_k + \lambda_k^* \hat{\alpha}_k^\dagger \hat{\sigma}_{ge}) \]  \hspace{1cm} (B.9)

where we have suppressed the phonon displacement operators. It is easy to see that
\(L_\omega(\hat{\sigma}_{eg}\hat{\sigma}_{ge})\) obeys an equation of motion given by

\[
\omega L_\omega(\hat{\sigma}_{eg}\hat{\sigma}_{ge}) = \langle \hat{\sigma}_{eg}\hat{\sigma}_{ge} \rangle_{NE} + \sum_k \lambda_k L_\omega(\hat{\sigma}_{eg}\hat{a}_k) - \sum_k \lambda_k^* L_\omega(\hat{a}_k^\dagger \hat{\sigma}_{ge})
\]

The non-equilibrium initial condition is chosen such that the atom is excited at \(t = 0\) which implies that \(\langle \hat{\sigma}_{eg} \hat{\sigma}_{ge} \rangle_{NE} = 1\). We now consider the equations of motion for \(L_\omega(\hat{\sigma}_{eg}\hat{a}_k)\) and \(L_\omega(\hat{a}_k^\dagger \hat{\sigma}_{ge})\). They are easily obtained and are given by:

\[
(\omega + (\omega_0 - \Delta - \omega_k))L_\omega(\hat{\sigma}_{eg}\hat{a}_k) = \sum_q \lambda_q^* L_\omega(\hat{\sigma}_{ee}\hat{a}_k \hat{a}_q^\dagger) - \sum_q \lambda_q L_\omega(\hat{\sigma}_{gg}\hat{a}_q^\dagger \hat{a}_k)
\]

\[
(\omega - (\omega_0 - \Delta - \omega_k))L_\omega(\hat{a}_k^\dagger \hat{\sigma}_{ge}) = -\sum_q \lambda_q L_\omega(\hat{\sigma}_{ee}\hat{a}_q \hat{a}_k^\dagger) + \sum_q \lambda_q L_\omega(\hat{\sigma}_{gg}\hat{a}_q^\dagger \hat{a}_k)\]

where we have used the non-equilibrium initial conditions: \(\langle \hat{\sigma}_{eg} \hat{a}_k \rangle_{NE} = \langle \hat{a}_k^\dagger \hat{\sigma}_{ge} \rangle_{NE} = 0\).

Using an approximate decoupling scheme of replacing the photon operators by their thermal averages obtained in the absence of two-level photon coupling, we simplify:

\[
L_\omega(\hat{\sigma}_{ee}\hat{a}_k \hat{a}_q^\dagger) \simeq \delta_{kq} (1 + \langle \hat{a}_k^\dagger \hat{a}_k \rangle) L_\omega(\hat{\sigma}_{ee}) \simeq L_\omega(\hat{\sigma}_{ee})
\]

\[
L_\omega(\hat{a}_k^\dagger \hat{\sigma}_{ge}) \simeq \delta_{kq} \langle \hat{a}_k^\dagger \hat{a}_k \rangle L_\omega(\hat{\sigma}_{gg}) \simeq 0
\]

where we have used the fact that in thermal equilibrium \(\langle \hat{a}_k^\dagger \hat{a}_k \rangle \simeq 0\). Using Eqs(B.11) and (B.12) in Eq(B.10) we obtain a closed equation for the \(L_\omega(\hat{\sigma}_{eg}\hat{\sigma}_{ge})\) given by

\[
L_\omega(\hat{\sigma}_{eg}\hat{\sigma}_{ge}) = \frac{\langle \hat{\sigma}_{eg}\hat{\sigma}_{ge} \rangle_{NE}}{\omega - \sum_k \frac{|\lambda_k|^2}{\omega - (\omega_0 - \Delta - \omega_k)} - \sum_k \frac{|\lambda_k|^2}{\omega + (\omega_0 - \Delta - \omega_k)}}
\]

Using the ansatz Eq(B.7) and the prescription outlined above to include the effects of phonons, we obtain the full solution in the presence of phonons as:

\[
L_\omega(\hat{\sigma}_{eg}\hat{\sigma}_{ge}) = \frac{\langle \hat{\sigma}_{eg}\hat{\sigma}_{ge} \rangle_{NE}}{\omega - \sum_n L_n \sum_k \frac{|\lambda_k|^2}{\omega - (\omega_0 - \Delta - m_l \omega_k)} - \sum_n L_n \sum_k \frac{|\lambda_k|^2}{\omega + (\omega_0 - \Delta - m_l \omega_k)}}
\]

Now using the fact that the nonequilibrium initial condition is \(\langle \hat{\sigma}_{eg}\hat{\sigma}_{ge} \rangle_{NE} = 1\) we find that this is exactly the same result Eq(3.47) as derived using the Heisenberg equations of motion method.

In this appendix we discussed a useful generalization of the method of thermody-
Appendix B. Method of generalized-Laplace transforms

Dynamical Green’s functions which is only applicable when the two-level atom is in thermal equilibrium. The method of generalized-Laplace transforms can be used in cases where the initial density matrix is non-canonical and the two-level atom is out of thermal equilibrium. We illustrated the method by applying it to study the population dynamics of a two-level atom coupled to a thermal reservoir of phonons and a general electromagnetic reservoir. The hierarchy of equations of motion generated for the various higher-order generalized-Laplace transforms were closed by an approximate decoupling of the atomic and radiative degrees of freedom. The decoupling was accomplished by replacing the photon operators with their thermal expectation values evaluated in the absence of any quantum-dot photon interaction. The Born approximation was employed to include processes up to second order in the dot-photon coupling constant. However, the Markov approximation was not made rendering the result valid for a general electromagnetic reservoir. The results obtained using the method of generalized-Laplace transforms was shown to be equivalent to those obtained using the Heisenberg equations of motion in Chapter III. The Heisenberg equations of motion become extremely cumbersome for multiple atoms or multilevel atomic systems. The method of generalized-Laplace transforms, on the other hand, provides us with an intuitive tool to obtain closed expressions for relevant quantum optical physical quantities with a well-defined methodology. The simplicity of the method of generalized-Laplace transforms lies in its ability to truncate the hierarchy of equations of motion in Fourier-space using a consistent set of approximations as illustrated above. This is difficult to accomplish using the Heisenberg equations of motion for more complicated systems.
Appendix C

Modified phonon correlation functions

In this appendix we provide a proof of $\langle \hat{C}_+(t)\hat{C}_-(t') \rangle = e^{-\langle \hat{\Phi}(0) - \hat{\Phi}(t-t') \rangle} e^{-\gamma_{\text{nonrad}}(t-t')}$ in the presence of direct nonradiative decay and $\langle \hat{C}_+(t)\hat{C}_-(t') \rangle = e^{-\langle \hat{\Phi}(0) - \hat{\Phi}(t-t') \rangle} e^{-\Gamma_{\text{damp}}(t-t')}$. Consider the Spin–Boson Hamiltonian written as

$$H = (\omega_0 + \hat{V})\hat{\sigma}_{ee} + H_p$$

(C.1a)

$$\hat{V} = \sum_q \eta_q (\hat{c}_q + \hat{c}^\dagger_q)$$

(C.1b)

The proof relies on the equivalence of thermal averages $\langle \hat{C}_+(t)\hat{C}_-(0) \rangle \equiv e^{-i\Delta t} \langle \hat{U}(t) \rangle$ where $\Delta$ is defined in Eq(2.37b), $\hat{U}(t) = e^{iH_p t} e^{-i (H_p + \hat{V}) t}$ and the trace is over the phonon degrees of freedom:

$$\langle \hat{U}(t) \rangle = \langle e^{iH_p t} e^{-i \sum_q \Omega_q (\hat{c}_q^\dagger + \hat{c}_q)^2} (\hat{c}_q + \hat{c}_q^\dagger) \rangle e^{i\Delta t}$$

(C.2)

$$= e^{i\Delta t} \langle e^{iH_p t} \hat{C}_- e^{-iH_p t} \hat{C}_+ \rangle$$

$$= e^{i\Delta t} \langle \hat{C}_-(t)\hat{C}_+(0) \rangle$$

where we have used the displacement properties of the operators $\hat{C}_\pm$:

$$\hat{C}_- f(\hat{c}_q, \hat{c}_q^\dagger) \hat{C}_+ = f(\hat{c}_q + \frac{\eta_q}{\Omega_q}, \hat{c}_q^\dagger + \frac{\eta_q}{\Omega_q})$$

(C.3)
Appendix C. Modified phonon correlation functions

valid for any arbitrary function \( f \). We also observe from Eq(3.10a) that \( \langle \hat{C}_-(t)\hat{C}_+(0) \rangle = \langle \hat{C}_+(t)\hat{C}_-(0) \rangle \)\[152]. Hence we conclude \( \langle \hat{C}_+(t)\hat{C}_-(0) \rangle \equiv e^{-i\Delta t}\langle e^{iH_p t}e^{-i(H_p+\hat{V})t} \rangle \) using Eq(C.2).

Now define a unitary operator \( \hat{U}(t) \):

\[
\hat{U}(t) = e^{iH_p t}e^{-i(H_p+\hat{V})t}
\]

such that \( \langle \hat{C}_+(t)\hat{C}_-(0) \rangle \equiv e^{-i\Delta t}\langle \hat{U}(t) \rangle \) and note that \( \hat{U}(0) = 1 \). It is easy to see that \( \hat{U}(t) \) obeys an equation of motion:

\[
\frac{\partial \hat{U}(t)}{\partial t} = e^{iH_p t}iH_p e^{-i(H_p+\hat{V})t} - e^{iH_p t}i(H_p + \hat{V})e^{-i(H_p+\hat{V})t}
\]

\[
= -e^{iH_p t}i\hat{V}e^{-i(H_p+\hat{V})t}
\]

\[
= -e^{iH_p t}i\hat{V}e^{-iH_p t}e^{iH_p t}e^{-i(H_p+\hat{V})t}
\]

\[
= -i\hat{V}(t)\hat{U}(t)
\]

which can be formally integrated and rewritten as:

\[
\hat{U}(t) = 1 - i \int_0^t dt_1 \hat{V}(t_1)\hat{U}(t_1)
\]

The solution can be written in the form of a Dyson series:

\[
\hat{U}(t) = \sum_{n=0}^{\infty} (-i)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} dt_n \hat{V}(t_1)\hat{V}(t_2)\cdots\hat{V}(t_n)
\]

The Dyson series can be compactly summed using a ”Time-Ordering” operator \( \hat{T} \):

\[
\hat{U}(t) = \hat{T} \exp(-i \int_0^t dt_1 \hat{V}(t_1))
\]

The evaluation of \( \langle \hat{U}(t) \rangle \) is most conveniently done using the ”Linked-Cluster” expansion as\[130]:

\[
\langle \hat{U}(t) \rangle = \sum_{n=0}^{\infty} (-i)^n U_n(t)
\]

\[
U_n(t) = \frac{1}{n!} \int_0^t dt_1 \cdots \int_0^t dt_n \langle \hat{T} \hat{V}(t_1)\hat{V}(t_2)\cdots\hat{V}(t_n) \rangle
\]

Since \( \hat{V}(t) \) describes the creation or destruction of a phonon, the \( \hat{V} \) operators always
exist in pairs. This means that only terms with even \( n \) contribute. Eq(C.9) can then be rewritten as:

\[
\langle \hat{U}(t) \rangle = \sum_{m=0}^{\infty} (-1)^m U_{2m}(t)
\]

\[
U_{2m}(t) = \frac{1}{(2m)!} \int_0^t dt_1 \cdots \int_0^t dt_{2m} \langle \hat{T}\hat{V}(t_1) \cdots \hat{V}(t_{2m}) \rangle
\]

According to Wick’s theorem the \( \hat{V} \) operators pair up to form phonon correlation function \( D(t_1 - t_2) \) as defined in Eq(5.10). Thus the \( m^{th} \) term has \( m \) phonon correlation functions:

\[
\langle \hat{T}\hat{V}(t_1) \cdots \hat{V}(t_{2m}) \rangle = i^m \sum_{\text{all combinations}} D(t_i - t_j) D(t_2 - t_s) D(t_u - t_{2m})
\]

Each phonon correlation function depends on two time variables \( D(t_i - t_j) \). The time integral over those two variables defines a function \( \phi(t) \):

\[
\phi(t) = i \int_0^t dt_1 \int_0^t dt_2 D(t_1 - t_2)
\]

The number of all such possible combinations is found[130] to be \( \frac{(2m)!}{2^m m!} \). Hence we obtain the result:

\[
U_{2m}(t) = \frac{1}{m!} \left( \frac{\phi(t)}{2} \right)^m
\]

which is summed to obtain:

\[
\langle \hat{U}(t) \rangle = \exp\left[ e^{-\frac{i}{2} \int_0^t dt_1 \int_0^t dt_2 D(t_1 - t_2)} \right]
\]

In the presence of an additional nonradiative decay term \( \gamma_{\text{nonrad}} \) we use the modified phonon correlation function

\[
D(t_1 - t_2) = -i \sum_q n_q^2 \left[ (N_q + 1) e^{-\omega_q t_1 - \omega_q t_2} + N_q e^{\omega_q (t_2 - t_1)} \right]
\]

in Eq(C.14) and obtain \( \langle \hat{C}_+(t)\hat{C}_-(t') \rangle = e^{-(\Phi(0) - \Phi(t_2 - t_1'))} e^{-\gamma_{\text{nonrad}} \tau_{t_1 - t_2}} \). In the presence of phonon damping we use the modified phonon correlation function Eq(5.37) in Eq(C.14) and obtain \( \langle \hat{C}_+(t)\hat{C}_-(t') \rangle = e^{-(\Phi(0) - \Phi(t_2 - t_1'))} e^{-\gamma_{\text{damp}} \tau_{t_1 - t_2}} \).
Bibliography


