UNIVERSITY OF HYDERABAD

DOCTORAL THESIS

Effects of multimode light-matter coupling on semiclassical and quantum Rabi oscillations of a two-level system

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A thesis submitted in partial fulfilment of the requirements for the award of the degree of
Doctor of Philosophy** in **Physics
by

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February 7, 2022

DECLARATION

I hereby declare that the matter embodied in this thesis entitled "**Effects of multimode light-matter coupling on semiclassical and quantum Rabi oscillations of a two-level system**" is an outcome of the theoretical research carried out by me at the School of Physics, University of Hyderabad, Hyderabad, India under the supervision of **Dr. Shyamal Biswas**.

I also declare that this thesis has not been submitted previously in part or in full to any other University or Institution for the award of any degree or diploma.

I hereby agree that my thesis can be deposited in the institutional repository.

I further declare that this thesis is based on my original work done under the supervision of Dr. Shyamal Biswas. I have cited all the relevant sources from which ideas and results have been taken. The Library report on the originality is enclosed in the end of the thesis.

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Date: February 7, 2022

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CERTIFICATE

This is to certify that the thesis entitled "**Effects of multimode light-matter coupling on semiclassical and quantum Rabi oscillations of a two-level system**" submitted by **Mr. Najirul Islam** (Reg. No. 17PHPH21) in partial fulfilment of the requirements for the award of Doctor of Philosophy in Physics is a bonafide work carried out by him under the direct supervision of Dr. Shyamal Biswas at the School of Physics, University of Hyderabad, Hyderabad, India.

This thesis is free from plagiarism and has not been submitted previously in part or in full to any other University or Institution for the award of any degree or diploma.

Further, the scholar (Mr. Najirul Islam) got the following research publications, which are related to the work of the thesis, before its submission for adjudication.

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3	PY803	Advanced Experimental Techniques	4	Pass
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Dedicated to my

Mother

List of Abbreviations

J-C	Jaynes-Cummings
et al	and others
K	Kelvin
$H.c.$	Hermitian Conjugate
m	Mass of a particle
$\vec{r}$	Position of a particle
t	Time
i	$\sqrt{-1}$
$\vec{d}$	Electric dipole moment
$\vec{\mu}$	Magnetic dipole moment
λ	Wavelength
Q	Quality factor
ω_0	Bohr frequency
Ω_R	Rabi frequency
$R_{2\to 1}(t)$	Rate of stimulated emission
$R_{1\to 2}(t)$	Rate of (stimulated) absorption
T	Temperature

Physical Constants

Charge of electron (e)	$1.60217662 \times 10^{-19}$ C
Bohr radius (a_0)	$5.29177210903(80) \times 10^{-11}$ m
Reduced Planck constant or Dirac constant ($\hbar$)	$1.0545718 \times 10^{-34}$ $m^2.kg.s^{-1}$
Speed of light (c)	299792458 $m.s^{-1}$
Free space permittivity (ϵ_0)	$8.8541878128(13) \times 10^{-12}$ $F.m^{-1}$
Boltzmann constant (k_B)	$1.38064852 \times 10^{-23}$ $m^2.kg.s^{-2}.K^{-1}$

Abstract

The thesis entitled "*Effects of multimode light-matter coupling on semiclassical and quantum Rabi oscillations of a two-level system*" presents the work done by us during the last four years and six months. The thesis consists of six chapters viz., [i] *introduction*, [ii] *Rabi model result for the Einstein B coefficient*, [iii] *multimode Jaynes-Cummings model results for the Einstein A and B coefficients*, [iv] *multimode Jaynes-Cummings model results for the collapse and the revival of the quantum Rabi oscillations in a lossy resonant cavity*, [v] *population dynamics of two-level systems for the generalized Einstein coefficients*, [vi] *conclusions & future scope*. The main contents and the discussion pertaining to the contents are systematically presented chapter wise.

Chapter 1: Introduction

Here we begin with the motivation of writing the thesis. Then we have mentioned the key findings in the thesis and the significance of our theoretical results. Then we have described the system of interest i.e. a two-level interacting with the thermal radiation field or the coherent field or a monochromatic light. Then we have briefly described the existing models and theories necessary for the rest of the chapters of the thesis. We have briefly shown how Dirac determined the Einstein B coefficient within the first order perturbation theory of quantum mechanics. We also have briefly shown how Weisskopf-Wigner determined Einstein's A coefficient within the quantum theory of electrodynamics. We have introduced the population dynamics of the two-level systems with the help of Einstein rate equations. We have explicitly shown failure of the first order time-dependent perturbation theory of quantum mechanics. We have introduced Rabi model and Jaynes-Cummings model for the explanation of the Rabi oscillations of a two-level system both from the semiclassical point of view and the quantum mechanical point of view, respectively. We also have introduced the Purcell effect in this regard. We have introduced the collapse and revival of the quantum Rabi oscillations. Finally, we have mentioned chapter-wise organization of the thesis.

Chapter 2: Rabi model result for the Einstein B coefficient

This chapter contains Rabi model result for the Einstein B coefficient. The system of interest for this chapter is mainly a two-level system (atom/molecule) in the thermal radiation field. Starting from the Rabi Hamiltonian, which is useful in arriving at non-perturbative results within the rotating wave approximation, we have found Einstein's B coefficient to be time-dependent: $B(t) = B_0|J_0(\Omega_R t)|$ for a two-level system in thermal radiation field. Here B_0 is the original Einstein B coefficient, Ω_R is the Rabi flopping (angular) frequency of the two level system, and J_0 is the zeroth order Bessel function of the first kind. Here the light-matter interaction is treated classically but the two-level system is treated quantum mechanically, and our result

can be considered as a semiclassical result. We, of course, get back the original B coefficient in the limiting case of $\Omega_R \to 0$. We also have obtained Rabi model result for the Einstein B coefficient for a monochromatic light incident of the two-level system.

Chapter 3: Multimode Jaynes-Cummings model results for the Einstein A and B coefficients

Here we have generalized the Einstein A and B coefficients from quantum field theoretic point of view by bringing the fundamental processes and the quantum Rabi oscillation in a single footing for the light-matter interactions for nonzero Rabi frequency . We have analytically obtained multimode Jaynes-Cummings model results for the quantum Rabi oscillation of a two-level system in a lossy resonant cavity containing (i) thermal photons and (ii) injected photons of a coherent field . We have renormalized the coupling constant for the light-matter interactions for these cases. The net transition probability calculated for 'vacuum' Rabi oscillation of a two-level system in a lossy Resonant cavity matches well with the seminal experimental data obtained by Brune *et al* [Phys. Rev. Lett. **76**, 1800 (1996)]. The net transition probability calculated for the quantum Rabi oscillations of two-level system interacting with an injected coherent field in a lossy Resonant cavity also matches well with the seminal experimental data obtained by Brune *et al*.

Chapter 4: Multimode Jaynes-Cummings model results for the collapse and the revival of the quantum Rabi oscillations in a lossy resonant cavity

Here we have numerically obtained theoretical results for the collapse and the revival of the quantum Rabi oscillations for low average number of coherent photons injected on a two-level system in a lossy resonant cavity . We have adopted the multimode Jaynes-Cummings model for the same and especially treated the Ohmic losses from the cavity. We have compared our results with two sets of experimental data for low average number of coherent photons ($\bar{n} = 0.85$ and 1.77) incident on a two-level system in the lossy resonant cavity. Our results match reasonably well with the experimental data, at least, better than the theoretical one obtained for only the resonant mode and no loss from the cavity under consideration.

Chapter 5: Population dynamics of two-level systems for the generalized Einstein coefficients

Here we have studied population dynamics of two-level systems interacting with both the thermal radiation field and the monochromatic light. While the interactions of the two-level systems and the monochromatic light have been treated classically (with the Rabi model), the interactions of the two-level systems and the thermal radiation field have been treated both classically (with the Rabi model) and quantum mechanically (with the multimode Jaynes-Cummings model). For the semiclassical cases we already have obtained the generalized (time-dependent) Einstein B coefficient. For the quantum mechanical case too we already have obtained the generalized (time-dependent) Einstein A and B coefficients. We have studied the population dynamics for all these cases with the help of Einstein rate equations where the original Einstein coefficients are replaced by the generalized Einstein coefficients. The A coefficient is, of course, kept unaltered for the semiclassical cases. Time-dependence of the generalized Einstein coefficients opens a path to go beyond Pauli-von Neumann formalism of the non-equilibrium statistical mechanics. The population dynamics allows us to further study the entropy production of a level-system. For the semiclassical cases, we have shown that the Rabi oscillation can drive the two-level system away from the thermodynamic equilibrium. On the other hand, for the quantum mechanical case, we have shown that the Rabi oscillation of a small Rabi frequency (Ω_R) can not drive the two-level system away from the thermodynamic equilibrium. However, reaching the thermodynamic equilibrium is prolonged due to the quantum Rabi oscillations in the two-level system.

Chapter 6: Conclusions and future scope

Here we have briefed the conclusions of the Ph.D. works, especially the summaries. We also have mentioned the future scopes of the Ph.D. works.

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Chapter 1

Introduction

1.1 Motivation

The system of our interest is a two-level system (atom/molecule) in a thermal or coherent radiation field. Fundamental processes such as spontaneous emission, stimulated emission and absorption have been widely discussed for such a system. Einstein's A and B coefficients characterise these fundamental processes, in particular, the A coefficient is related to the spontaneous emission and the B coefficient is related to the stimulated emission or absorption for a two-level system in the thermal radiation field [1]. While Weisskopf-Wigner [2] determined Einstein's A coefficient within the quantum theory of electrodynamics, Dirac [3] determined Einstein's B coefficient within the first order time-dependent perturbation theory of quantum mechanics. Dirac determined the B coefficient from the well known transition probability [4]

$$P_{2\rightarrow 1}(t) \simeq \Omega_R^2 \frac{\sin^2([\omega-\omega_0]t/2)}{(\omega-\omega_0)^2} \tag{1.1}$$

where $\Omega_R = |\langle\psi_1|\vec{d}\cdot\vec{E}_0|\psi_2\rangle|/\hbar$ represents the light-matter coupling as well as the Rabi frequency of the two-level system, $\omega_0 = [E_2 - E_1]/\hbar$ is the Bohr frequency of the two-level system of energy eigenvalues E_2 and E_1 ($E_2 > E_1$), ω is the frequency of the sinusoidal electromagnetic wave ($\vec{E}_0\cos(\omega t)$) incident on the two-level system, and t is the time. It is considered for the above transition probability that the two-level system initially ($t = 0$) was at the upper level. For the validity of perturbation we have $\frac{\Omega_R}{|\omega-\omega_0|} \ll 1$ [4]. Calculation of the B coefficient needs consideration of the above transition probability for all possible frequencies of the thermal radiation field incident on the two-level system [4]. However, it is clear from the above expression that most of the transitions occur for frequencies at around the resonance ($\omega \rightarrow \omega_0$) [4].

Interestingly, there is a problem of normalization of the transition probability at the resonance ($\omega \rightarrow \omega_0$). The transition probability takes the form $P_{2\rightarrow 1}(t) \rightarrow \frac{\Omega_R^2 t^2}{4}$ at the resonance. This form of the transition probability is not normalizable rather $P_{2\rightarrow 1}(t) \rightarrow \infty$ for $t \rightarrow \infty$ for any finite small value of Ω_R for $\omega \rightarrow \omega_0$. Hence the 1^{st} order perturbation theory of the quantum mechanics fails at around the resonance even for a non-zero weak light-matter coupling of the sinusoidal perturbation. Since most of the contributions in the B coefficient come from around the resonance, the expression of the B coefficient determined by Dirac is questionable even for the weak coupling regime [4, 5]. Another drawback of the 1^{st} order perturbation theory is that the periodicity in stimulated transition is lost at the resonance $(\omega \to \omega_0)$. The periodicity can be found in non-pertubative model such as the Rabi model [6] which is able to explain the nuclear magnetic resonance.

Thus we are motivated to determine the B coefficient within a non-perturbative model, such as the Rabi model, which though is often used for strong coupling regime, can also be used for weak coupling regime as it is an exactly solvable model for the entire range of light-matter coupling.

The Rabi model, however, treats the light-matter interaction classically. The Jaynes-Cummings model, on the other hand, is another exactly solvable model for light-matter interactions. This is a quantum mechanical model where light-matter interactions are treated within the quantum field theory. We are also interested to determine the Einstein A and B coefficients within the multimode Jaynes-Cummings model.

There are experimental data available for the collapse and the revival of the quantum Rabi oscillations of a two-level system in a lossy resonant cavity. We are also motivated to explain these data within the multimode Jaynes-Cummings model.

We are also motivated to study the population dynamics of a two-level system in thermal radiation field with the model dependent Einstein coefficients.

The population dynamics allows us to study non-equilibrium statistical mechanics of the two-level system in thermal radiation field. We are motivated to determine entropy production of the system.

Let us now introduce various topics related to our findings.

1.2 Einstein's A and B coefficients

In the early 20^{th} century, within the era of the old quantum mechanics, Einstein proposed the ideas of spontaneous emission, stimulated emission and stimulated absorption rate coefficients which are commonly known as Einstein's A and B coefficients [1]. Spontaneous emission is a fundamental process undergone on a two-level system (atom/molecule) such that the system transits spontaneously from its excited state ($|\psi_2\rangle$) to its ground state ($|\psi_1\rangle$) and emits a photon having energy similar to the energy difference of the two states of the two-level system [2]. Vacuum fluctuations around the two-level system play a significant role for the spontaneous emission [2]. The Einstein A coefficient is defined as the rate of spontaneous emission from the excited state to the ground state of the two-level system [4, 7, 8].

Stimulated emission, on the other hand, is another fundamental process undergone on a two-level system such that the system transits from its excited state ($|\psi_2\rangle$) to its ground state ($|\psi_1\rangle$) once a photon incidents on it and the system emits a photon. The characteristics of the emitted photon in stimulated emission have phase, polarization, frequency and direction of propagation similar to the incident photon of the electromagnetic field [4]. These characteristics raise the possibility of the light amplification i.e. if there is a bottle of two-level systems (atoms/molecules), all in the excited state, and the bottle is triggered with a single photon, then a chain reaction may take place. The first photon would produce two photons, these two photons would produce four photons, and so on [4]. This is the principle behind the light amplification by stimulated emission of radiation (LASER) [4]. The Einstein B_{21} coefficient is defined as the rate of stimulated emission per unit energy density ($u(\omega_0)$) per unit frequency interval around the resonance and it is denoted as $B_{21} = R_{2\to1}/u(\omega_0)$ [4, 7, 8]. Here, by resonance, we mean the matching of the (angular) frequency of the incident photon (ω) and the Bohr frequency ($\omega_0 = [E_2 - E_1]/\hbar$) of the two-level system having energy E_2 in the state $|\psi_2\rangle$ and energy E_1 in the state $|\psi_1\rangle$.

Stimulated absorption (or simply the absorption) is another fundamental process undergone on a two-level system such that the system transits from its ground state to its excited state once a photon incidents on it and the photon is absorbed by the system [4]. The Einstein B_{12} coefficient is defined as the rate of stimulated absorption per unit energy density ($u(\omega_0)$) per unit frequency interval around the resonance and it is denoted as $B_{12} = R_{1\to2}/u(\omega_0)$ [4, 7, 8].

A schematic diagram for the fundamental processes related to the light-matter interactions for a two-level system are shown in figure 1.1.

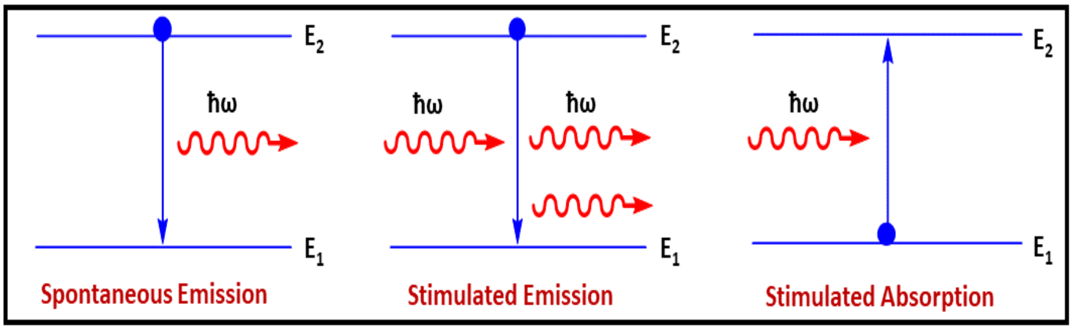

FIGURE 1.1: Schematic diagram for the fundamental processes undergone on a two-level system.

Einstein's rate equations for the occupation probabilities P_2 (for the excited state) and P_1 (for the ground state) of a two-level system in the thermal radiation field in a blackbody cavity are given by [1, 4]

$$\frac{\mathrm{d}P_2}{\mathrm{d}t} = -AP_2 - P_2B_{21}u(\omega_0) + P_1B_{12}u(\omega_0) \tag{1.2}$$

and

$$\frac{\mathrm{d}P_1}{\mathrm{d}t} = AP_2 + P_2B_{21}u(\omega_0) - P_1B_{12}u(\omega_0). \tag{1.3}$$

The occupation probabilities can take the form $P_i = N_i/N$ (where $(i = 1,2)$) where N_i is the occupation number of the two-level systems in the state $|i\rangle$ and N is the total number of the two-level systems in the thermal radiation field. A schematic diagram for the occupation of the two-level systems is shown in figure 1.2. We, of course, have the relation $P_1 + P_2 = 1$ for the conservation of the total probability.

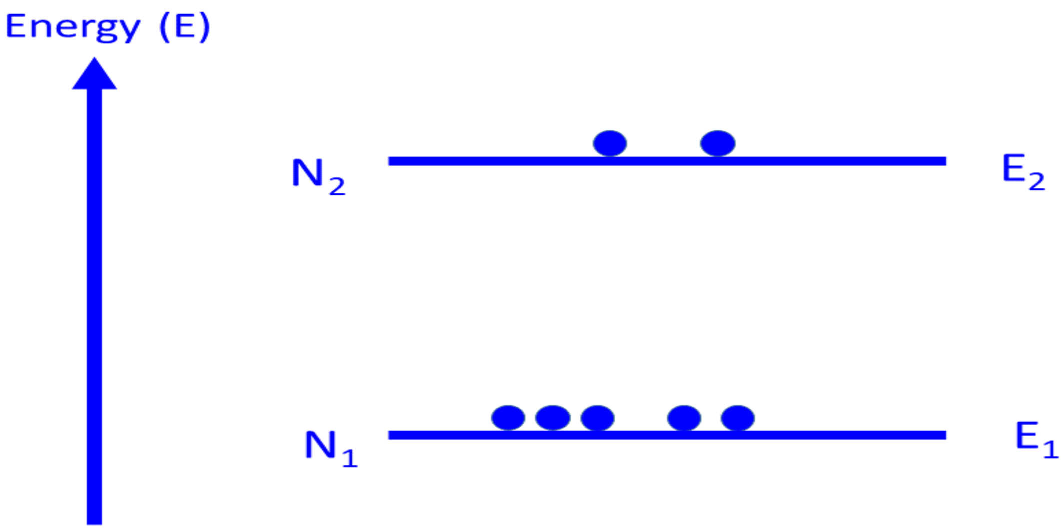

FIGURE 1.2: Occupation of the two-level systems (blue circles).

It is clear from Eqn. (1.2) that the population of the excited state ($NP_2 = N_2$) is (i) reduced at the rate A due to the spontaneous emission, (ii) reduced at the rate $B_{21}u(\omega_0)$ due to the stimulated emission, and (iii) increased at the rate $B_{12}u(\omega_0)$ due to the stimulated absorption. On the other hand, it is clear from Eqn. (1.3) that the population of the ground state ($NP_1 = N_1$) is (i) increased at the rate A due to the spontaneous emission, (ii) increased at the rate $B_{21}u(\omega_0)$ due to the stimulated emission, and (iii) decreased at the rate $B_{12}u(\omega_0)$ due to the stimulated absorption [4, 7]. In thermal equilibrium, the occupation probabilities of the two-level system becomes constant ($\frac{dP_2}{dt} = \frac{dP_1}{dt} = 0$) and they take the form of the Boltzmann probability $P_i \propto \mathrm{e}^{-E_i/k_B T}$ where T is the temperature of the blackbody cavity. Thus from the above two equations we get [4]

$$u(\omega_0) = \frac{A}{(P_1/P_2)B_{12} - B_{21}} \tag{1.4}$$

for $P_1/P_2 = \mathrm{e}^{[E_2-E_1]/k_B T}$ under the consideration of the thermal equilibrium at temperature T. Now comparing this formula for the energy density per unit frequency interval around the resonance with Planck's blackbody radiation formula [9]

$$u(\omega_0) = \frac{\hbar\omega_0^3}{\pi^2 c^3}\left[\frac{1}{\mathrm{e}^{\hbar\omega_0/k_B T} - 1}\right] \tag{1.5}$$

we get [1]

$$B = B_{21} = B_{12} \tag{1.6}$$

and [1]

$$\frac{A}{B} = \frac{\hbar\omega_0^3}{\pi^2 c^3} \tag{1.7}$$

where c is the speed of light. This is how Einstein determined the A and B coefficients [1]. The Einstein coefficients are well known to the scientific community in connection with the atomic spectral lines and the laser transitions. The equality of the stimulated emission rate and the stimulated absorption rate was also a key to reach Plank's blackbody radiation formula. The Einstein coefficients quantify intrinsic properties of atoms/molecules and is independent of the populations and of the radiation field.

1.3 Two-level system

A two-level system, also known as a two-state system, is a quantum mechanical system which has only two non-degenerate energy eigenstates. There are infinitely many energy eigenstates for an atom or a molecule. Hilbert space is infinite dimensional for such quantum mechanical systems [4]. Electric dipole transitions of the atoms/molecules are the common study of interest. Hydrogen atom emits sharp spectral lines as a consequence of the electric dipole transitions. The spectral lines, of course, are formed as a result of allowed transitions between the two energy eigenstates [10]. For many experimental realizations, electric dipole transitions of an atom/molecule are considered between its only two energy eigenstates. The atom/molecule under consideration for these two energy eigenstates can be treated as a two-level system. The Hilbert space of a two-level system is, of course, two dimensional. The two-level system has an advantage of easily studying its dynamics analytically without making any approximation.

The two-level consideration of atomic or molecular systems in low concentration in resonance is of great importance especially in laser transitions. The simplest two-level system in quantum mechanics is a spin 1/2 particle of energy eigenvalues $\mu_B B$ and $-\mu_B B$ in a magnetic field ($\vec{B} = B\hat{k}$). The interaction of a two-level system with the electromagnetic field is of great importance and it has huge applications in quantum optics [11].

Let the bare Hamiltonian of a two-level system be $\hat{H}_0$. Let the energy eigenstates of the two-level system be $|\psi_1\rangle$ and $|\psi_2\rangle$ such that $\hat{H}_0|\psi_1\rangle = E_1|\psi_1\rangle$ and $\hat{H}_0|\psi_2\rangle = E_2|\psi_2\rangle$. Here, E_1 and E_2 ($E_2 > E_1$) are two energy eigenvalues. We further have the orthonormality condition $\langle\psi_1|\psi_2\rangle = \delta_{12}$.

The quantum mechanical state of the two-level system at time t can be written as linear combination (superposition) of the energy eigenstates $|\psi_1\rangle$ and $|\psi_2\rangle$ as [5]

$$|\Psi(t)\rangle = c_1 \mathrm{e}^{-iE_1 t/\hbar}|\psi_1\rangle + c_2 \mathrm{e}^{-iE_2 t/\hbar}|\psi_2\rangle \tag{1.8}$$

where c_1 and c_2 are the probability amplitudes of the states ψ_1 and ψ_2, respectively and follows the normalization condition $|c_1|^2 + |c_2|^2 = 1$ [4].

1.3.1 Transition dipole moment

The transition dipole moment decides whether a transition is possible under the electric dipole interaction. In electrostatics, if two equal charges $\pm q$ are separated by a distance $\vec{r}$, then the dipole moment of the system of the two charges can be written as $\vec{d} = q\vec{r}$. The direction of the dipole moment is considered from the location of the negative to the location of the positive charge. In quantum mechanics, the dipole interaction resulting in the atomic transition from the initial state $|\psi_2\rangle$ to the final state $|\psi_1\rangle$ is characterised by the transition dipole moment d_{21} such that $d_{21} = \langle\psi_2|\vec{d}\cdot\hat{k}|\psi_1\rangle$ where $\vec{d}$ is the dipole moment operator for the two-level system and $\hat{k}$ is the unit vector along the z-axis [4, 12]. Since the dipole operator is a Hermitian operator, we can conveniently assume the transition dipole moment to be a real quantity. In that case, we have $d_{21} = d_{12}$. Otherwise we can take only the absolute value of $\langle\psi_2|\vec{d}\cdot\hat{k}|\psi_1\rangle$ as the transition dipole moment. If d_{21} is zero then there will be no dipole transition and the transition is said to be forbidden [5]. The atomic spectroscopic selection rule ($\triangle l = \pm 1$) determines whether a transition is possible. The transition dipole moment determines the strength of the light-matter interactions. The transition dipole moment is also defined to be as a vector $\vec{d}_{21} = \langle\psi_2|\vec{d}|\psi_1\rangle$. This vector definition is useful for the determination of the Einstein A and B coefficients.

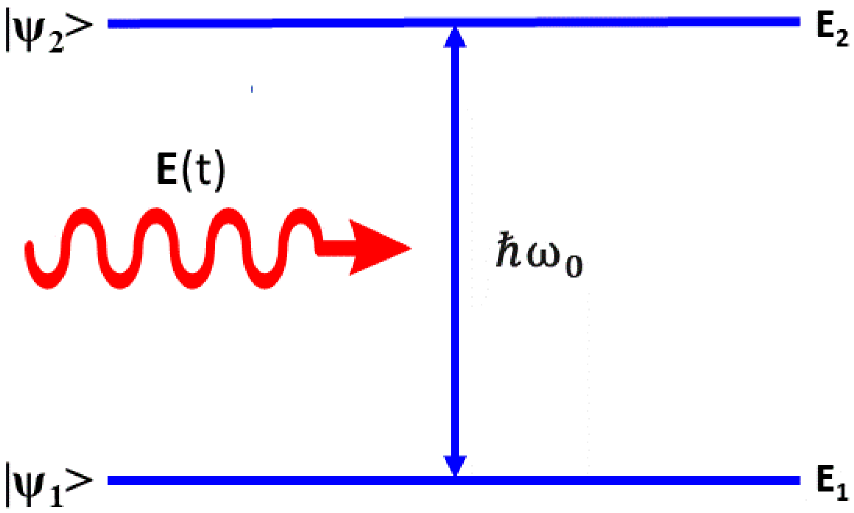

FIGURE 1.3: A typical interaction between the light and the matter (a two-level system). The fundamental processes for the light-matter interactions are shown in figure 1.1.

1.3.2 Interaction of a two-level system with the electromagnetic field

The wave-particle duality of light has its own significance in explaining various experimental outcomes. While both the semiclassical light-matter interactions and the quantum light-matter interactions describe the Rabi oscillation in a two-level system, the former cannot explain the spontaneous emission and the collapse-revival phenomena [13]. However, the stimulated emission from a two-level system and the stimulated absorption by a two-level system can be explained by both the semiclassical theory and the quantum theory for the light-matter interactions. Effective Hamiltonian for a two-level system interacting with an electromagnetic wave via its electric dipole moment, can be written within the electric dipole approximation ($\vec{k} \cdot \vec{r} \ll 1$[1]) as [7, 11, 14, 15, 16]

$$\begin{aligned} \hat{H} &= \hat{H}_0 + \hat{H}'(t) \\ &= \hat{H}_0 - \hat{\vec{d}} \cdot \hat{\vec{E}}(t) \end{aligned} \tag{1.9}$$

where $\hat{H}_0$ is the bare Hamiltonian for the two-level system, $\hat{\vec{d}}$ is the electric dipole moment operator of the two-level system, and $\hat{\vec{E}}(t)$ is the time (t) dependent electric field operator [8]. A schematic diagram for a typical light-matter interaction for a two-level system in shown in figure 1.3. The operator over the electric field is removed in the semiclassical theory. While for the classical case of the oscillatory electric field ($\vec{E}(t)$) with the (angular) frequency ω we have $\vec{E}(t) = \vec{E}_0 \cos(\omega t)$ [4, 11], for the quantum (field theoretic) case of the same we have $\hat{\vec{E}}(t) = i \sum_{\vec{k}s} \hat{e}_{\vec{k}} \left(\frac{\hbar\omega_{\vec{k}}}{2\epsilon_0 V}\right)^{1/2} [\hat{a}_{\vec{k}s} e^{-i\omega_{\vec{k}} t} -$

[1]Here, $\vec{r}$ is position of an electron in an atom/molecule.

$\hat{a}^{\dagger}_{\vec{k}s} e^{i\omega_{\vec{k}}t}]$ where $\hat{a}_{\vec{k}s}$ is the annihilation operator which annihilates a photon (boson) of the wave-vector $\vec{k}$ and the polarization s in the Fock space, $\hat{a}^{\dagger}_{\vec{k}s}$ is the creation operator which creates a photon (boson) of the wave-vector k and the polarization s in the Fock space, $\omega_{\vec{k}}$ is the frequency of the photon of the wave-vector k, $\hat{e}_{\vec{k}}$ is the unit vector for the polarization of the photon of the wave-vector $\vec{k}$, and V is the effective volume of a cavity where the light-matter interactions are taking place [11, 8].

In the classical case the light-matter coupling term $-\hat{\vec{d}} \cdot \hat{\vec{E}}(t)$ is replaced by $-\vec{d} \cdot \vec{E}(t)$ where $\vec{d}$ is the electric dipole moment [4]. On the other hand, in the quantum field theoretic case, the light-matter coupling term $-\hat{\vec{d}} \cdot \hat{\vec{E}}(t)$ requires $\hat{\vec{d}} \cdot \hat{e}_{\vec{k}}$ in operator form $\hat{\vec{d}} \cdot \hat{e}_{\vec{k}} = \langle\psi_1|\vec{d} \cdot \hat{e}_{\vec{k}}|\psi_2\rangle \left[|\psi_1\rangle\langle\psi_2| + |\psi_2\rangle\langle\psi_1|\right]$ where $= \langle\psi_1|\vec{d} \cdot \hat{e}_{\vec{k}}|\psi_2\rangle$ is assumed to be as a real quantity [8]. While the operator $|\psi_1\rangle\langle\psi_2| = \sigma_-$ is called as the lowering operator for the electric dipole transition (emission), the operator $|\psi_2\rangle\langle\psi_1| = \sigma_+$ is called as the raising operator for the electric dipole transition (absorption) [8]. It should be mentioned that the explicit time dependence of the coupling term $\hat{\vec{d}} \cdot \hat{\vec{E}}(t)$ is often ignored in the Schrödinger picture of the quantum field theoretic models, such as the Jaynes-Cumming model, because the quantized electromagnetic field is not treated as an external object with given time dependence [17]. In the quantum field theoretic models, the quantized electromagnetic field is included in the degree of freedom of the entire system [17]. The light-matter coupling $-\hat{\vec{d}} \cdot \hat{\vec{E}}$ has four types of terms, e.g. $\sigma_-\hat{a}_{\vec{k}s}$, $\sigma_+\hat{a}_{\vec{k}s}$, $\sigma_-\hat{a}^{\dagger}_{\vec{k}s}$, and $\sigma_+\hat{a}^{\dagger}_{\vec{k}s}$. The interaction terms involving the operators $\sigma_-\hat{a}_{\vec{k}s}$ and $\sigma_+\hat{a}^{\dagger}_{\vec{k}s}$ oscillate so fast in the interaction picture that they can be neglected under the rotating wave approximation [8]. These terms also leads to the energy non-conserving (off-shell) processes [11].

1.3.3 Rotating wave approximation

The rotating wave approximation (RWA) is a approximation applicable for light-matter interactions around the resonance. The RWA is applied to get approximate analytic form for the transition probability of a two-level system [14]. The RWA is applied not only to the exactly solvable models such as Rabi model [6] and Jaynes-Cummings model [18], but also to the case of time-dependent sinusoidal perturbation in the first order perturbation theory of quantum mechanics [4]. This approximation is used to neglect the rapidly oscillating terms which appear in the interaction picture. Suppose a two-level system with $E_2 - E_1 = \hbar\omega_0$ is interacting with a single-mode resonant electromagnetic field of frequency ω. The terms like $e^{\pm i(\omega-\omega_0)t}$ and $e^{\pm i(\omega+\omega_0)t}$ appear in the interaction picture. One can neglect the terms $e^{\pm i(\omega+\omega_0)t}$ comparing with terms $e^{\pm i(\omega-\omega_0)t}$ in the interaction picture for frequency (ω) of the incident electromagnetic wave near around the Bohr frequency (ω_0). The fast oscillating terms quickly averages to zero in an appreciable time scale. The RWA can also be conveniently written as $\omega + \omega_0 \gg |\omega - \omega_0|$ [8, 14].

1.4 Light-matter coupling regimes

A semiclassical theory for the light-matter interactions was introduced by Einstein almost a century back [1]. In the early eighties, it was possible for atoms in optical and microwave cavities to couple with the cavity modes. This led to the foundation of the cavity quantum electrodynamics (QED) [7, 19]. The most interesting feature of the cavity QED is that, the spontaneous emission from excited atoms or molecules can be greatly suppressed or enhanced by placing them in mirrors or in cavities, such as Fabry-Perot cavity, by virtue of the Purcell effect [20, 21]. Experimentalists basically engineer the vacuum inside the cavity to observe the Purcell effect [21]. Recently with the great advent of technologies it has become possible to explore the quantum dynamics of a two-level system in the deep and ultra strong coupling regimes [22].

Strength of the light-matter coupling for a two-level system is usually analysed at the resonance condition which is achieved by tuning the cavity in such a way that frequency of one of the cavity mode becomes equal to the Bohr frequency of the system. The relative strength of the light matter interactions in the cavity is determined based on the three parameters as follows [7]:

i). $\kappa = \frac{\omega_0}{Q}$: the photon decay rate of the cavity of the quality factor Q,

ii). γ: the non-resonant decay rate, and

iii). g_{ω_0}: the light-matter coupling constant.

The non-resonant decay rate γ, however, is related to the Einstein A coefficient by the relation $\gamma = \frac{A}{2}(1 - \frac{\Delta\Omega}{4\pi})$ where $\Delta\Omega$ is the solid angle subtended by the cavity mode [7].

By the weak light-matter interactions we mean $g_{\omega_0} \ll \sup\{\gamma, \kappa\}$ where $\sup\{\gamma, \kappa\}$ represents the larger of κ and γ [7].

On the other hand, by the strong light-matter interactions, we mean $g_{\omega_0} \gg \sup\{\gamma, \kappa\}$ [7].

While the quantum Rabi oscillation is studied for strong light-matter interactions ($g_{\omega_0} \gg \sup\{\gamma, \kappa\}$ [7]), the Einstein rate equations are often applied for weak light-matter interactions ($g_{\omega_0} \ll \sup\{\gamma, \kappa\}$ [7]). Incidentally, the Jaynes-Cummings model gives results in both the weak coupling regime and the strong coupling regime as far as the rotating wave approximation is applicable. The Jaynes-Cummings model, however, is not applicable in the ultrastrong coupling ($g_{\omega_0} \sim \omega_0$) and deep strong coupling ($g_{\omega_0} \gg \omega_0$) regimes [23, 24]. The quantum Rabi model which generalizes the Jaynes-Cummings model is applicable in these regimes [22, 25].

1.5 Time-dependent perturbation theory of quantum mechanics

The time-dependent perturbation theory of quantum mechanics was originally developed by Dirac [3]. Let us consider an orthonormal and complete set of functions

($\{\phi_n\}$) which are the solutions to the time-independent Schrödinger equation

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V^{(0)}(r)\right]\phi_n(\vec{r}) = E_n^{(0)}\phi_n(\vec{r}) \tag{1.10}$$

such that $\int \phi_n^*(\vec{r})\phi_{n'}(\vec{r})\mathrm{d}^3\vec{r} = \delta_{n,n'}$ holds for the orthonormality and $\sum_n \phi_n^*(\vec{r})\phi_n(\vec{r}') = \delta^3(\vec{r}-\vec{r}')$ holds for the completeness [4]. Here, $E_n^{(0)}$ is energy eigenvalue of the system (e.g. a point particle) in the state $|\phi_n\rangle$.

Let us now consider a time-dependent Hamiltonian [26]

$$H = -\frac{\hbar^2}{2m}\nabla^2 + V^{(0)}(r) + \lambda V(\vec{r},t) \tag{1.11}$$

where $H^{(0)} = -\frac{\hbar^2}{2m}\nabla^2 + V^{(0)}(r)$ is the time-independent part (original Hamiltonian) and $\lambda V(\vec{r},t)$ is the time-dependent part ($H'(t)$) of the Hamiltonian H. The time-dependent part can be treated as a perturbation if $\lambda \ll 1$. The quantum mechanical state ($\psi(\vec{r},t)$) of the system follows Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\psi(\vec{r},t) = H\psi(\vec{r},t) \tag{1.12}$$

and takes the form of the superposition of the energy eigenstates as

$$\psi(\vec{r},t) = \sum_n c_n(t)\mathrm{e}^{-iE_n^{(0)}t/\hbar}\phi_n(\vec{r}) \tag{1.13}$$

where $\{c_n(t)\}$ are time-dependent coefficients. These coefficients are time-independent for the case of time-independent perturbation theory.

Let the system initially was in the state $\psi(\vec{r},t) = \mathrm{e}^{-iE_k^{(0)}t/\hbar}\phi_k(\vec{r})$ and the perturbation was switched on at $t = t_0$. To the first order in λ, the time-dependent coefficients can be determined as

$$c_n(t) \simeq \delta_{n,k} + \frac{\lambda}{i\hbar}\int_{t_0}^{t} \mathrm{e}^{i[E_n^{(0)}-E_k^{(0)}]t'/\hbar}\langle\phi_n|\,\hat{V}(t')\,|\phi_k\rangle\,\mathrm{d}t' \tag{1.14}$$

for $t > t_0$. On the other hand, we have $c_n(t) = \delta_{n,k}$ for $-\infty < t \le t_0$. The transition probability comes into the consideration for $t > t_0$ and it is defined as $P_{k\to n}(t) = |c_n(t) - c_n(t_0)|^2$ for the transition of the system from the sate $|\phi_k\rangle$ to the state $|\phi_n\rangle$. Here-from we get the transition probability within the first order as [26]

$$P_{k\to n}(t) \simeq \left|\frac{\lambda}{i\hbar}\int_{t_0}^{t} \mathrm{e}^{i[E_n^{(0)}-E_k^{(0)}]t'/\hbar}\langle\phi_n|\,\hat{V}(t')\,|\phi_k\rangle\,\mathrm{d}t'\right|^2. \tag{1.15}$$

1.5.1 Sinusoidal perturbation

For a sinusoidal perturbation we take the time-dependent perturbing term as $\lambda V(\vec{r},t) = H'(\vec{r},t) = V(r)\cos(\omega t)$ where ω is the (angular) frequency of the perturbation. Thus

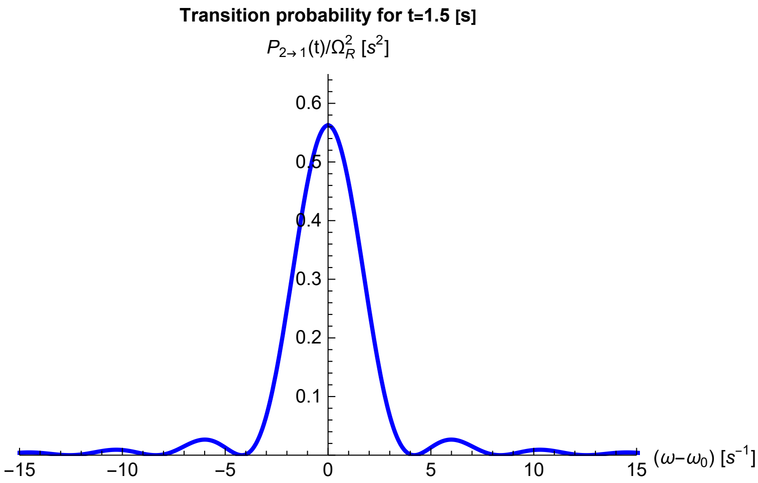

FIGURE 1.4: Transition probability as a function of frequency of the electromagnetic field. The plot follows Eqn. (1.18).

we recast Eqn. (1.14) for $t_0 = 0$ as

$$c_n(t) - c_n(0) \simeq -\frac{V_{nk}}{2\hbar}\left[\frac{e^{i[\omega-\omega_0]t} - 1}{\omega - \omega_0} - \frac{e^{-i[\omega_0+\omega]t} - 1}{\omega_0 + \omega}\right] \tag{1.16}$$

where $V_{nk} = \langle\phi_n|\,\hat{V}\,|\phi_k\rangle$ and $\omega_0 = [E_k^0 - E_n^0]/\hbar$. Now applying the rotating wave approximation ($\omega + \omega_0 \gg |\omega - \omega_0|$) we get the above transition probability as

$$P_{k\to n}(t) = |c_n(t) - c_n(0)|^2 \simeq \frac{|V_{nk}|^2}{\hbar^2}\frac{\sin^2(\omega-\omega_0)t/2}{[\omega-\omega_0]^2} \tag{1.17}$$

1.5.2 Sinusoidal perturbation for a two-level system

Let a two-level system (atom) be interacting with the electric field part ($\vec{E}(t)$) of an electromagnetic field. Under the electric dipole approximation the perturbing term, which represents the light-matter interaction, can be written as $V(r)\cos(\omega t) = -\vec{d}\cdot\vec{E}(t) = -\vec{d}\cdot\vec{E}_0\cos(\omega t)$ where $\vec{E}_0 = E_0\hat{k}$ is the amplitude of the electric field, $\vec{d} = -e\vec{r}$ is the electric dipole moment of the two-level system, $\vec{r}$ is the position of the electron with respect to the nucleus of two-level system, and $\hat{k}$ is the unit vector along the z-axis. Let us consider $|\phi_k\rangle = |\psi_2\rangle$ with the energy eigenvalue E_2 and $|\phi_n\rangle = |\psi_1\rangle$ with the energy eigenvalue E_1. Bohr frequency of the two-level system is $\omega_0 = [E_2 - E_1]/\hbar > 0$. Now we get the transition probability for the two level-system from Eqn. (1.17) as [4]

$$P_{2\to 1}(t) \simeq \Omega_R^2\frac{\sin^2([\omega-\omega_0]t/2)}{[\omega-\omega_0]^2} \tag{1.18}$$

where $\Omega_R = \frac{|\langle\psi_2|\vec{d}\cdot\vec{E}_0|\psi_1\rangle|}{\hbar}$ is the Rabi frequency [6]. Above formula of the transition probability is plotted in figure 1.4. It is clear from the figure that most of the transitions take place near around the resonance ($\omega = \omega_0$).

1.5.3 Dirac's determination of the Einstein B coefficient

Although Einstein obtained a relation between the Einstein coefficients (see Eqn. (1.7)) he could not determine either of the coefficients because of limitations of the old quantum mechanics. While the Einstein A coefficient was determined by Weisskopf-Wigner [2] within the quantum field theory for the electrodynamics, the Einstein B coefficient was determined by Dirac [3] within the 1st order perturbation theory of quantum mechanics. Before going to the derivation of the A coefficient let us see how Dirac determined the B coefficient [3]. Dirac determined the Einstein B coefficient for a two-level system system in large blackbody cavity ($V \to \infty$). Thermal radiation field inside the blackbody cavity interact with the two-level system in this regard.

The energy density of the incident monochromatic electromagnetic field, as described in the previous subsection, can be written as $u = \frac{\epsilon_0}{2} E_0^2$ [4]. Thus in terms of the energy density, we recast the transition probability of Eqn. (1.18) as [4]

$$P_{2\to 1}(t) \simeq \frac{2u}{\epsilon_0 \hbar^2} d_{21}^2 \frac{\sin^2([\omega - \omega_0]t/2)}{[\omega - \omega_0]^2} \tag{1.19}$$

where the transition dipole moment $d_{21} = \langle \psi_2 | \vec{d} \cdot \hat{k} | \psi_1 \rangle$ is assumed real and $\hat{k}$ is a unit vector along the z-axis. If the transition dipole moment is not real then we have to replace it by its absolute value ($d_{21} \to |d_{21}|$). All the frequencies ($0 < \omega < \infty$) of the electromagnetic fields contribute in the light-matter interactions for the two-level system in the thermal radiation field. The thermal radiation field is also unpolarized. For such a case we replace u by $u(\omega)$ in Eqn. (1.19) and call $u(\omega)$ as energy density per unit frequency interval. If the z-axis be the direction of propagation of the electromagnetic field, then the unit vector for the polarization $\hat{e}_{\vec{k}}$ can be oriented arbitrarily in the $x - y$ plane. Now we further have to integrate the transition probability over all possible frequencies to get the net transition probability. We also have to take averaging $|\vec{d} \cdot \hat{e}_{\vec{k}}|^2$ over all the directions of incidence of the electromagnetic fields and all the independent polarizations of the electromagnetic fields [4]. Thus we get the net transition probability as [4]

$$P_{2\to 1}(t) \simeq \frac{2|\vec{d}_{21}|^2}{3\epsilon_0 \hbar^2} \int_0^\infty u(\omega) \frac{\sin^2([\omega - \omega_0]t/2)}{[\omega - \omega_0]^2} d\omega \tag{1.20}$$

where $\vec{d}_{21} = \langle \psi_2 | \vec{d} | \psi_1 \rangle$ is the transition dipole moment. Here, the factor $\frac{1}{3}$ comes from averaging over all the directions of incidence and the two independent polarizations [4]. However, since most of the transitions are taking place at around the resonance ($\omega = \omega_0$), we replace $u(\omega)$ with $u(\omega_0)$ in Eqn. (1.20). It is clear from Eqn. (1.20) that most of the contributions in the net transition probability are coming from around the removal singularity in the sinc function i.e. from around the resonance

($\omega \to \omega_0$). We can further assume $\omega_0 t \to \infty$. Thus we recast Eqn. (1.20) as [4]

$$P_{2\to 1}(t) \simeq \frac{\pi|\vec{d}_{21}|^2}{3\epsilon_0\hbar^2}u(\omega_0)t \tag{1.21}$$

for $\omega_0 t \to \infty$. Note that the net transition probability for a two-level system in the thermal radiation field is linear with the time t for the limiting case $\omega_0 t \to \infty$. This result is compatible with the Fermi's golden rule [27]. It is clear from Eqn. (1.21) that the rate ($R_{2\to 1} = \frac{d}{dt}P_{2\to 1}(t)$) of transitions i.e. the rate of stimulated emission is independent of time for $\omega_0 t \to \infty$, and it takes the form

$$R_{2\to 1} = \frac{\pi|\vec{d}_{21}|^2}{3\epsilon_0\hbar^2}u(\omega_0). \tag{1.22}$$

Now by following the definition ($B_{21} = R_{2\to 1}/u(\omega_0)$), we get the Einstein B coefficient from Eqn. (1.22) as [3]

$$B_{21} = \frac{\pi|\vec{d}_{21}|^2}{3\epsilon_0\hbar^2}. \tag{1.23}$$

This is Dirac's determination of the Einstein B coefficient [3].

1.6 Weisskopf-Wigner determination of the Einstein A coefficient

The Einstein A coefficient was determined by Weisskopf and Wigner within the quantum field theory for a two-level system interacting with the vacuum modes of the electromagnetic field [2]. No photons are involved in making a spontaneous emission from the two-level system [2]. What it requires are the quantum (vacuum) fluctuations of the electromagnetic field around the system [2].

Let the two bare states of the two-level system be given by $|\psi_1\rangle$ with energy eigenvalue $E_1 = 0$ and $|\psi_2\rangle$ with energy eigenvalue $E_2 > 0$. Bohr frequency of the system is $\omega_0 = (E_2 - 0)/\hbar$. Let the two-level system be kept inside a blackbody cavity of volume $V \to \infty$. The effective Hamiltonian for the two-level system, electromagnetic field, and the light-matter interactions can be written within the rotating wave approximation after quantization of the electromagnetic field as [11]

$$\hat{H} = \hbar\omega_0 |\psi_2\rangle\langle\psi_2| + \sum_{\vec{k}s}\hbar\omega_{\vec{k}}\hat{a}^{\dagger}_{\vec{k}s}\hat{a}_{\vec{k}s} - \sum_{\vec{k}s}[\hbar g_{\vec{k}s}|\psi_2\rangle\langle\psi_1|\hat{a}_{\vec{k}s} + H.c.] \tag{1.24}$$

where the first term represents the bare Hamiltonian of the two-level system, the second term represents the Hamiltonian of the electromagnetic field, the third term represents the light-matter interactions, $\hat{a}_{\vec{k}s}$ is the bosonic annihilation operator which annihilates a photon of energy $\hbar\omega_{\vec{k}}$, polarization s and momentum $\hbar\vec{k}$ (having dispersion $\omega_k = ck$) in the Fock space, $g_{\vec{k}s} = i\sqrt{\frac{\omega_{\vec{k}}}{2\hbar\epsilon_0 V}}\langle\psi_1|\vec{d}\cdot\hat{\epsilon}_{\vec{k}s}|\psi_2\rangle$ is the light-matter coupling constant, $\hat{\epsilon}_{\vec{k}s}$ is the unit-vector for the polarization of a photon in the mode $\vec{k}$, and $\vec{d}$ is the electric dipole moment operator for the two-level system.

Let us consider that the two-level system is initially ($t = 0$) in the excited state $|\psi_2\rangle$ and there are no photons at time $t = 0$. Therefore, the combined state of the two-level system and field can be written at $t = 0$ as $|\psi(0)\rangle = |\psi_2, \{0\}\rangle$. After some time the two-level system makes a spontaneous transition to the ground state $|\psi_1\rangle$ after emitting a photon. Therefore, the combined state of the two-level system and field at time t can be written as [2]

$$|\Psi(t)\rangle = a(t)\mathrm{e}^{-i\omega_0 t}|\psi_2, \{0\}\rangle + \sum_{\vec{k}s} b_{\vec{k}s}(t)\mathrm{e}^{-i\omega_{\vec{k}} t}|\psi_1, \{1_{\vec{k}s}\}\rangle \tag{1.25}$$

where $|\psi_1, \{1_{\vec{k}s}\}\rangle$ represents the combined state of the two-level system (in the ground state) and the emitted photon (of mode $\vec{k}$ and polarization s), $a(t)$ represents the amplitude of decay due to the spontaneous emission, and $b_{\vec{k}s}(t)$ is the amplitude for a radiation (of wave-vector $\vec{k}$ and polarization state s) due to the spontaneous emission. Now solving the Schrödinger equation, $i\hbar\frac{\partial|\Psi(t)\rangle}{\partial t} = \hat{H}|\Psi(t)\rangle$, for the initial conditions as mentioned above, we get [11]

$$\frac{\mathrm{d}a(t)}{\mathrm{d}t} = -\sum_{\vec{k}s}|g_{\vec{k}s}|^2\int_0^t \mathrm{e}^{-i(\omega_{\vec{k}}-\omega_0)(t-t')}a(t')dt' \tag{1.26}$$

and [11]

$$\frac{\mathrm{d}b_{\vec{k}s}(t)}{\mathrm{d}t} = ig^*_{\vec{k}s}\mathrm{e}^{i(\omega_{\vec{k}}-\omega_0)t}a(t). \tag{1.27}$$

The discrete set of modes set to be continuum for $V \to \infty$. Let us now consider the Weisskopf-Wigner approximation ($a(t') = a(t)$), i.e. the amplitude $a(t)$ varies with the constant rate A for $A \ll \omega_0$. Such an approximation is compatible with the weak light-matter coupling regime. Under this approximation and for $V \to \infty$, Eqn. (1.26) takes the form [2, 11]

$$a(t) = \mathrm{e}^{-At/2} \tag{1.28}$$

where [2]

$$A = \frac{\omega_0^3|\vec{d}_{21}|^2}{3\pi\epsilon_0\hbar c^3} \tag{1.29}$$

is the Einstein A coefficient and $\vec{d}_{21} = \langle\psi_2|\hat{\vec{d}}|\psi_1\rangle$ is the transition dipole moment. Since the amplitude $a(t)$ decay with the rate $A/2$ due to the spontaneous emission, the energy ($\sim |a(t)|^2$) decays with the constant rate A. Thus the Einstein A coefficient is the natural decay rate. Now from Eqn. (1.27), we get Lorentzian broadening

[2]

$$\lim_{t\to\infty} |b_{\vec{k}s}(t)|^2 = \frac{|g_{\vec{k}s}|^2}{A^2/4 + [\omega_{\vec{k}} - \omega_0]^2} \tag{1.30}$$

for the frequencies of the emitted photons around the Bohr frequency. It is interesting to note that the width of this distribution is the Einstein A coefficient. It should be mentioned in this regard that the A coefficient becomes constant because of the consideration of the Weisskopf-Wigner approximation. The A coefficient may not necessarily be constant beyond this approximation [13].

1.7 Population dynamics and entropy production for two-level systems

The Einstein rate equations (Eqns. (1.2) ans (1.3)) allow us to study nonequilibrium statistical mechanics e.g. population dynamics and entropy production of two-level systems in the thermal radiation field.

1.7.1 Population dynamics

Let us consider an ideal classical gas of two-level systems in a large ($V \to \infty$) black-body cavity at a temperature T. Let P_1 be the probability of occupation of the ground state ($|\psi_1\rangle$) of the system and P_2 be the probability of occupation of the excited state ($|\psi_2\rangle$) of the system. Bohr frequency of the two-level system has already been mentioned to be as $\omega_0 = [E_2 - E_1]/\hbar$. We already have mentioned in Section 1.2 that the population dynamics of the gas of the two-level systems follows from the Einstein rate equations [1, 4, 28]:

$$\frac{\mathrm{d}P_2}{\mathrm{d}t} = -AP_2 - P_2R_{21} + P_1R_{12} \tag{1.31}$$

and

$$\frac{\mathrm{d}P_1}{\mathrm{d}t} = AP_2 + P_2R_{21} - P_1R_{12}. \tag{1.32}$$

where $R_{21} = B_{21}u(\omega_0) = R_{12} = B_{12}u(\omega_0) = R$ is the rate of stimulated transitions of a two-level system at the resonance ($\omega = \omega_0$). Above two rate equations, however, can be combined as

$$\frac{\mathrm{d}P_2}{\mathrm{d}t} = R - [A + 2R]P_2 \tag{1.33}$$

by virtue of the conservation of the occupation probabilities i.e.

$$P_1 + P_2 = 1. \tag{1.34}$$

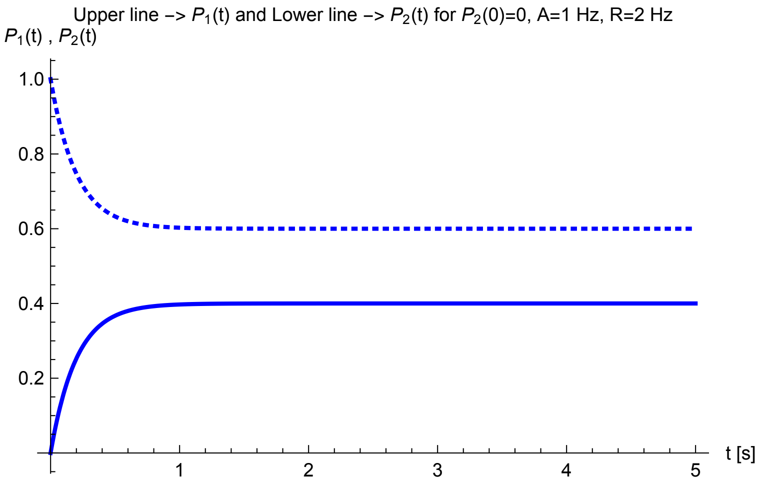

FIGURE 1.5: The solid line ($P_2(t)$) follows Eqn. (1.35) for the initial condition $P_2(0) = 0$ and the parameters as mentioned in the plot label. The dotted line ($P_1(t)$) follows Eqn. (1.34) for the same initial condition and the same parameters.

Initial condition: $P_2(0) = 0$

If initially i.e. at time $t = 0$, the two-level system is found to be in the ground state i.e. $P_2(0) = 0$, then Eqn. (1.33) has the physical solution for the occupation probability $P_2(t)$ as

$$P_2(t) = \frac{R}{A+2R}\left[1 - \mathrm{e}^{-[A+2R]t}\right]. \tag{1.35}$$

We plot this probability in figure 1.5. For $t \to \infty$, the two level systems come in equilibrium with the thermal radiation field and the occupation probability $P_2(t)$ reaches the Boltzmann probability $P_2(\infty) = \frac{R}{A+2R} = \frac{\mathrm{e}^{-E_2/k_BT}}{\mathrm{e}^{-E_1/k_BT}+\mathrm{e}^{-E_2/k_BT}}$ [1, 4].

Initial condition: $P_2(0) = 1$

If initially i.e. at time $t = 0$, the two-level system is found to be in the excited state i.e. $P_2(0) = 1$, Eqn. (1.33) has the physical solution for the occupation probability $P_2(t)$ as

$$P_2(t) = \frac{R}{A+2R} + \left[1 - \frac{R}{A+2R}\right]\mathrm{e}^{-[A+2R]t}. \tag{1.36}$$

For $t \to \infty$, the two level systems come in equilibrium with the thermal radiation field and the occupation probability $P_2(t)$ reaches the same Boltzmann probability $P_2(\infty) = \frac{\mathrm{e}^{-E_2/k_BT}}{\mathrm{e}^{-E_1/k_BT}+\mathrm{e}^{-E_2/k_BT}}$ [1, 4]. The Boltzmann probability does not depend on the initial condition.

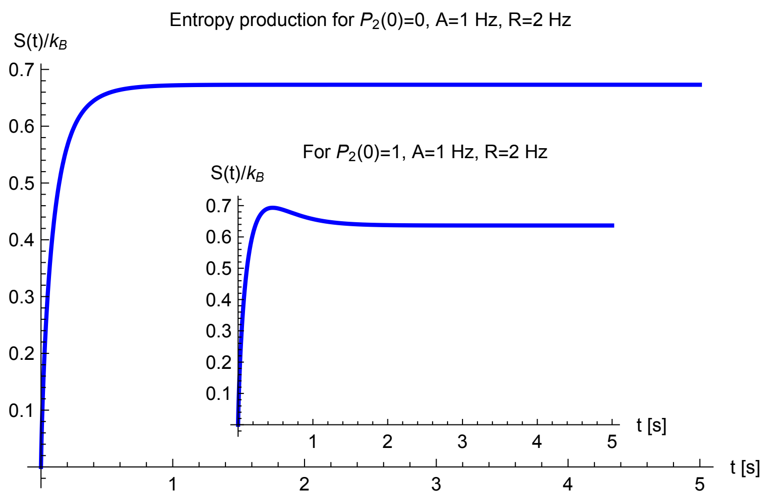

FIGURE 1.6: Entropy production of a two-level system in a blackbody cavity. The plot follows Eqn. (1.37) for the initial condition $P_2(0) = 0$ and the parameters as mentioned in the plot label. The plot in the inset follows Eqn. (1.37) for the initial condition $P_2(0) = 1$ and the same parameters as mentioned in the plot label.

1.7.2 Entropy production

Entropy production of a single two-level system in the blackbody cavity can be described by the formula [10]

$$S(t) = -k_B[P_1(t)\ln(P_1(t)) + P_2(t)\ln(P_2(t))] \tag{1.37}$$

where $S(t)$ denotes the entropy of the two-level system at time t and $P_1(t)$ & $P_2(t)$ represents the occupation probabilities which obey the Einstein rate equations (Eqns. (1.31) and (1.32)). Above form of the entropy production formula is compatible with the form prescribed by Pauli and von Neumann [29, 30].

We plot the entropy production in figure 1.6 for both the initial conditions $P_2(0) = 0$ and $P_2(0) = 1$. It is interesting to note that the entropy always increases with time if the two-level system initially occupies the ground state. Otherwise the entropy may decrease at some interval of time. However, the entropy averaged over both the realizations of the initial condition may not decrease in time as far as the light-matter couplings are considered to be weak. Incidentally, the light-matter couplings are considered to be weak for a two-level system in a blackbody cavity of volume $V \to \infty$. Otherwise, for strong light-matter interactions, the two-level system would be treated as an open system where the second law of thermodynamics is not applicable.

1.8 Failure of the 1^{st} order time-dependent perturbation theory of quantum mechanics at around the resonance ($\omega \to \omega_0$)

Let us recall the quantum mechanical result

$$P_{2\to1}(t) \simeq \Omega_R^2 \frac{\sin^2([\omega-\omega_0]t/2)}{[\omega-\omega_0]^2} \tag{1.38}$$

for the transition probability for a time-dependent sinusoidal perturbation on a two-level system as described in Eqn. (1.18). In Section 1.5.3 we have noticed that this is the key result for the derivation of the Einstein B coefficient for a two-level system in a large blackbody cavity. We have noticed that most of the contributions in the B coefficient come from around the resonance ($\omega \to \omega_0$).

The time-dependent perturbation theory is valid for $\frac{\Omega_R}{|\omega-\omega_0|} \ll 1$ [4]. Interestingly, there is a problem of normalization of the transition probability at the resonance ($\omega \to \omega_0$). The transition probability takes the form $P_{2\to1}(t) \to \frac{\Omega_R^2 t^2}{4}$ at the resonance. This form of the transition probability is not normalizable rather $P_{2\to1}(t) \to \infty$ for $t \to \infty$ for any finite small value of Ω_R for $\omega \to \omega_0$. Hence the 1^{st} order perturbation theory of the quantum mechanics fails at around the resonance even for a non-zero weak light-matter coupling ($\Omega_R/A \ll 1$) of the sinusoidal perturbation.

Since most of the contributions in the B coefficient come from around the resonance, the expression of the B coefficient determined by Dirac is questionable even for the weak coupling regime [4, 5].

Another drawback of the 1^{st} order perturbation theory is that the periodicity in stimulated transition is lost at the resonance ($\omega \to \omega_0$). The periodicity can be found in non-pertubative model such as the Rabi model [6] which is able to explain the nuclear magnetic resonance.

Hence we are motivated to determine the B coefficient within a non-perturbative model, such as Rabi model, which though is often used for strong coupling regime, can also be used for weak coupling regime as it is an exactly solvable model for the entire range of light-matter coupling.

Rabi model, however, treats the light-matter interaction classically. The Jaynes-Cummings model [18] on the other hand is another exactly solvable model for light-matter interaction. This is quantum mechanical model where light-matter interaction is treated with the quantum field theory. We are also interested to determine the Einstein A and B coefficients within the multimode Jaynes-Cummings model.

There are experimental data available for the collapse and the revival of the quantum Rabi oscillations of a two-level system in a lossy resonant cavity. We are also motivated to explain these data within the multimode Jaynes-Cummings model.

We are also motivated to study the population dynamics of a two-level system in thermal radiation field with the model dependent Einstein coefficients.

The population dynamics allows us to study non-equilibrium statistical mechanics of the two-level system in thermal radiation field. We are motivated to determine entropy production of the system.

Let us now introduce the two exactly solvable models.

1.9 Exactly solvable models for light-matter interactions

By the virtue of the existence of spin, electron has an intrinsic angular momentum as well as an intrinsic magnetic moment. The Stern-Gerlach experiment first demonstrated the space quantization of the angular momentum of an atom as well as the existence of spin of an electron. Extending the theory related to the Stern-Gerlach experiment, Rabi *et al* first theorised and observed the nuclear magnetic resonance (NMR) in molecular beams in 1939 [6]. The NMR is a phenomenon where a nuclei is perturbed by a weakly oscillating magnetic field and produce a electromagnetic field of frequency characteristic of the nucleus. The NMR spectroscopy is a result of magnetic properties of certain atomic nuclei. Rabi *et al* in their experiment considered the problem of a spin-1/2 magnetic dipole undergoing precession in a magnetic field. Rabi obtained an expression for the probability that a spin-1/2 atom incident on a Stern-Gerlach apparatus would be flipped from the $+1/2$ or $-1/2$ state to the $-1/2$ or $+1/2$ state, respectively, by an applied radio-frequency magnetic field [11]. The periodic flipping of the states is coined as the Rabi oscillation [6].

Just as the spin-1/2 system undergoes Rabi oscillation by the action of an oscillating magnetic field, a two-level system (atom/molecule) also shows Rabi oscillation by the action of an electromagnetic field [11]. While the interaction of a two-level system with magnetic field is described by $-\vec{\mu} \cdot \vec{B}$ under the magnetic dipole interaction, the interaction with electric field is described by $-\vec{d} \cdot \vec{E}$ under the electric dipole interaction [4, 5].

1.9.1 Rabi model

Let us consider a two-level system (atom/molecule) having the excited state $|\psi_2\rangle$ and the ground state $|\psi_1\rangle$ with corresponding energy eigenvalues E_2 and E_1 ($E_2 > E_1$), respectively. The Bohr frequency of the two-level system is $\omega_0 = [E_2 - E_1]/\hbar$. Let us further consider that the two-level system is interacting with a plane monochromatic electromagnetic wave incident on it. Let the electric field part of the incident electromagnetic wave be $\vec{E} = \vec{E_0} \cos(\omega t)$ where $\vec{E}_0$ be the amplitude of oscillation of the electric field and ω be the (angular) frequency of oscillation of the electromagnetic field. Let the electric dipole moment of the two-level system be $\vec{d}$. The Rabi Hamiltonian for the two-level system interacting with the electromagnetic field

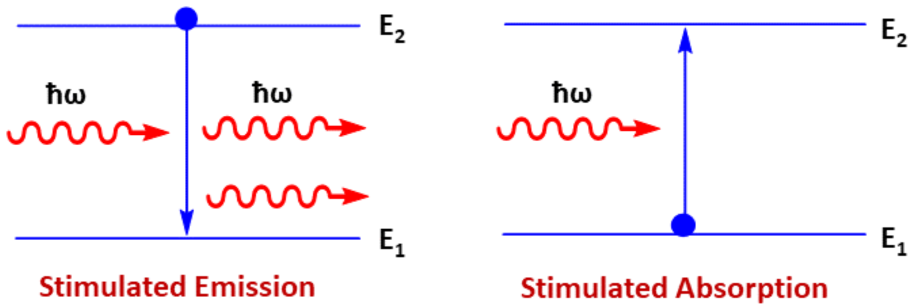

FIGURE 1.7: Schematic diagram for the fundamental processes related to the Rabi model.

within rotating wave approximation ($\omega + \omega_0 \gg |\omega - \omega_0|$) and the electric dipole approximation is given by [6, 5]

$$\hat{H} = \underbrace{E_1|\psi_1\rangle\langle\psi_1| + E_2|\psi_2\rangle\langle\psi_2|}_{\text{Bare part of the Hamiltonian: } \hat{H}_0} \underbrace{- \frac{\vec{E}_0 \cdot \vec{d}}{2}\left[\mathrm{e}^{i\omega t}|\psi_1\rangle\langle\psi_2| + \mathrm{e}^{-i\omega t}|\psi_2\rangle\langle\psi_1|\right]}_{\text{Interaction part of the Hamiltonian: } \hat{H}'(t)}. \tag{1.39}$$

The Rabi Hamiltonian describes the Rabi model. It should be mentioned in this regard that the Rabi model is a semiclassical model because the bare part of the Hamiltonian is treated quantum mechanically and the light-matter interaction part is treated classically. While the first term of the interaction part containing $|\psi_1\rangle\langle\psi_2|$ leads to an electric dipole transition from the excited state to the ground state, the second term of the interaction part containing $|\psi_2\rangle\langle\psi_1|$ leads to an electric dipole transition from the ground state to the excited state. The operator $|\psi_1\rangle\langle\psi_2|$ is called as the lowering operator and the operator $|\psi_2\rangle\langle\psi_1|$ is called as the raising operator. These are also called as transition operators. Schematic diagram for the fundamental processes related to the Rabi model is shown in figure 1.7.

The state of the system at time t can be written as [6]

$$|\psi(t)\rangle = c_1(t)\mathrm{e}^{-iE_1t/\hbar}|\psi_1\rangle + c_2(t)\mathrm{e}^{-iE_2t/\hbar}|\psi_2\rangle \tag{1.40}$$

where $c_1(t)$ is the probability amplitude of the bare state $|\psi_1\rangle$ and $c_2(t)$ is the probability amplitude of the bare state $|\psi_2\rangle$. From conservation of the total probability we have

$$|c_1(t)|^2 + |c_2(t)|^2 = 1. \tag{1.41}$$

Time evolution of the probability amplitude can, however, be determined from the Schrödinger equation

$$i\hbar\frac{\partial|\psi(t)\rangle}{\partial t} = \hat{H}|\psi(t)\rangle. \tag{1.42}$$

It should be mentioned in this regard that if the system is initially ($t = 0$) found in the excited state ($|\psi_2\rangle$), then $|c_1(t)|^2$ would be called as the transition probability ($P_{2\to 1}(t) = |c_1(t)|^2$) or the probability of a stimulated emission from the excited state to the ground state. We also have $|c_2(t)|^2 = P_{1\to 2}(t)$ as the probability of a stimulated absorption [4]. On the other hand, if the system is initially found in the ground state ($|\psi_1\rangle$), then $|c_2(t)|^2$ would be called as the transition probability ($P_{1\to 2}(t) = |c_2(t)|^2$) or the probability of a stimulated absorption. In this case, we also have $|c_1(t)|^2 = P_{2\to 1}(t)$ as the probability of a stimulated emission.

If we move on to the interaction picture from the Schrödinger picture, we have following equation for the time-dependent probability amplitudes [5]

$$i\hbar \begin{pmatrix} \dot{c_1}(t) \\ \dot{c_2}(t) \end{pmatrix} = \begin{pmatrix} 0 & V_{12}\mathrm{e}^{-i\omega_0 t} \\ V_{21}\mathrm{e}^{i\omega_0 t} & 0 \end{pmatrix} \begin{pmatrix} c_1(t) \\ c_2(t) \end{pmatrix} \tag{1.43}$$

where $V_{12} = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2}\mathrm{e}^{i\omega t}$, $V_{21} = -\frac{\langle\psi_2|\vec{E}_0\cdot\vec{d}|\psi_1\rangle}{2}\mathrm{e}^{-i\omega t}$, and $V_{11} = V_{22} = 0$. Since $\vec{E}_0\cdot\vec{d}$ is an Hermitian operator, we are assuming that $\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle$ be a real quantity so that $\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle = \langle\psi_2|\vec{E}_0\cdot\vec{d}|\psi_1\rangle$ holds. Otherwise we replace $\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle$ with $|\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle|$ and $\langle\psi_2|\vec{E}_0\cdot\vec{d}|\psi_1\rangle$ with $|\langle\psi_2|\vec{E}_0\cdot\vec{d}|\psi_1\rangle|$ for convenience [5]. Now Eqn. (1.43) leads to the following coupled differential equations

$$i\hbar\frac{\mathrm{d}}{\mathrm{d}t}c_1(t) = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2}\mathrm{e}^{i[\omega-\omega_0]t}c_2(t) \tag{1.44}$$

and

$$i\hbar\frac{\mathrm{d}}{\mathrm{d}t}c_2(t) = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2}\mathrm{e}^{-i[\omega-\omega_0]t}c_1(t). \tag{1.45}$$

Let us now solve the coupled differential equations (Eqns. (1.44) and (1.45)) for the initial conditions $c_1(0) = 1$ and $c_2(0) = 0$. It is clear from the initial conditions that the two-level system initially was in the ground state ($|\psi_1\rangle$). Let us now take the ansatz

$$\left.\begin{aligned} c_1(t) &= B(t)\mathrm{e}^{i(\omega-\omega_0)t/2} \\ c_2(t) &= A(t)\mathrm{e}^{-i(\omega-\omega_0)t/2} \end{aligned}\right\} \text{where } |B^2(t)| + |A^2(t)| = 1 \text{ due to Eqn. (1.41).}$$

Eqns. (1.44) and (1.45) take the forms [31]

$$i\hbar\dot{B}(t) - \hbar\frac{(\omega-\omega_0)}{2}B(t) = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2}A(t) \tag{1.46}$$

$$i\hbar\dot{A}(t) + \hbar\frac{(\omega-\omega_0)}{2}A(t) = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2}B(t) \tag{1.47}$$

with $B(0) = 1$ and $A(0) = 0$. Let us now take another ansatz

$$\left.\begin{aligned} B(t) &= B_0 e^{i\Omega' t} \\ A(t) &= A_0 e^{i\Omega' t} \end{aligned}\right\} \text{provided that } A_0 \text{ and } B_0 \text{ are constants.}$$

Substituting the above forms of $B(t)$ and $A(t)$ in Eqns. (1.46) and (1.47) we get non-trivial solutions of $B(t)$ and $A(t)$ if the following [31]

$$\begin{vmatrix} \hbar[\Omega' + (\omega - \omega_0)/2] & -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2} \\ -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{2} & \hbar[\Omega' - (\omega - \omega_0)/2] \end{vmatrix} = 0 \tag{1.48}$$

holds by resulting in

$$\Omega' = \pm\frac{1}{2}\left[\frac{|\langle\psi_1|E_0.\vec{d}|\psi_2\rangle|^2}{\hbar^2} + (\omega - \omega_0)^2\right]^{1/2}. \tag{1.49}$$

Now we write the general solutions for $B(t)$ and $A(t)$ as [31]

$$B(t) = \alpha e^{i\Omega' t} + \beta e^{-i\Omega' t} \tag{1.50}$$

$$A(t) = \alpha r_\alpha e^{i\Omega' t} + \beta r_\beta e^{-i\Omega' t} \tag{1.51}$$

where $r_\alpha = -\frac{\Omega' + [\omega - \omega_0]/2}{\gamma} = \frac{\gamma}{\Omega' - [\omega - \omega_0]/2}$, $r_\beta = \frac{\Omega' - [\omega - \omega_0]/2}{\gamma} = -\frac{\gamma}{\Omega' + [\omega - \omega_0]/2}$ and $\gamma = \langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle/2\hbar$. Now applying the initial conditions we have $B(0) = \alpha + \beta = 1$ and $A(0) = \alpha r_\alpha + \beta r_\beta = 0$. Here-from we get $\alpha = \frac{r_\beta}{r_\beta - r_\alpha}$ and $2i\alpha r_\alpha = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{i2\hbar\Omega'}$. Now Eqn. (1.51) can be recast as [31]

$$\begin{aligned} A(t) &= 2i\alpha r_\alpha \sin(\Omega' t) \\ &= -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{i2\hbar}\left[\frac{\sin(\Omega' t)}{\Omega'}\right] \end{aligned} \tag{1.52}$$

Therefore $c_2(t)$ takes the form $c_2(t) = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{i2\hbar\Omega'} e^{-i[\omega-\omega_0]t/2}\sin(\Omega' t)$. Thus the transition probability $P_{1\to 2}(t) = |c_2(t)|^2$ takes the form [6]

$$P_{1\to 2}(t) = \Omega_R^2 \frac{\sin^2(\Omega t/2)}{\Omega^2} \tag{1.53}$$

where $\Omega_R = \frac{|\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle|}{\hbar}$ is the Rabi frequency, $\Omega = \pm\sqrt{\Omega_R^2 + \Delta^2} = 2\Omega'$ is the generalized Rabi frequency, and $\Delta = \omega - \omega_0$ is the detuning parameter. The other probability amplitude, on the other hand, takes the form $c_1(t) = -\frac{\langle\psi_1|\vec{E}_0\cdot\vec{d}|\psi_2\rangle}{i\hbar\Omega}\left[r_\beta e^{i\Omega t/2} + r_\alpha e^{-i\Omega t/2}\right]e^{i[\omega-\omega_0]t/2}$ [31]. However, from the conservation of the total probability now we can safely write

$$P_{2\to 1}(t) = |c_1(t)|^2 = 1 - \Omega_R^2 \frac{\sin^2(\Omega t/2)}{\Omega^2} \tag{1.54}$$

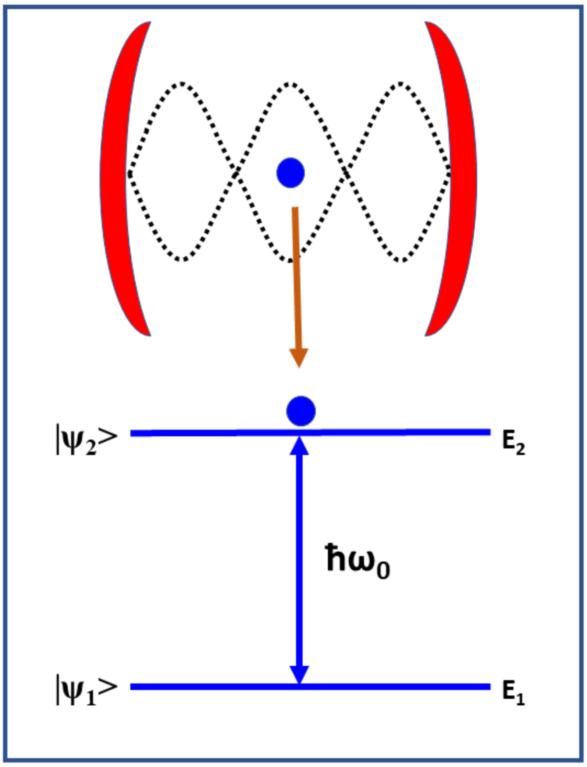

FIGURE 1.8: A two-level system in an optical cavity.

It should be mentioned in this regard that if the initial conditions are now reversed i.e. if the two-level system is initially ($t = 0$) found in the excited state, then the transition probability ($P_{2\to 1}(t)$) takes the form

$$P_{2\to 1}(t) = \Omega_R^2 \frac{\sin^2(\Omega t/2)}{\Omega^2} \tag{1.55}$$

of Eqn. (1.53).

It is to be noted that the transition probability oscillates periodically in the time domain with the generalized Rabi frequency Ω. It is also to be noted that this oscillation, called the Rabi oscillation, does not stop even at the resonance ($\omega \to \omega_0$). The transition probability oscillates with the Rabi frequency (Ω_R) in the resonance. Such as oscillation was observed in connection with the NMR [6].

1.9.2 Jaynes-Cummings model

Let us consider a two-level system (atom/molecule) having the excited state $|\psi_2\rangle$ and the ground state $|\psi_1\rangle$ with corresponding energy eigenvalues E_2 and E_1 ($E_2 > E_1$), respectively. The Bohr frequency of the two-level system is $\omega_0 = [E_2 - E_1]/\hbar$. Let us further consider that the two-level system be placed in a 1-D optical cavity and the system is interacting with a single quantized mode corresponding to the frequency ω of the cavity's electromagnetic field as shown in figure 1.8. Let the light-matter interaction in this case takes place due the coupling of the electric dipole moment $\vec{d}$ of the two-level system and the electric field part ($\vec{E} = \vec{E}_0 \cos(\omega t)$) of the electromagnetic field under the electric dipole approximation. Let z-axis be the cavity axis. The Jaynes-Cummings Hamiltonian for the two-level system interacting with the electromagnetic field within rotating wave approximation ($\omega + \omega_0 \gg |\omega - \omega_0|$) and the electric dipole approximation is given by [18, 8, 15]

$$\hat{H} = \underbrace{\frac{1}{2}\hbar\omega_0\sigma_3}_{\text{System}} + \underbrace{\hbar\omega\hat{a}^\dagger\hat{a}}_{\text{Field}} \underbrace{-i\hbar g[\sigma_+\hat{a} - \sigma_-\hat{a}^\dagger]}_{\text{System-Field Interaction}} \tag{1.56}$$

where we are following the notations [8]: $\sigma_+ = |\psi_2\rangle\langle\psi_1|$ is the raising operator for the two-level system, $\sigma_- = |\psi_1\rangle\langle\psi_2|$ is the lowering operator for the two-level system, $\sigma_1 = [\sigma_+ + \sigma_-]$, $\sigma_2 = -i[\sigma_+ - \sigma_-]$, $\sigma_3 = |\psi_2\rangle\langle\psi_2| - |\psi_1\rangle\langle\psi_1|$ is the inversion operator for the two-level system, $\hat{a}$ ($\hat{a}^\dagger$) annihilates (creates) a photon of energy $\hbar\omega$ in the Fock space, $g = \sqrt{\frac{\omega}{2\hbar\epsilon_0 V}}\langle\psi_1|\vec{d}\cdot\hat{e}|\psi_2\rangle$ is the coupling constant (assumed real) for the light-matter interaction, $\hat{e}$ is the unit vector for the polarization of the cavity field, and V is the effective volume of the cavity. It should be mentioned that the operators σ_1, σ_2, and σ_3 obey Pauli spin algebra and the operators $\hat{a}$ and $\hat{a}^\dagger$ obey the bosonic commutation relation ($[\hat{a}, \hat{a}^\dagger] = 1$). The Jaynes-Cummings Hamiltonian is explicitly time-independent in the Schrödinger picture because the quantized electromagnetic field is not treated as an external object with given time dependence [17].

The Jaynes-Cummings Hamiltonian describes the Jaynes-Cummings model. The first term ($\frac{1}{2}\hbar\omega_0\sigma_3$) of the Jaynes-Cumming Hamiltonian represents the bare Hamiltonian of the two-level system, the second term ($\hbar\omega\hat{a}^\dagger\hat{a}$) of the Jaynes-Cummings Hamiltonian represents the bare Hamiltonian of the (second) quantized electromagnetic field, and the third term ($-i\hbar g[\sigma_+\hat{a} - \sigma_-\hat{a}^\dagger]$) of the Jaynes-Cummings Hamiltonian represents the light-matter interaction for the two-level system. The first two terms together form the bare part (i.e. non-interacting part) of the Jaynes-Cummings Hamiltonian. It should be mentioned in this regard that the Jaynes-Cummings model is a purely quantum mechanical model because the atomic/molecular part of the Hamiltonian is treated quantum mechanically, the electromagnetic field part of the Hamiltonian is treated quantum field theoretically, and the light-matter interaction part of the Hamiltonian is also treated quantum field theoretically. The Jaynes-Cumming model is a model for cavity quantum electrodynamics. While the first term of the interaction part corresponds to an absorption (annihilation) of a photon by the two level system, the second term of the interaction part corresponds to an emission (creation) of a photon from the two-level system. Schematic diagram for the fundamental processes related to the Jaynes-Cummings model is shown in figure 1.9. It should also be mentioned in this regard that if the position of the two-level system in the cavity becomes significant, then the coupling constant has to be modified. If the cavity-mirrors are situated at $z = 0$ and $z = L$ on the z-axis and the two-level system is found at $z = z_0$, then the coupling constant would be modified to as $g = \sqrt{\frac{\omega}{\hbar\epsilon_0 V}}\sin(kz_0)\langle\psi_1|\vec{d}\cdot\hat{e}|\psi_2\rangle$ where $k = \omega/c$ is the wave-number. The fundamental processes related to the Jaynes-Cummings model are shown in figure

1.9. Jaynes-Cummings model is mainly used for the theoretical explanation of the quantum Rabi oscillations of a two-level system in an optical cavity [8]. It is also used to describe the phenomena of the collapse and the revival of the quantum Rabi oscillations [8].

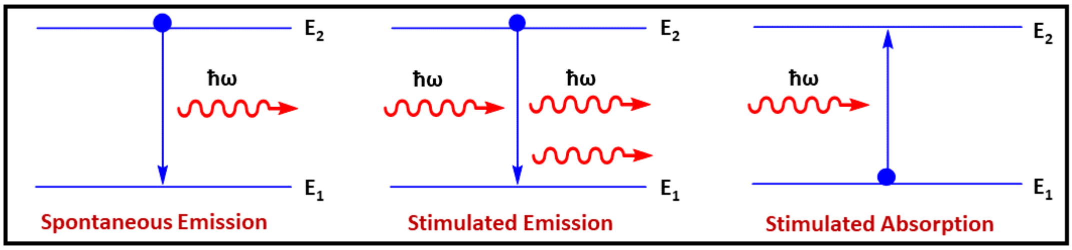

FIGURE 1.9: Schematic diagrams for the fundamental processes related to the Jaynes-Cummings model.

It is to be noted that the light-matter interaction part of the Hamiltonian commutes with non-interacting part (i.e. bare part) of the Hamiltonian. Thus the eigenstates of the Jaynes-Cummings Hamiltonian can be written as linear combination of the bare states $(|n,\psi_2\rangle, |n+1,\psi_1\rangle)$ [8, 15, 32]. Bare states representation of the Hamiltonian for n photons, however, is read as [15, 32]

$$\begin{aligned}\hat{H}_n &= \begin{bmatrix} \langle n,\psi_2|\hat{H}|n,\psi_2\rangle & \langle n+1,\psi_1|\hat{H}|n,\psi_2\rangle \\ \langle n,\psi_2|\hat{H}|n+1,\psi_1\rangle & \langle n+1,\psi_1|\hat{H}|n+1,\psi_1\rangle \end{bmatrix} \\ &= \begin{bmatrix} \frac{\omega_0}{2}\hbar + n\hbar\omega & +i\hbar g\sqrt{n+1} \\ -i\hbar g\sqrt{n+1} & -\frac{\omega_0}{2}\hbar + (n+1)\hbar\omega \end{bmatrix}. \end{aligned} \tag{1.57}$$

Here-from we get the energy eigenstates (as well as the dressed states) of the two-level system, as

$$|n,+\rangle = \cos(\theta_n/2)\,|n,\psi_2\rangle + \sin(\theta_n/2)\,|n+1,\psi_1\rangle \tag{1.58}$$

and

$$|n,-\rangle = -\sin(\theta_n/2)\,|n,\psi_2\rangle + \cos(\theta_n/2)\,|n+1,\psi_1\rangle \tag{1.59}$$

where $\tan(\theta_n) = -\frac{2g\sqrt{n+1}}{\omega-\omega_0}$ follows from the orthonormality of the dressed states [32]. Energy eigenvalues corresponding to the dressed states are given by[2] [15, 32]

$$E_\pm = \hbar\omega(n+\frac{1}{2}) \pm \frac{\hbar\Omega_n}{2} \tag{1.60}$$

where $\Omega_n = \sqrt{(\omega-\omega_0)^2 + 4g^2(n+1)}$ is the generalized n-photon Rabi frequency. Here E_+ is the energy eigenvalue of the dressed state $|n,+\rangle$ and E_- is the energy eigenvalue of the dressed state $|n,-\rangle$. Let us now express the bare states in terms of

[2]Here, the upper sign corresponds to $|n,+\rangle$ and the lower sign corresponds to $|n,-\rangle$.

the dressed states, as

$$|n, \psi_2\rangle = \cos(\theta_n/2)\,|n,+\rangle - \sin(\theta_n/2)\,|n,-\rangle \tag{1.61}$$

and

$$|n+1, \psi_1\rangle = \sin(\theta_n/2)\,|n,+\rangle + \cos(\theta_n/2)\,|n,-\rangle \tag{1.62}$$

It is clear from the diagonal elements of the matrix in Eqn. (1.57) that the energy eigenvalues of the bares states $|n+1, \psi_1\rangle$ and $|n, \psi_2\rangle$ are

$$E_{\pm}^{(0)} = (n+1/2)\hbar\omega \pm \hbar[\omega - \omega_0]/2 \tag{1.63}$$

where the upper sign $(+)$ is applicable for the bare state $|n+1, \psi_1\rangle$ and the lower sign $(-)$ is applicable for the bare state $|n, \psi_2\rangle$. Energy eigenvalues of the dressed states and the bare states are plotted with respect to the detuning parameter in figure 1.10. It is clear from the figure that the energy eigenvalues of the bare states cross each other at the resonance ($\omega = \omega_0$). This crossing, however, is avoided for the energy eigenvalues of the dressed states. It is clear from Eqn. (1.60) and Eqn. (1.63) that the dress state energy eigenvalues reach the bare state energy eigenvalues in the limiting case of $g \to 0$.

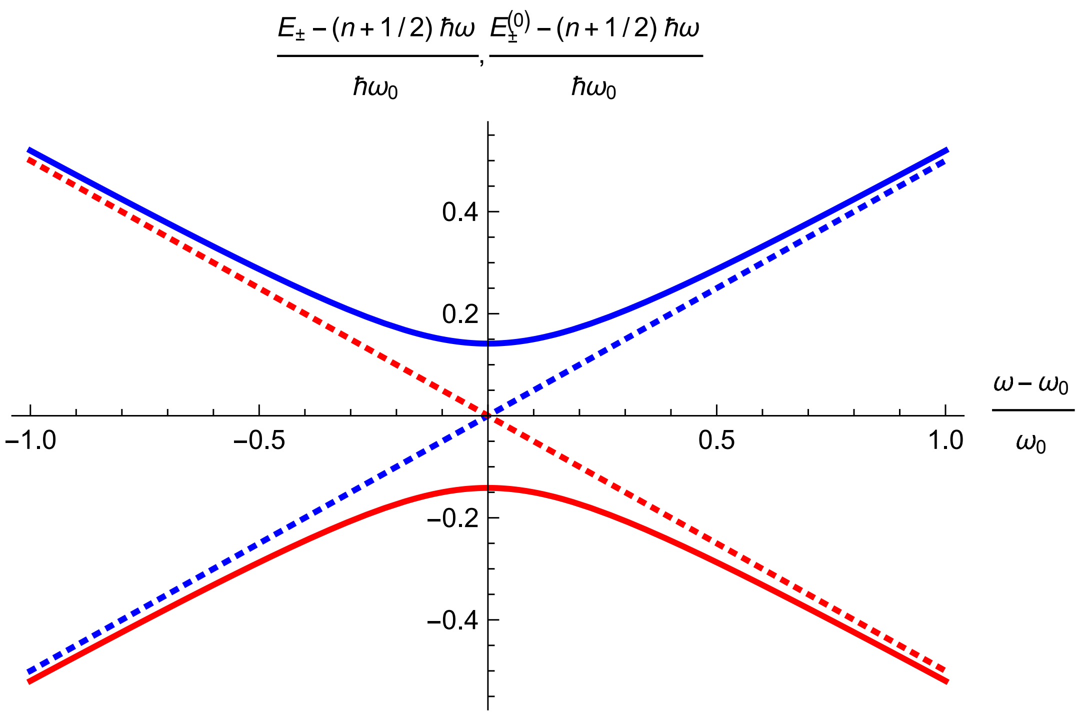

FIGURE 1.10: Energy eigenvalues of the dressed states and the bare states. The upper solid line ($[E_+ - (n+1/2)\hbar\omega]/\hbar\omega_0$) and the lower solid line ($[E_- - (n+1/2)\hbar\omega]/\hbar\omega_0$) follow Eqn. (1.60) for $g/\omega_0 = 0.1$ and $n = 1$. The right upper dotted line ($[E_+^{(0)} - (n+1/2)\hbar\omega]/\hbar\omega_0$) and the right lower dotted line ($[E_-^{(0)} - (n+1/2)\hbar\omega]/\hbar\omega_0$) follow Eqn. (1.63) for $n = 1$.

The quantum mechanical state $|\psi(t)\rangle$ of the two-level system follows Schrödinger equation for its time evolution. Thus if the system initially ($t = 0$) were in the upper state with n photons, then its time evolution takes place, as $|\psi(t)\rangle = \mathrm{e}^{-i\hat{H}t/\hbar}|n,\psi_2\rangle = \mathrm{e}^{-i\hat{H}t/\hbar}[\cos(\theta_n/2)|n,+\rangle - \sin(\theta_n/2)|n,-\rangle]$. Since the dressed states are stationary states, we can write

$$|\psi(t)\rangle = \cos(\theta_n/2)\mathrm{e}^{-iE_+t/\hbar}|n,+\rangle - \sin(\theta_n/2)\mathrm{e}^{-iE_-t/\hbar}|n,-\rangle\,. \tag{1.64}$$

Here-from we get the transition probability to the ground state with $n+1$ photons, as [18, 8, 15]

$$\begin{aligned} P_{2\to1}^{n\to n+1} &= |\langle n+1,\psi_1|\psi(t)\rangle|^2 \\ &= |[\sin(\theta_n/2)\langle n,+| + \cos(\theta_n/2)\langle n,-|][\cos(\theta_n/2)\mathrm{e}^{-iE_+t/\hbar}|n,+\rangle \\ &\quad - \sin(\theta_n/2)\mathrm{e}^{-iE_-t/\hbar}|n,-\rangle]|^2 \\ &= 4g^2\times(n+1)\times\frac{\sin^2\left(\frac{\sqrt{(\omega-\omega_0)^2+4g^2(n+1)}t}{2}\right)}{(\omega-\omega_0)^2+4g^2(n+1)}. \end{aligned} \tag{1.65}$$

This is the Jaynes-Cummings model result for the transition probability $P_{2\to1}^{n\to n+1}(t)$ for the initial condition that the two-level system initially ($t = 0$) was in the excited state $|\psi_2\rangle$. This transition probability can be recast as

$$P_{2\to1}^{n\to n+1}(t) = \frac{4g^2[n+1]}{\Omega_{n,\Delta}^2}\sin^2\left(\frac{\Omega_{n,\Delta}t}{2}\right) \tag{1.66}$$

where $\Omega_{n,\Delta} = \sqrt{\Delta^2 + 4g^2(n+1)}$ is generalized n-photon (quantum) Rabi frequency and $\Delta = (\omega - \omega_0)$ is the detuning parameter. At the resonance ($\omega \to \omega_0$), the generalized n-photon quantum Rabi frequency simply becomes the n-photon Rabi frequency $\Omega_n = 2g\sqrt{n+1}$. It is also to be noted that this transition (emission) probability is non-zero even if $n = 0$ i.e. even if no photons are incident on the two-level system. This nontrivial result for $n = 0$ can not be obtained from the semiclassical Rabi model as described in Section 1.9.1. Quantum vacuum fluctuations which are compatible with the Jaynes-Cumming model are necessary to get this non trivial result. While the transition $|n,\psi_2\rangle \to |n+1,\psi_1\rangle$ for $n = 0$ would be called as spontaneous emission, the transition $|n,\psi_2\rangle \to |n+1,\psi_1\rangle$ for $n \neq 0$ would be called as stimulated emission. On the other hand, the transition $|n+1,\psi_1\rangle \to |n,\psi_2\rangle$ would be called as stimulated absorption. When there is no field inside the cavity, i.e. $n = 0$, the transition probability in Eqn. (1.66) oscillates with the 0-photon Rabi frequency $\Omega_0 = 2g$ at the resonance. Such an oscillation is known as vacuum Rabi oscillation [11, 13, 14]. The semicalssical Rabi model can not, of course, explain the vacuum Rabi oscillation.

1.10 Collapse and revival

The Jaynes-Cummings model as described in Section 1.9.2 is able to describe the quantum Rabi oscillations of a two-level system (atom/molecule) in an optical cavity. The quantum Rabi oscillations, however, are most prominent at the resonance ($\omega \to \omega_0$). The optical cavity is often tuned with the resonant frequency and becomes a resonant cavity. There are many quantum Rabi oscillations of the transition probabilities (Eqn. (1.66)) with different n-photon Rabi frequencies in a single two-level system. Collapse of the Rabi oscillations takes place when they interfere destructively. The quantum Rabi oscillations for different photons numbers dephase in this situation with the phase difference π. On the other hand, revival of the Rabi oscillations takes place when they interfere constructively. The quantum Rabi oscillations for two consecutive photons numbers n and $n+1$ rephase in this situation with the phase difference 2π.

The collapse and the revival of the quantum Rabi oscillations have been observed for the case of the injection of the coherent electromagnetic field to a two-level system (atom) in a lossy resonant cavity [33]. Observation of the collapse and the revival, however, were first reported for the investigation of the dynamics of the interaction of a single Rydberg atom with the resonant mode of an electromagnetic field in a superconducting cavity [34]. The phenomena of the collapse and the revival become even more interesting if a large number of coherent photons are injected to a two-level system in a resonant cavity [35]. A theory for the collapse and the revival was proposed in this regard by Eberly *et al* [36] even before the experiment was carried out by Rempe *et al* [34]. We describe the theory given by Eberly *et al* [36] in the following.

For the description of the collapse and the revival of the quantum Rabi oscillations we take the Jaynes-Cumming model result for the transition probability obtained in Eqn. (1.66). We take $\Delta = 0$ for the consideration of the light-matter coupling for only the resonant mode ($\omega = \omega_0$). We further have to consider the statistical weight of the n-photon Rabi oscillation. The statistical weight can be determined from the Poisson distribution $p_n = \frac{\bar{n}^n e^{-\bar{n}}}{n!}$ where p_n represents the probability of occupation of n coherent photons for average number of coherent photons $\bar{n}$. Thus we get the net transition (emission) probability from Eqn. (1.66) as [36]

$$P_{2\to1}(t) = \sum_{n=0}^{\infty} \frac{\bar{n}^n e^{-\bar{n}}}{n!} \sin^2\left(gt\sqrt{n+1}\right). \tag{1.67}$$

It should be mentioned in this regard that we have considered the two-level system to be in the excited state ($|\psi_2\rangle$) at time $t = 0$ while deriving the above equation. In spite of large amount of simplicity of the Jaynes-Cumming model, the net transition probability $P_{2\to1}(t)$ is a sum of infinite series [36]. This tells us that the net transition probability varies with various different n-photon Rabi frequencies $\Omega_n = 2g\sqrt{n+1}$. Another important parameter related to the dynamics of

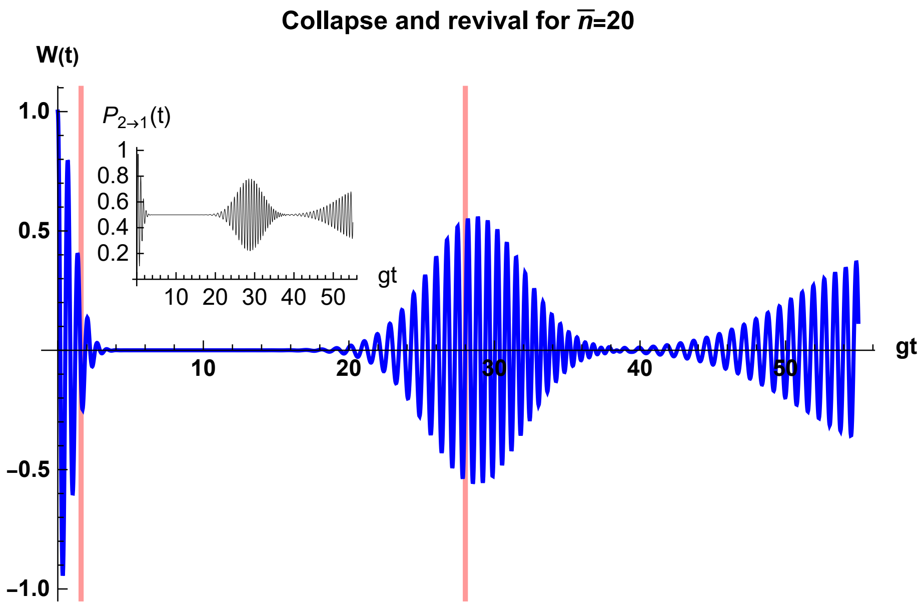

FIGURE 1.11: Collapse and revival of the quantum Rabi oscillations of a two-level system in a resonant cavity. The plot follows Eqn. (1.68) for the average number of injected coherent photons $\bar{n} = 20$. The plot in the inset follows Eqn. (1.67) for the same average number of injected coherent photons.

the Jaynes-Cummings model is the population inversion $W(t) = P_{1\to2}(t) - P_{2\to1}(t)$ where $P_{1\to2}(t) = 1 - P_{2\to1}(t)$ is the net transition probability from the ground state ($|\psi_1\rangle$) to the excited state ($|\psi_2\rangle$) [36]. Thus we get the population inversion from Eqn. (1.67) as [11, 14, 36, 37]

$$\begin{aligned} W(t) &= \sum_{n=0}^{\infty} \frac{\bar{n}^n e^{-\bar{n}}}{n!} [\cos^2(gt\sqrt{n+1}) - \sin^2(gt\sqrt{n+1})] \\ &= \sum_{n=0}^{\infty} \frac{\bar{n}^n e^{-\bar{n}}}{n!} \cos(2gt\sqrt{n+1}) \end{aligned} \tag{1.68}$$

While the net transition probability $P_{2\to1}(t)$ ranges from 0 to 1, the population inversion $W(t)$ oscillates ranges from $+1$ to -1. However, both the population inversion $W(t)$ and the net transition probability $P_{2\to1}(t)$ describe the collapse and the revival of the quantum Rabi oscillations [36, 33].

We know that the peak of the Poisson distribution takes place at $n = \bar{n}$ for $\bar{n} \gg 1$. The most significant contribution to the quantum Rabi frequency comes from the symmetric spread $2\triangle n$ of n about the peak at $n = \bar{n}$ [38]. The symmetric spread $2\triangle n$ is twice of the standard deviation $\triangle n$. For the Poisson distribution the standard deviation is $\triangle n = \bar{n}^{1/2}$. Therefore the dephasing condition of the quantum Rabi oscillations can be set for the collapse time t_c as [38]

$$\begin{aligned} & [\Omega_{\bar{n}+\Delta n} - \Omega_{\bar{n}-\Delta n}]t_c = \pi \\ \text{or} \quad & 2g[(\bar{n}+1+\sqrt{\bar{n}})^{1/2} - (\bar{n}+1-\sqrt{\bar{n}})^{1/2}]t_c = \pi. \end{aligned} \tag{1.69}$$

From this equation we estimate the collapse time as $t_c \simeq \frac{\pi}{2g}$ for $\bar{n} \gg 1$. On the other hand, the rephasing condition of the quantum Rabi oscillations can be set for the revival time t_r as [38, 39, 35]

$$\begin{aligned} & [\Omega_{\bar{n}+1} - \Omega_{\bar{n}}]t_r = 2\pi \\ \text{or} \quad & 2g[(\bar{n}+2)^{1/2} - (\bar{n}+1)^{1/2}]t_r = 2\pi. \end{aligned} \tag{1.70}$$

From this equation we estimate the revival time as $t_r \simeq \frac{2\pi\sqrt{\bar{n}}}{g}$ for $\bar{n} \gg 1$.

We plot the population inversion (Eqn. (1.68)) in figure 1.11 for $\bar{n} = 20$. The collapse time $t_c \simeq \frac{\pi}{2g}$ is indicated by a vertical line at $gt \simeq 1.6$. The revival time $t_r \simeq \frac{2\sqrt{n}\pi}{g}$ is indicated by another vertical line at $gt \simeq 28$. We also plot the net transition probability (Eqn. (1.67)) in the inset of the same figure for the same parameters. It is clear from the inset that the collapse and the revival are also captured by the net transition probability.

In the (semi) classical limit we have $V \to \infty$. This implies that $g = \sqrt{\frac{\omega}{2\hbar\epsilon_0 V}}\langle\psi_1|\vec{d}\cdot\hat{\epsilon}|\psi_2\rangle \to 0$. We also have $\bar{n} \to \infty$ in the classical limit so the Rabi frequency $\Omega_R = 2g\sqrt{\bar{n}+1} \simeq 2g\sqrt{\bar{n}}$ remains constant. Since Ω_R is a constant and $g \to 0$, the collapse time $t_c \simeq \frac{\pi}{2g} \to \infty$. This result indicates that the collapse and the revival phenomena are absent in the classical limit.

1.11 Purcell effect

The most interesting feature of the cavity quantum electrodynamics is that, the spontaneous emission from excited atoms or molecules can be greatly suppressed or enhanced by placing them in mirrors or in cavities, such as Fabry-Perot cavity, by virtue of the Purcell effect which is nothing but an enhancement or suppression of the rate of the spontaneous emission (A) from the atoms or molecules [20, 21]. Experimentalists engineer the vacuum inside the cavity to observe the Purcell effect [21]. Mode quality factor (Q) of resonant cavity plays an important role in this regard. Let us now briefly describe the Purcell effect [20].

The rate of spontaneous emission in the free space, often called as the Einstein A coefficient, follows from the exponential decay $N_2(t) = N_2(0)\mathrm{e}^{-At}$ of the population ($N_2(t)$) of the excited state of a two-level system interacting with the quantized electromagnetic field in the vacuum. In 1946 Purcell described that the spontaneous emission probability of a two-level system coupled to a resonant optical cavity is increased over its bulk value (A) by a factor [20]

$$F_P = \frac{3(2\pi c/\omega_0 n)^3 Q}{4\pi^2 V} \tag{1.71}$$

where ω_0 is the Bohr frequency of the two-level system, V is the effective volume (also called as mode volume) of the cavity, and n is the refractive index of the medium inside the cavity. The enhanced value of the spontaneous emission rate of a two-level system in the resonant cavity of the mode quality factor Q can be given by

$$A^{cavity}_{enhanced} = A \times F_P \tag{1.72}$$

where $A = \frac{\omega_0^3 |\vec{d}_{21}|^2}{3\pi\epsilon_0 \hbar c^3}$ is the Einstein A coefficient [2] of the two-level system in the free space. For cavities with very large quality factor, the enhanced spontaneous rate inside the cavity increases linearly with the quality factor Q [14]. A requirement to observe enhanced spontaneous emission is that the cavity mode volume $(V \sim (\frac{2\pi c}{\omega_0})^3)$ must be small in size so that the quantum (vacuum) fluctuations strongly affect the two-level system [7]. The experimental observation of the Purcell effect was made in 1987 by Heinzen [40] for visible spontaneous emission from atoms. The Purcell effect was also later observed in a microscopic optical cavity and a microcavity [41, 42].

The Purcell factor can be heuristically derived as follows. The density of states of photons in the resonant cavity around the resonance frequency $\omega = \omega_0$ is given by $\rho_c \sim \frac{1}{V \triangle\omega}$ [43] where $\triangle\omega$ is the spread of the frequency around the resonance. The density of states of photons at around the resonance in the bulk medium, on the other hand, is $\rho_f = \frac{\omega_0^2 n^3}{\pi^2 c^3}$ [43]. The mode quality factor is defined as $Q = \frac{\omega_0}{\triangle\omega}$ [43]. Thus we get the Purcell factor as $F_P = \frac{\rho_c}{\rho_f} \sim \frac{\pi^2 (c/\omega_0 n)^3 Q}{V}$. The numerical pre-factors are missing due to the heuristic derivation. Detailed derivation of the Purcell factor is found in Ref. [43].

While for $F_P > 1$ the spontaneous emission rate is enhanced, for $F_P < 1$ the spontaneous emission rate is inhibited or suppressed. If the dimension of cavity is small than the atomic transition wavelength, then the rate of spontaneous emission would be inhibited [44]. Such an inhibition was experimentally observed by Hulet, Hilfer, and Kleppner in 1985 [45].

1.12 Experiments on two-level systems

We already have mentioned in connection with the Einstein A and B coefficients that the stimulated emission is a fundamental process undergone on a two-level system such that the system transits from its excited state ($|\psi_2\rangle$) to its ground state ($|\psi_1\rangle$) once a photon incidents on it and the system emits a photon. The characteristics of the emitted photon in stimulated emission have phase, polarization, frequency and direction of propagation similar to the incident photon of the electromagnetic field [4]. These characteristics raise the possibility of the light amplification i.e. if there is a bottle of two-level systems (atoms/molecules), all in the excited state, and the bottle is triggered with a single photon, then a chain reaction may take place. The first photon would produce two photons, these two photons would produce four photons, and so on [4]. This is the principle behind both the microwave amplification by stimulated emission of radiation (MASER) and the light amplification by stimulated emission of radiation (LASER) [4]. Ladenburg confirmed the existence of stimulated emission phenomena in 1928 [46]. Fabrikant predicted the use of stimulated emission to amplify short waves in 1939 [46]. Kastler proposed the method of optical pumping, as a consequence of stimulated absorption in three or four level systems, in the early 1950s [47]. Weber proposed the idea to use stimulated emission phenomena to make microwave amplifier in 1951 [48]. Using Weber's idea, in 1953, Townes, Gordon, and Zeiger invented the first microwave amplifier device which amplify microwave radiation ($\lambda \sim (1mm - 1m)$) popularly known as MASER (microwave amplification by stimulated emission of radiation) [46]. This MASER was incapable to produce continuous output. This problem was sorted out by Basov and Prokhorov using more than two energy levels. The LASER was eventually invented by Maiman in 1960 for the radiation in the visible (higher frequency) range [46]. However, both the LASER and the MASER need three-level or higher level systems (atoms/molecules) to make the optical pumping in action.

1.12.1 Micromaser

A single-atom maser is called as a micromaser [11]. The development of the micromaser eases detailed study of the light-matter interactions for a two-level system (atom) in a resonant cavity [11]. The micromaser was first realized in a superconducting cavity by Meschede, Walther, and Müller in 1985 [49]. In a micromaser set up, a large number of two-level atoms are injected into a high Q superconducting cavity so that only one atom is present inside the cavity at any instant of time. This, of course, depends on the rate of injection of the two-level system. Higher value of the mode quality factor (Q) of the cavity guarantees the atom-field interaction time to be much less than the radiation decay time. This allows a two-level system to strongly couple to the cavity field through periodic energy exchanges between the cavity field and the two-level atom [11].

Micromaser captures the cavity quantum electrodynamics (CQED) for the light-matter interactions for a two-level system (atom) in a high Q resonant cavity [21]. The photon statistics of a micromaser has interesting applications towards generation of number states, quantum measurement, manipulation of atoms, quantum information, quantum computation, *etc* [11].

1.12.2 Rydberg atoms

The two-level systems in a micromaser setup are often realized with the Rydberg atoms. A Rydberg atom is an ordinary atom where one of its electrons, say the valence election, is excited to the very high principal quantum number. The state of an atom with a very high principal quantum number is called as the Rydberg state. Alkali atoms of very high principal quantum number, such as Rubidium atom (^{87}Rb) of the principal quantum number $n = 50$, are often chosen to be as Ryaberg atoms.

The electronic binding energy of a Rydberg state is given by [14]

$$E_{n,l} = -\frac{R_H}{[n-\delta_l]^2} \tag{1.73}$$

where $R_H \simeq 13.6$ eV is the Rydberg constant, $n = 1, 2, 3, ...$ is the principal quantum number, $l = 0, 1, 2, ..., n-1$ is the azimuthal quantum number, and δ_l is the "quantum defect" due to the hydrogenic "core". Here the "quantum defect" δ_l corrects for the deviation of the binding energy from the purely hydrogenic situation and δ_l is small for high l. The Rydberg state, for which l takes the highest value ($l = n-1$) and the magnetic quantum number m takes the highest or the lowest value ($|m| = n-1$), is called as the circular Rydberg state. A Rydberg atom in the Circular Rydberg states with only the principal quantum numbers n and $n+1$ can be realized as a two-level system. The electric dipole transition is allowed between these two states according to the selection rules $\triangle l = \pm 1$ and $\triangle m = \pm 1$ because the principle quantum number, the azimuthal quantum number, and the magnetic quantum number changes as $n \leftrightarrow n+1$, $n-1 \leftrightarrow n$, and $\pm(n-1) \leftrightarrow \pm n$, respectively [14]. Circular Rydberg states are used in the micromaser setups for the experiments on the cavity quantum electrodynamics (CQED) [33].

There are several reasons to consider the circular Rydberg atoms for CQED experiments. A few reasons are as follows. Only one electric dipole transition is allowed, $n-1 \leftrightarrow n$, $|m-1| \leftrightarrow |m|$, so that such states closely approximate to a two-level system [14]. The Bohr radius of a Rydberg atom scales as $a_0 n^2$. The transition dipole moment $d_{21} = \langle \psi_2 | \vec{d} \cdot \hat{k} | \psi_1 \rangle$ for such a case becomes $d_{21} \sim e n^2 a_0$. For $n = 1$, we have $d_{21} \sim 1390$ A.U. ($ea_0 = 1$ A.U.) [14]. Such a transition dipole moment is about 300 times larger than the transition dipole moment for a typical optical transition [14]. Incidentally, the light-matter coupling constant is proportional to the transition dipole moment. Thus circular Rybderg states are considered for strong light-matter coupling. The Bohr frequency for the above electric dipole transition takes the form $\omega_0 = [E_{n+1,n} - E_{n,n-1}]/\hbar \simeq \frac{2R_H}{\hbar n^3}$ for $n \gg 1$. For $n \sim 50$, the (angular) frequency of the emitted radiation becomes $\omega \sim \omega_0 \sim 6$ GHz. This frequency corresponds to the wavelength $\lambda = \frac{2\pi c}{\omega_0} \sim 8$ mm. Here-from one can estimate the experimentally achievable dimension of the resonant cavity to support a standing microwave field. Another reason for considering the circular Rydberg atom for the CQED experiment is that the circular Rydberg states have low spontaneous emission rate and long excited state lifetime ($\tau = \frac{1}{A} = \tau_0 n^5$) [14, 50, 51]. For $n \sim 50$ we get $\tau \sim 10^{-1}$s which is a very long life time in comparison to an ordinary life time $\tau_0 \sim 10^{-9}$ (corresponding to $n = 1$) [14]. Another advantage with the circular Rydberg atom is that it can be selectively ionized by an applied electric field to achieve selective state detection [14].

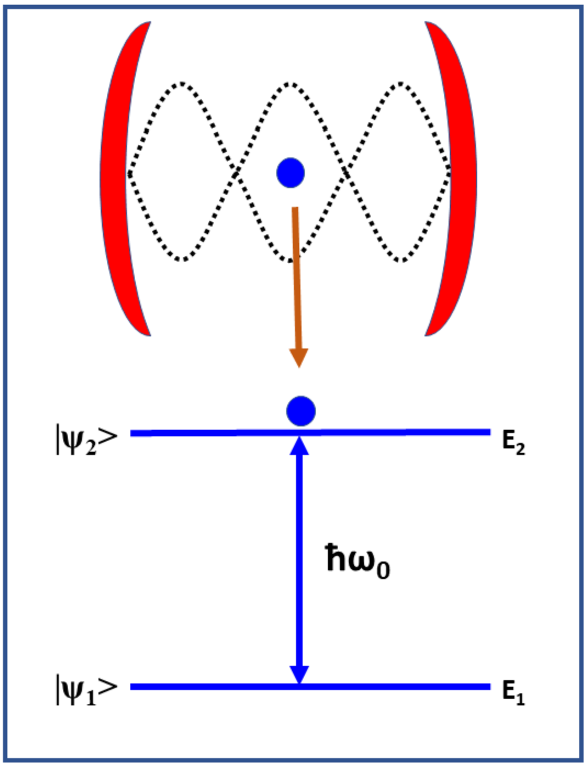

FIGURE 1.12: Schematic diagram for the cavity quantum electrodynamics. A two-level system (atom/molecule) (blue circle) in an optical cavity (red walls) is interacting with a cavity mode (black dotted lines).

1.12.3 A few experimental results on the cavity quantum electrodynamics

Cavity quantum electrodynamics (CQED) is the study of the light-matter interactions in an optical cavity where quantum nature of both the matter (e.g. two-level atom) and the electromagnetic field is significant. The most interesting feature of the cavity quantum electrodynamics is that, the spontaneous emission from an excited atom or a molecule can be greatly suppressed or enhanced by placing it inside an optical cavity, by virtue of the Purcell effect [20, 21]. Micromaser captures the cavity quantum electrodynamics (CQED) for the light-matter interactions for a two-level system in a high Q resonant cavity [21]. A Rydberg atom in the superposition of its two circular states is considered as the two-level system in this regard [33]. A schematic diagram for a model of the CQED is shown in figure 1.12. There have been many experimental investigations on the CQED since late 1980s [21, 52].

The 2012 Nobel Prize for Physics was shared by Haroche and Wineland "for ground-breaking experimental methods that enable measuring and manipulation of individual quantum systems" [52, 53]. A significant part of these experimental methods is subject to the CQED [52].

Vacuum Rabi oscillation, collapse and revival of the quantum Rabi oscillations, *etc* of the net transition probability ($P_{2\to1}(t)$) of a two-level atom were observed as results of experimental studies of the CQED [33]. Experimental data of a few of such

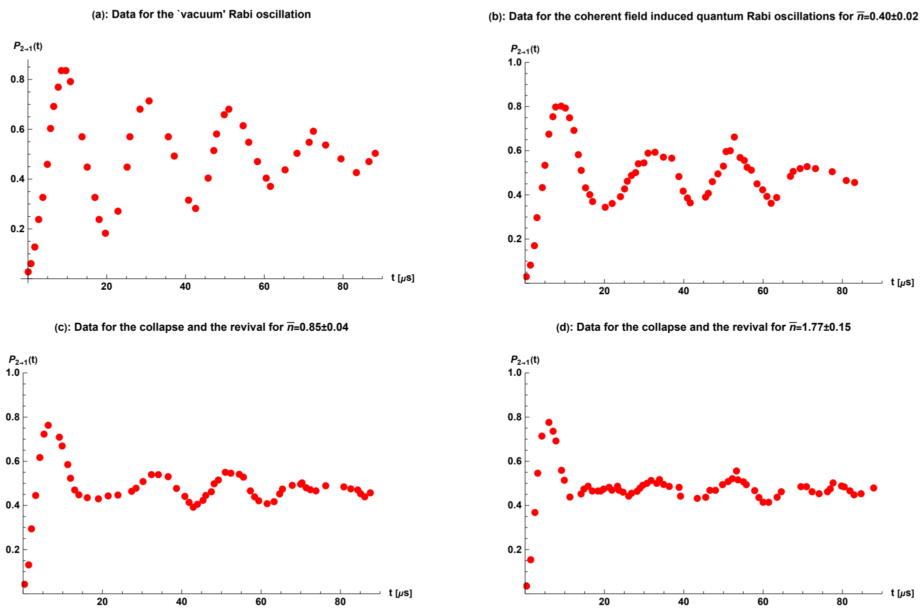

FIGURE 1.13: (a): Experimental data (circles) for the 'vacuum' Rabi oscillation of the net transition probability $P_{2\to 1}(t)$ of a two-level system for the Bohr frequency $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz, Rabi frequency $\Omega_R = 2\pi \times 47 \times 10^3$ Hz and average number of thermal photons $\bar{n} = 0.0489$. The data are adapted for the circular Rydberg states (with the principal quantum number $n = 50$ and $n = 51$) of ^{87}Rb atoms in a lossy resonant cavity of the Q-factor $Q = 7 \times 10^7$ and size $\pi(50/2)^2 \times 27$ mm^3 at the temperature $T = 0.8$ K [33].
(b): Experimental data for the coherent field induced quantum Rabi oscillations in the two-level system. The data are adapted from the same source [33] for the same parameters as mentioned above except for Rabi frequency and the average number of injected photons $\bar{n} = 0.4 \pm 0.02$ in the lossy resonant cavity.
(c): Experimental data for the coherent field induced quantum Rabi oscillations in the two-level system showing the collapse and the revival. The data are adapted from the same source [33] for the same parameters as mentioned above except for Rabi frequency and the average number of injected photons $\bar{n} = 0.85 \pm 0.04$ in the lossy resonant cavity.
(d): Experimental data for the coherent field induced quantum Rabi oscillations in the two-level system showing the collapse and the revival. The data are adapted from the same source [33] for the same parameters as mentioned above except for Rabi frequency and the average number of injected photons $\bar{n} = 1.77 \pm 0.15$ in the lossy resonant cavity.

observations on the two-level system in a lossy resonant cavity are shown in figure 1.13. These experimental data, however, have not been theoretically explained with the consideration of the multimodes into account.

1.13 Objectives

We already have discussed the failure of the 1st order time-dependent perturbation theory of quantum mechanics at around the resonance ($\omega \to \omega_0$) in Section 1.8. This failure motivates us to determine the Einstein B coefficient within a non-perturbative model, such as the Rabi model [6].

The Rabi model, however, treats the light-matter interaction classically. The Jaynes-Cummings model, on the other hand, is another exactly solvable model for light-matter interaction. This is a quantum mechanical model where light-matter interaction is treated with the quantum field theory. We are also interested to determine the Einstein A and B coefficients within the multimode Jaynes-Cummings model.

The experimental data obtained by Brune *et al* [33] for the net transition probability of a two-level system (^{87}Rb atom) in a lossy resonant cavity of a micromaser setup, have been shown in figure 1.13. While figure 1.13-a represents the data for the 'vacuum' Rabi oscillation of the two-level system, the data in 1.13-b represents the quantum Rabi oscillations of the two-level system for very low coherent field injected on it. Figure 1.13-c and figure 1.13-d, on the other hand, represent experimental data for the collapse and the revival of the quantum Rabi oscillations of the two-level system for low coherent field injected on it. These data have not been theoretically explained so far taking multimodes into account. Our aim is to explain these data within the multimode Jaynes-Cummings model [13].

The generalized Einstein coefficients lead interesting features of the occupation probabilities of a two-level system. We also aim to study the population dynamics of a two-level system in thermal radiation field with the model dependent Einstein coefficients. The population dynamics allows us to study non-equilibrium statistical mechanics of the two-level system in the thermal radiation field. We also aim to determine the entropy production of the two-level system in this regard.

Let us now mention the organization of the thesis below.

1.14 Organization of the thesis

The thesis entitled "*Effects of multimode light-matter coupling on semiclassical and quantum Rabi oscillations of a two-level system*" presents the work done by us during the last four years and six months. The thesis consists of six chapters viz., [i] *introduction*, [ii] *Rabi model result for the Einstein B coefficient*, [iii] *multimode Jaynes-Cummings model results for the Einstein A and B coefficients*, [iv] *multimode Jaynes-Cummings model results for the collapse and the revival of the quantum Rabi oscillations in a lossy resonant* *cavity*, [v] *population dynamics of two-level systems for the generalized Einstein coefficients*, [vi] *conclusions & future scope*. The main contents and the discussion pertaining to the contents are systematically presented chapter wise.

Chapter 1: Introduction

Here we begin with the motivation of writing the thesis. Then we have mentioned the key findings in the thesis and the significance of our theoretical results. Then we have described the system of interest i.e. a two-level interacting with the thermal radiation field or the coherent field or a monochromatic light. Then we have briefly described the existing models and theories necessary for the rest of the chapters of the thesis. We have briefly shown how Dirac determined the Einstein B coefficient within the first order perturbation theory of quantum mechanics. We also have briefly shown how Weisskopf-Wigner determined Einstein's A coefficient within the quantum theory of electrodynamics. We have introduced the population dynamics of the two-level systems with the help of Einstein rate equations. We have explicitly shown failure of the first order time-dependent perturbation theory of quantum mechanics. We have introduced Rabi model and Jaynes-Cummings model for the explanation of the Rabi oscillations of a two-level system both from the semiclassical point of view and the quantum mechanical point of view, respectively. We also have introduced the Purcell effect in this regard. We have introduced the collapse and revival of the quantum Rabi oscillations. Finally, we have mentioned chapter-wise organization of the thesis.

Chapter 2: Rabi model result for the Einstein B coefficient

This chapter contains Rabi model result for the Einstein B coefficient. The system of interest for this chapter is mainly a two-level system (atom/ molecule) in the thermal radiation field. Starting from the Rabi Hamiltonian, which is useful in arriving at non-perturbative results within the rotating wave approximation, we have found Einstein's B coefficient to be time-dependent: $B(t) = B_0|J_0(\Omega_R t)|$ for a two-level system in thermal radiation field. Here B_0 is the original Einstein B coefficient, Ω_R is the Rabi flopping (angular) frequency of the two level system, and J_0 is the zeroth order Bessel function of the first kind. Here the light-matter interaction is treated classically but the two-level system is treated quantum mechanically, and our result can be considered as a semiclassical result. We, of course, get back the original B coefficient in the limiting case of $\Omega_R \to 0$. We also have obtained Rabi model result for the Einstein B coefficient for a monochromatic light incident of the two-level system.

Chapter 3: Multimode Jaynes-Cummings model results for the Einstein A and B coefficients

Here we have generalized the Einstein A and B coefficients from quantum field theoretic point of view by bringing the fundamental processes and the quantum Rabi oscillation in a single footing for the light-matter interactions for nonzero Rabi frequency. We have analytically obtained multimode Jaynes-Cummings model results for the quantum Rabi oscillation of a two-level system in a lossy resonant cavity containing (i) thermal photons and (ii) injected photons of a coherent field. We have renormalized the coupling constant for the light-matter interactions for these cases. The net transition probability calculated for 'vacuum' Rabi oscillation of a two-level system in a lossy Resonant cavity matches well with the seminal experimental data obtained by Brune *et al* [Phys. Rev. Lett. **76**, 1800 (1996)]. The net transition probability calculated for the quantum Rabi oscillations of two-level system interacting with an injected coherent field in a lossy Resonant cavity also matches well with the seminal experimental data obtained by Brune *et al*.

Chapter 4: Multimode Jaynes-Cummings model results for the collapse and the revival of the quantum Rabi oscillations in a lossy resonant cavity

Here we have numerically obtained theoretical results for the collapse and the revival of the quantum Rabi oscillations for low average number of coherent photons injected on a two-level system in a lossy resonant cavity . We have adopted the multimode Jaynes-Cummings model for the same and especially treated the Ohmic losses from the cavity. We have compared our results with two sets of experimental data for low average number of coherent photons ($\bar{n} = 0.85$ and 1.77) incident on a two-level system in the lossy resonant cavity. Our results match reasonably well with the experimental data, at least, better than the theoretical one obtained for only the resonant mode and no loss from the cavity under consideration.

Chapter 5: Population dynamics of two-level systems for the generalized Einstein coefficients

Here we have studied population dynamics of two-level systems interacting with both the thermal radiation field and the monochromatic light. While the interactions of the two-level systems and the monochromatic light have been treated classically (with the Rabi model), the interactions of the two-level systems and the thermal radiation field have been treated both classically (with the Rabi model) and quantum mechanically (with the multimode Jaynes-Cummings model). For the semiclassical cases we already have obtained the generalized (time-dependent) Einstein B coefficient. For the quantum mechanical case too we already have obtained the generalized (time-dependent) Einstein A and B coefficients. We have studied the population dynamics for all these cases with the help of Einstein rate equations where the original Einstein coefficients are replaced by the generalized Einstein coefficients. The A coefficient is, of course, kept unaltered for the semiclassical cases. Time-dependence of the generalized Einstein coefficients opens a path to go beyond Pauli-von Neumann formalism of the non-equilibrium statistical mechanics. The population dynamics allows us to further study the entropy production of a level-system. For the semiclassical cases, we have shown that the Rabi oscillation can drive the two-level system away from the thermodynamic equilibrium. On the other hand, for the quantum mechanical case, we have shown that the Rabi oscillation of a small Rabi frequency (Ω_R) can not drive the two-level system away from the thermodynamic equilibrium. However, reaching the thermodynamic equilibrium is prolonged due to the quantum Rabi oscillations in the two-level system.

Chapter 6: Conclusions and future scope

Here we have briefed the conclusions of the Ph.D. works, especially the summaries. We also have mentioned the future scopes of the Ph.D. works.

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Chapter 2

Rabi model result for the Einstein B coefficient

2.1 Introduction

Einstein's A and B coefficients are quite known to the scientific community in connection with the formation of spectral lines involving the fundamental processes, such as spontaneous emission, stimulated absorption and stimulated emission, undergone on a two-level system (atom or molecule) in the presence of an oscillatory electromagnetic field, say laser light, thermal radiation, *etc.* [1]. While the A coefficient is the rate of spontaneous emission from a higher energy level to a lower energy level of the two-level system caused by vacuum fluctuations of electromagnetic field, the B coefficient is the rate of stimulated absorption B_{12} (or emission B_{21}) of (or from) the same system in the radiation field for unit energy density of the radiation per unit (angular) frequency interval around the Bohr frequency [1, 2]. Einstein's A and B coefficients are of very high importance, as because, the spectral lines have huge applications almost everywhere in the modern science, engineering, and technology. These coefficients also determine density of photons in thermal equilibrium when the probability of transitions for a two-level system reaches a steady state.

Historically, almost a century back—during the era of old quantum mechanics when time-dependent perturbation theory was not known [1], Einstein's A and B coefficients were proposed to be time-independent. These coefficients, for the two-level system in thermal radiation field at an absolute temperature T, were determined in terms of fundamental constants by Dirac, Weisskopf, and Wigner in the quantum mechanics era within (i) the frameworks of the time-dependent perturbation theory for the light-matter interactions and (ii) the quantum field theory of the stimulated emission, the stimulated absorption, and the spontaneous emission of radiation [3, 4]. However, neither Einstein's semi-classical theory of radiation [1] nor Dirac's first order quantum mechanical perturbation theory of radiation [3] predicted regularity in the stimulated transitions (absorption and emission) though the electromagnetic field incident on the two-level system oscillates in a regular manner. This regularity was predicted a decade later by Rabi [5]. He and his collaborators showed resonance in the two-level system in the course of stimulated absorption and emission within a nonperturbative model which is now known as the Rabi model [5, 6]. For the two-level system (having the electric dipole moment $\vec{d}$ and the Bohr (angular) frequency ω_0 corresponding to energy eigenstates $|\psi_1\rangle$ and $|\psi_2\rangle$) in presence of an oscillatory electromagnetic field with the electric field component $\vec{E} = \vec{E}_0 \cos(\omega t)$, Rabi *et al.* obtained a generalized (angular) frequency for the transition induced flopping of the two states, as $\Omega = \sqrt{(\omega - \omega_0)^2 + \Omega_R^2}$ which is now known as the Rabi formula where $\Omega_R = |\langle\psi_1|\vec{d}\cdot\vec{E}_0|\psi_2\rangle|/\hbar$ is the Rabi flopping frequency [6].

It it quite known, that, the generalized Rabi flopping frequency (Ω) tends to the Rabi flopping frequency (Ω_R) at resonance ($\omega \to \omega_0$), i.e., where the probability of the stimulated transitions for the stimulated emission from the initial ($t = 0$) state $|\psi_2\rangle$ to the final state $|\psi_1\rangle$ at time t, say $P_{2\to1}(t) = \Omega_R^2 \frac{\sin^2(\Omega t/2)}{\Omega^2} = \Omega_R^2 \frac{\sin^2(\sqrt{(\omega-\omega_0)^2+\Omega_R^2}t/2)}{(\omega-\omega_0)^2+\Omega_R^2}$, is sharply peaked [6, 7, 8, 9]. The expression of transition probability (involving the Rabi flopping frequency) is quite successful, as it gives a reliable value of the nuclear magnetic moment to experimentalists [6]. Later experimentalists found this expression quite successful for atoms, molecules, semiconductors, Bose–Einstein condensates, many-bodies, *etc.* exposed in laser light [10, 11, 12, 13, 14, 15, 16]. One can get the perturbation result (which is compatible with Fermi's golden rule) [3, 7] back if one assumes $|\omega - \omega_0| \gg \Omega_R$ in the Rabi formula. But, condition for the time-dependent perturbation ($P_{2\to1}(t) \ll 1 \; \forall \; t$, i.e., $\Omega_R^2/(\omega - \omega_0)^2 \ll 1$) does not hold [7] at the resonance ($\omega \to \omega_0$) at least for $t \to \infty$ however weak the light-matter coupling ($\sim \Omega_R$) may be. The problem with the upper limit of time ($0 \leq t < \infty$) can not be avoided to get the frequency matching condition ($\delta(\omega - \omega_0)$ [1]) for the dipole-transitions stimulated by a sinusoidal perturbation [3, 7]. Thus, the divergence of the transition probability questions soundness of the 1st order perturbation theory at the resonance for $t \to \infty$. The soundness can, of course, be restored only for $\Omega_R \to 0$ so that $\lim_{\Omega_R\to0,\omega\to\omega_0} \Omega_R \delta(\omega - \omega_0) = \text{constant} \lesssim 1$.

Since the condition for the sinusoidal perturbation with non-vanishing light-matter coupling ($\sim \Omega_R$) is not satisfied [7] at the resonance ($\omega \to \omega_0$) where the stimulated transitions (emission) are most probable, the first order perturbation result ($B_{12} = B_{21} = \frac{\pi}{3\epsilon_0\hbar^2}|\langle\psi_1|\vec{d}|\psi_2\rangle|^2$ [3, 7] [2]) for the B coefficient obtained by Dirac [3] is not reliable for $\Omega_R \nrightarrow 0$. This is a problem with the quantum mechanical perturbation theory, and it remains so even in the weak coupling regime ($\Omega_R/A \ll 1$) as long as the coupling ($\sim \Omega_R$) does not tend to zero keeping the natural decay rate (i.e., the A coefficient) fixed to a nonzero value. Consequently, a question arises: what

[1]The frequency matching condition follows from the limiting case of the square root of the stimulated transition probability $\lim_{t\to\infty} \sqrt{P_{2\to1}(t)} = \lim_{t\to\infty} \Omega_R \frac{\sin([\omega-\omega_0]t/2)}{\omega-\omega_0} = \Omega_R 2\pi\delta(\omega - \omega_0)$ within the 1st order sinusoidal perturbation.

[2]If degeneracy of the two states are g_1 and g_2, respectively, then B_{21}/B_{12} would be given by $B_{21}/B_{12} = g_1/g_2$ [2].

would be the reliable expression for the B coefficient in the weak coupling regime? This issue could have been addressed with a nonperturbative model such as the Rabi model [5, 6], but has surprisingly been overlooked for the last eight decades, though there are several works done in the intermediate regime ($0 \lessapprox \Omega_R/A \lesssim 1$) with the consideration of moderate system-bath (i.e., system-radiation field) interactions. These interactions are often (i) semiclassically modelled as perturbation terms in the Bloch–Redfield (Markovian master) equation within the density matrix formalism [17, 18], (ii) quantum mechanically modelled as non-perturbation terms in the Schrodinger equation within the generalized Weisskopf-Wigner (natural) decay formalism for discrete and continuum modes [19], and (iii) quantum mechanically modelled as non-perturbation terms in the Nakajima–Zwanzig-type (non-Markovian master) equation within the density matrix formalism [20, 21].

While the stimulated transition rates are time-independent in the (quantum) Markovian master equations and are solvable for the case of the time-dependent perturbation on the system [22, 18, 17], they are time-dependent in the (quantum) non-Markovian master equations and are usually hard to solve [20, 21]; the explicit time-dependent terms in the stimulated transition rates render the non-Markovian master equations analytically intractable. Of course, some simplified versions of the non-Markovian master equations can be solved either in the limiting cases of weak [23] and linear [24] interactions between the system and the bath or in the limiting case of structured bath even for strong interactions [25]. Nevertheless, population dynamics of the open quantum system of our interest, i.e., the two-level system in the thermal radiation field, has not been described so far through exact analytical solutions of the non-Markovian master equations.

The semiclassical Rabi model, we are considering, though is a non-perturbative one, gives exact results even in the weak coupling regime, as the model is exactly solvable for the entire range of coupling constant ($\sim \Omega_R$). Hence, we aim (i) to get a single reliable expression of Einstein's B coefficient from the (semiclassial) Rabi model not only for the weak coupling regime ($\Omega_R/A \ll 1$) but also for the entire regime including the strong coupling regime ($\Omega_R/A \gg 1$), (ii) to generalize Einstein's rate equations with the reliable B coefficient for the two-level system in the thermal radiation field.

The next section of this chapter begins with the Rabi model for the two-level system in a sinusoidally oscillating electromagnetic field [5, 6, 7, 8]. Then we write down transition probabilities for the electric-dipole transitions among the two (energy) levels, and recast the transition probabilities for the two-level system in the thermal radiation by integrating over all possible frequencies and polarizations of the thermal radiation field. This result significantly differs from the perturbation result. This result, however, is a semiclassical result [3]. We get oscillatory type time-dependent B coefficient, from the corresponding transition probability (i.e., the stimulated emission's probability) for the thermal photons. We also consider monochromatic radiation field side by side throughout the chapter.

2.2 Two-level system in thermal radiation field

2.2.1 Rabi model

The Rabi Hamiltonian for the two-level system having electric dipole moment $\vec{d}$ in the oscillatory electromagnetic field (with the electric field component $\vec{E} = \vec{E}_0 \cos(\omega t)$) is given by [5, 6, 8, 26]

$$\begin{aligned} H &= E_1|\psi_1\rangle\langle\psi_1| + E_2|\psi_2\rangle\langle\psi_2| \\ &\quad -\frac{\vec{E}_0 \cdot \vec{d}}{2}[\mathrm{e}^{i\omega t}|\psi_1\rangle\langle\psi_2| + \mathrm{e}^{-i\omega t}|\psi_2\rangle\langle\psi_1|] \end{aligned} \tag{2.1}$$

where $|\psi_1\rangle$ and $|\psi_2\rangle$ constitute a set of two orthonormal states of the two-sate system (in absence of the external field) with energy eigenvalues E_1 and E_2 ($E_2 > E_1$) respectively. The third term of the Hamiltonian represents the classical interaction between the two-level system (atom or molecule) and the external electromagnetic field. The interaction term, although is not a perturbation, is consistent with the rotating wave approximation ($\omega_0 + \omega \gg |\omega_0 - \omega|$) which is also used in the time-dependent perturbation theory [6, 7]. Validity of the rotating wave approximation, however, is not questioned at the resonance ($\omega \to \omega_0$) as long as $\omega_0 = (E_2 - E_1)/\hbar$ is fairly large, say $\omega_0 \gg \Omega_R$. Thus, the Rabi model is applicable for large Bohr frequency (ω_0) of the two-level system, and incidentally, the Schrödinger equation for the two-level system corresponding to the Rabi Hamiltonian is exactly solvable under the transformation into the interaction-picture [6, 7]. The Rabi model is of course an integrable one due to the presence of a discrete symmetry in it [28]. The energy eigenvalues of the Rabi Hamiltonian in Eqn. (2.1) thus takes the form $E_{\mp} = [E_2 + E_1]/2 \mp \frac{\hbar}{2}\sqrt{(\omega - \omega_0)^2 + \Omega_R^2}$ [6]. Corresponding eigenstates are now dressed due to the light-matter coupling resulting the energy eigenvalues different from those (E_1, E_2) of the uncoupled bare states $(|\psi_1\rangle, |\psi_2\rangle)$. Both the eigenstates eventually are linear combinations of the uncoupled bare states: $|\psi_-\rangle = \cos(\theta)|\psi_1\rangle + \sin(\theta)|\psi_2\rangle$ & $|\psi_+\rangle = -\sin(\theta)|\psi_1\rangle + \cos(\theta)|\psi_2\rangle$ for $\tan(\theta) = \Omega_R/[\sqrt{(\omega - \omega_0)^2 + \Omega_R^2} - (\omega - \omega_0)]$ [29].

Eventually, the two-level system will always be in the superposition state $|\psi\rangle$ of the two energy eigenstates as well as of the uncoupled bare states as long as energy of the system is not measured. Thus, time evolution of the state of the system takes

[3] Semiclassical results, in this respect, are found with the quantum mechanical treatment of the two-level system and the classical treatment of the light-matter interactions [7, 8]. Inclusion of the operator algebra for photon-annihilation and photon-creation operators would make the treatment quantum electrodynamic (QED) [27].

the form [4] $|\psi(t)\rangle = c_1(t)\mathrm{e}^{-iE_1t/\hbar}|\psi_1\rangle + c_2(t)\mathrm{e}^{-iE_2t/\hbar}|\psi_2\rangle$, where $c_1(t)$ is the transition probability amplitude for the dipole-transition from the uncoupled bare state $|\psi_2\rangle$ to $|\psi_1\rangle$ and $c_2(t)$ is that from the uncoupled bare state $|\psi_1\rangle$ to $|\psi_2\rangle$ [3, 5, 6]. Transformation of the Schrodinger equation for the two-level system corresponding to the Rabi Hamiltonian into the interaction picture results in the transition probability [6, 7, 8]

$$P_{2\to 1}(t) = |c_1(t)|^2 = \Omega_R^2 \frac{\sin^2(\Omega t/2)}{\Omega^2} \tag{2.2}$$

once we use the initial condition that the system was initially ($t = 0$) only in the state $|\psi_2\rangle$. Here, $\Omega = \sqrt{(\omega-\omega_0)^2 + \Omega_R^2} = (\omega-\omega_0)\sqrt{1+[\frac{\Omega_R}{\omega-\omega_0}]^2}$ is the generalized Rabi flopping (angular) frequency and $\Omega_R = |\langle\psi_1|\vec{d}\cdot\vec{E}_0|\psi_2\rangle|/\hbar$ is the Rabi flopping frequency for the stimulated emission from the state $|\psi_2\rangle$ to the state $|\psi_1\rangle$. Also, we keep in mind that

$$P_{1\to 2}(t) = |c_2(t)|^2 = 1 - P_{2\to 1}(t) \tag{2.3}$$

is the transition probability for the stimulated absorption from the state $|\psi_1\rangle$ to the state $|\psi_2\rangle$.

Eqs. (2.2) and (2.3) hold good for a linearly polarized monochromatic light (having energy density, $u = \frac{1}{2}\epsilon_0 E_0^2$ [5]) incident on the two-level system.

2.2.2 Rabi model result for the Einstein B coefficient

Let us now consider the system be placed at the thermal radiation field in the free space [6] where all possible frequencies of the incident light are present with two independent arbitrary polarization directions. While averaging over the polarization states there results in factor $1/3$ [7] in the r.h.s. of Eqn. (2.2), contribution of the thermal radiation of all possible frequencies leads to an integration in the r.h.s. of Eqn. (2.2) over ω with the weight-factor $u(\omega)$. This follows from Planck's distribution formula (or Bose-Einstein statistics for photons). Thus, we get the net transition probability from Eqn. (2.2) as

$$P_{2\to 1}(t) = \frac{|\langle\psi_1|\vec{d}|\psi_2\rangle|^2}{3\hbar^2}\int_0^\infty \frac{2u(\omega)}{\epsilon_0}\frac{\sin^2(\Omega t/2)}{\Omega^2}d\omega, \tag{2.4}$$

[4] It takes another form, viz., $|\psi(t)\rangle = c_-\mathrm{e}^{-iE_-t/\hbar}|\psi_-\rangle + c_+\mathrm{e}^{-iE_+t/\hbar}|\psi_+\rangle$, in the basis of the energy eigenstates with time-independent coefficients ($c_\mp$) resulting in no transitions between the dressed eigenstates $|\psi_+\rangle$ and $|\psi_-\rangle$.

[5] Actual energy density, $u = \epsilon_0 E_0^2\cos^2(\omega t)$, where magnetic field part also contributes equally is averaged out here, as because, (i) ω goes to ω_0 at the resonance, and (ii) electromagnetic field oscillates many times within a single Rabi cycle for $\omega_0 \gg \Omega_R$ [7].

[6] Here free space is an idealization of a big black-body cavity of volume $V \to \infty$. The two-level system would not come to equilibrium with the thermal radiation field in ideal free space.

where [30, 7]

$$u(\omega) = \frac{\hbar\omega^3}{\pi^2 c^3}\left[\frac{1}{e^{\hbar\omega/k_B T} - 1}\right] \tag{2.5}$$

is the average energy density of the thermal radiation (electromagnetic) field per unit (angular) frequency interval [7]. The average energy density of the thermal radiation (electromagnetic) field per unit (angular) frequency interval $u(\omega)$ in Eqn. (2.5) incidentally represents the average contribution for the thermal photons at the temperature T and is responsible for the stimulated emission.

The generalized Rabi frequency (Ω), by the definition, ranges form $-\sqrt{\omega_0^2 + \Omega_R^2}$ to $-\Omega_R$ and Ω_R to ∞ following the avoided crossing as ω varies from 0 to ω_0 and ω_0 to ∞. The integrand in Eqn. (2.4) is peaked at the resonant frequency, $\omega = \omega_0$ (i.e., at $\Omega = \pm\Omega_R$), so that most of the integration comes from ω close to ω_0. Further considering ω_0 to be fairly large (i.e., $\omega_0 \gg \Omega_R$ which is compatible with the rotating-wave approximation), we recast Eqn. (2.4) as

$$\begin{aligned} P_{2\to 1}(t) &\simeq \frac{2d_{12}^2 u(\omega_0)}{3\epsilon_0\hbar^2}\left[\int_{-\infty}^{-\Omega_R}\frac{\sin^2(\Omega t/2)}{\Omega^2}\frac{1}{\sqrt{1-(\Omega_R/\Omega)^2}}d\Omega \right.\\ &\quad \left. + \int_{\Omega_R}^{\infty}\frac{\sin^2(\Omega t/2)}{\Omega^2}\frac{1}{\sqrt{1-(\Omega_R/\Omega)^2}}d\Omega\right] \\ &= \frac{2d_{12}^2 u(\omega_0)}{3\epsilon_0\hbar^2}\frac{\pi}{2\Omega_R}\,{}_1F_2(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-\frac{\Omega_R^2 t^2}{4})\Omega_R t \end{aligned} \tag{2.6}$$

where ${}_1F_2$ is a generalized hypergeometric function [8], $d_{12} = |\langle\psi_1|\vec{d}|\psi_2\rangle| = |\langle\psi_2|\vec{d}|\psi_1\rangle|$ is the transition dipole moment, and the lower limit $\Omega = -\sqrt{\omega_0^2 + \Omega_R^2}$ (which follows from the avoided crossing) has been replaced [9] by $-\infty$ as the typical full-width of the transition probability, $\Delta\Omega = 4\pi/t$, is well contained (for reasonable values of t) within the lowest possible value ($-\sqrt{\omega_0^2 + \Omega_R^2}$) and the highest possible value (∞) of Ω for fairly large ω_0. All these approximations for evaluating the above integrations are also applied in the time-dependent perturbative calculation keeping $\Omega_R \to 0$ [3, 7]. We differ from the perturbation result [3, 7] only by keeping $\Omega_R \neq 0$.

The natural question arises about quantifying the Rabi flopping frequency for the two-level system in the thermal radiation field. Eqn. (2.6) is the generalization of Eqn. (2.2) with proper normalization for all the frequencies of the thermal radiation around ω_0. Thus for $t \to \infty$ and $\omega \to \omega_0$, the right hand sides of both Eqn. (2.2) and Eqn. (2.6) are averaged out to $1/2$ under the consideration that $u(\omega_0)$ remains fixed for stimulated emission and stimulated absorption processes. Thus, we get the Rabi

[7] Here, time averaging is taken in the very short time scale of $1/\omega_0$. Grand canonical ensemble averaging is further taken for the thermal photons.

[8] ${}_1F_2(\{1/2\},\{1,3/2\},-(1/4)a^2t^2) = \frac{1}{t}\int_0^t J_0(a\tau)d\tau$

[9] Correction to the integration, for this replacement, quickly vanishes as $[-\pi/2 + \mathrm{Si}(\omega_0 t) + O(\Omega_R^2/\omega_0^2)]t$.

flopping frequency for the two-level system in the thermal radiation field as

$$\Omega_R = \frac{2\pi d_{12}^2 u(\omega_0)}{3\epsilon_0 \hbar^2}. \tag{2.7}$$

This form of the Rabi flopping frequency is quite general for the two-level system. Now, using Eqs. (2.6) and (2.7), we get the net transition probability for the stimulated emission as

$$P_{2\to1}(t) = \frac{\Omega_R t}{2}\, {}_1F_2(\{\frac{1}{2}\}, \{1, \frac{3}{2}\}, -\frac{\Omega_R^2 t^2}{4}). \tag{2.8}$$

Eqn. (2.8) is our semiclassical result for the Rabi flopping for thermal radiation. Time-averaged transition probability was alternatively calculated within the same (semiclassical) Rabi model not for multi-frequency components rather for a single frequency component of the incident electromagnetic field decades back by Shirley [9]. His result on the time-averaged transition probability is significantly different from our net time-dependent transition probability, presented in Eqn. (2.8), where contributions of all the frequency components of the thermal radiation field are averaged out with the Planck's distribution. It needs a full quantum electrodynamic (QED) description to capture all the quantum mechanical features of the interactions of the two-level system (atom/molecule) with the thermal (as well as coherent) radiation field. Such a QED description was given long before by Jaynes and Cummings but for interactions with a single cavity mode in a resonant cavity [27]. However, our semiclassical result is still useful for interactions in the free space with the broadband modes near around the resonance frequency. It would be important in relating the Rabi model with Einstein's rate (master) equations which are useful in describing nonequilibrium phenomena in terms of the fundamental processes.

The rate of stimulated emission ($R_{2\to1}(t) = |\frac{d}{dt}P_{2\to1}(t)|$) is the modulus of the time-derivative of the net transition probability $P_{2\to1}(t)$. Here, the modulus comes into definition of the rate of the transitions because the transition probability is now time dependent and there is an oscillation in it. If the time-derivative is negative then it refers to backward transition, say stimulated absorption, whose rate may be denoted by $R_{1\to2}(t)$. By convention, we don't want to say $R_{2\to1}(t) = -R_{1\to2}(t)$, rather we want to say $R_{2\to1}(t) = R_{1\to2}(t)$ for all the time. Hence we need the modulus in the definition of the rate of the stimulated emission. Thus we get the rate of the stimulated emission from Eqn. (2.6) as

$$\begin{aligned} R_{2\to1}(t) &= \Bigg| \frac{d_{12}^2 u(\omega_0)}{3\epsilon_0\hbar^2} \Bigg[\int_{-\infty}^{-\Omega_R} \frac{\sin(\Omega t)}{\Omega} \frac{1}{\sqrt{1-(\Omega_R/\Omega)^2}} d\Omega \\ &\quad + \int_{\Omega_R}^{\infty} \frac{\sin(\Omega t)}{\Omega} \frac{1}{\sqrt{1-(\Omega_R/\Omega)^2}} d\Omega \Bigg] \Bigg| \\ &= \frac{\pi d_{12}^2 u(\omega_0)}{3\epsilon_0\hbar^2} |J_0(\Omega_R t)| \end{aligned} \tag{2.9}$$

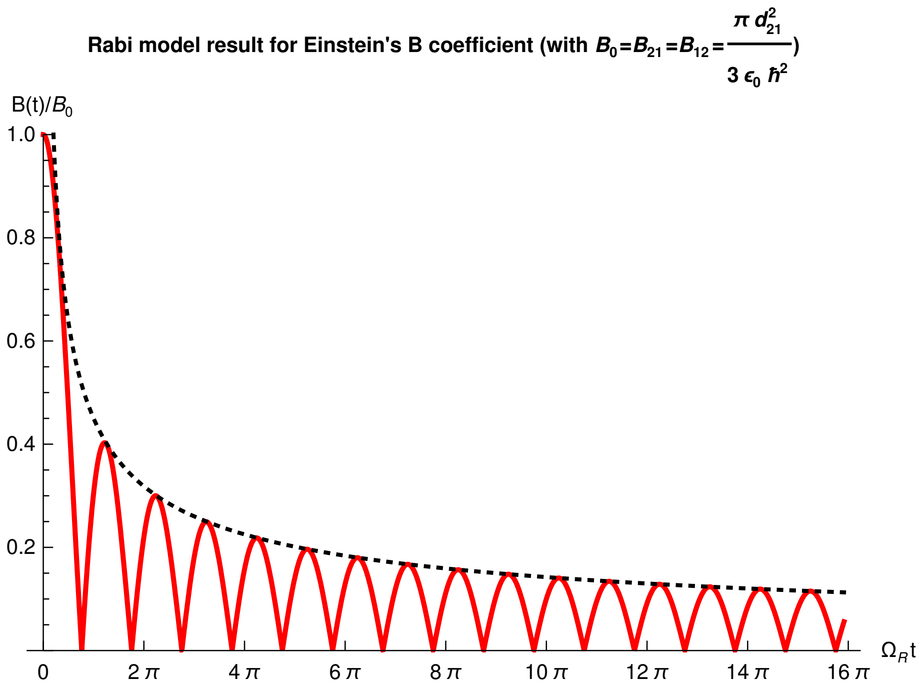

FIGURE 2.1: The solid line follows Eqn. (2.11) and represents the Rabi model result for the Einstein B coefficient. The dotted line ($B(t)/B_0 \equiv \sqrt{2/\pi\Omega_R t}$) represents envelope for the oscillations in the B coefficient.

where $J_0(\Omega_R t)$ is the Bessel function of the order zero of the first kind. The rate of absorption ($R_{1\to 2}(t) = |\frac{d}{dt}P_{1\to 2}(t)|$) from the state $|\psi_1\rangle$ to $|\psi_2\rangle$, on the other hand, is just the opposite, i.e., $R_{1\to 2}(t) = R_{2\to 1}(t)$ as $P_{2\to 1}(t) + P_{1\to 2}(t) = 1$. From Eqs. (2.3) and (2.9), it is clear that the rate of stimulated absorption from the state $|\psi_1\rangle$ to the state $|\psi_2\rangle$ would be

$$R_{1\to 2}(t) = R_{2\to 1}(t) = \frac{\pi d_{12}^2 u(\omega_0)}{3\epsilon_0\hbar^2}|J_0(\Omega_R t)|. \tag{2.10}$$

Einstein's B coefficient was already defined as the rate of stimulated transitions (emission or absorption) per unit energy density of radiations per unit (angular) frequency interval around the Bohr frequency. However, the rate of transitions in Eqn. (2.10) quasi-periodically alters its sign in the course of time as if the absorption becomes emission and vice-versa whenever there is an alternation of the sign. Thus, by following the definition of the B coefficient, we get Einstein's B coefficient ($B(t) = R_{1\to 2}(t)/u(\omega_0) = R_{2\to 1}(t)/u(\omega_0)$) from Eqs. (2.9) and (2.10), as

$$B(t) = B_0|J_0(\Omega_R t)| \tag{2.11}$$

where $B_0 = \frac{\pi d_{12}^2}{3\epsilon_0\hbar^2}$ is the original B coefficient obtained by Dirac [3, 7]. The time-dependent coefficient $B(t)$ in Eqn. (2.11) is the Rabi model result for the Einstein B coefficient. We have got this result published in Ref. [31]. The time-dependent coefficient $B(t)$ can be called as the generalized Einstein B coefficient as $B(t)$ becomes the

Einstein B coefficient ($B_0 = B_{21} = B_{12}$) in the limit $\Omega_R \to 0$. The generalized B coefficient would be unaltered if we alter the initial conditions, by taking $c_1(0) = 1$ and $c_2(0) = 0$, as we always have $R_{1\to 2}(t) = R_{2\to 1}(t)$. We show the time-dependence in the B coefficient in figure 2.1. We are not able to compare this result with the existing experimental data because they have not been obtained by any direct measurement (as far as we know); rather, experimentalists apply time-dependent perturbation theory for the indirect measurement of the B coefficient [32].

2.3 Generalized Einstein B coefficient for monochromatic radiation field

For the case of a monochromatic light (having a single polarization perpendicular to a fixed direction of propagation and time averaged [10] energy density $u = \frac{1}{2}\epsilon_0 E_0^2$) incident on the two-level system, we don't need to average over the directions of polarizations and frequencies as done in Eqn. (2.4). For such a case, we get the rate of stimulated emission (or absorption) from Eqn. (2.2) as

$$R_{2\to 1}(t) = R_{1\to 2}(t) = \left|\frac{d}{dt}P_{2\to 1}(t)\right| = \frac{d_{12}^2 u}{\epsilon_0 \hbar^2}\frac{|\sin(\Omega t)|}{\Omega}. \tag{2.12}$$

Now, if we define the B coefficient ($B(t) = R_{1\to 2}(t)/[u/\Omega] = R_{2\to 1}(t)/[u/\Omega]$) for a monochromatic wave, as the rate of the stimulated transitions (emission and absorption) per unit time average energy density per unit generalized Rabi flopping frequency, then it would be

$$B(t) = \frac{3B_0}{\pi}|\sin(\Omega t)|. \tag{2.13}$$

This is the Rabi model result for the Einstein B coefficient for a monochromatic radiation field. We show the time-dependence in this form of the B coefficient in figure 2.2. We have got this result published in Ref. [31].

2.4 Conclusion

We have shown that the Rabi model result for Einstein's B coefficient depends on time and the Rabi flopping frequency for the two-level system (atom or molecule) in the thermal radiation field at an absolute temperature T. This result is accurate for fairly large Bohr frequency ($\omega_0 \gg \Omega_R$ [11]) and fairly higher temperature ($k_B T \gtrsim \hbar\Omega_R$), and is significantly different from the perturbation result which is not reliable near the resonance in the Rabi flopping. Our analytical result regarding the

[10] Here, time averaging is taken in the very short time scale of $1/\omega_0$.

[11] This is also a requirement for the rotating wave approximation, which is inbuilt in the Rabi model, to be valid.

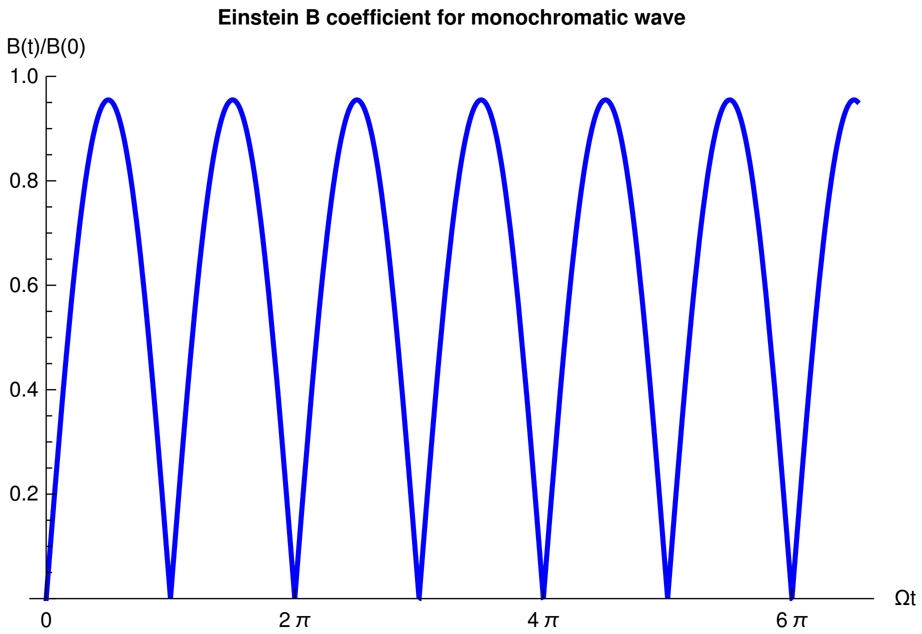

FIGURE 2.2: Plot follows Eqn. (2.13) and represents the Rabi model result for the Einstein B coefficient for a monochromatic radiation field.

B coefficient is an invitation for the experimentalists to do direct measurement of the B coefficient.

While the Rabi flopping usually is studied for strong light-matter interactions ($\Omega_R/A \gg 1$), Einstein's rate equations are usually applied for weak light-matter interactions ($\Omega_R/A \ll 1$). Incidentally, the Rabi model, which gives exact results in both the weak coupling regime and the strong coupling regime, is not phenomenologically different from the fundamental processes' point of view. We have been interested in bringing the Rabi flopping and the rate equations together in a single footing for this reason. We have been specially interested in the intermediate regime $0 \lessapprox \Omega_R/A \lesssim 1$.

The generalized Einstein B coefficient obtained in Eqn. (2.11) may take place in the time-dependent rate coefficients $R_{1\to 2}(t)$ and $R_{2\to 1}(t)$ in the Einstein rate equations if the time-independent rate coefficients are replaced by the time-dependent rate coefficients. Population dynamics would have to be studied in terms of the time-dependent rate coefficients in such a case. Such a population dynamics with the time-dependent rate coefficients would be interesting for analysing the entropy production of a two-level system in the thermal radiation field. In Chapter-5 we will study the population dynamics and entropy production for both the time-dependent forms of the B coefficient obtained in Eqns. (2.11) and (2.13).

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Chapter 3

Multimode Jaynes-Cummings model results for the Einstein A and B coefficients

3.1 Introduction

Seminal experimental work of Brune *at al* [1] regarding the quantum Rabi oscillations (or flopping) of the occupation of the two energy eigenstates of ^{87}Rb atom in a lossy resonant cavity at finite temperatures opened the possibilities of experimental [2, 3, 4] and theoretical [5] study of the cavity quantum electrodynamics (QED) specially in the field of measuring and manipulation of individual quantum systems a quarter century back [1, 6, 7, 8]. The most interesting feature of the cavity-QED is that, the spontaneous emission from excited atoms or molecules can be greatly suppressed or enhanced by placing them in mirrors or in cavities, such as Fabry-Perot cavity, by virtue of the Purcell effect [9, 2]. Experimentalists basically engineer the vacuum inside the cavity to observe the Purcell effect [2]. Mode quality factor (Q) of resonant cavity plays an important role in this regard.

A two-level system (atom or molecule) in the free space once makes a spontaneous emission, say at time $t = 0$, the emitted photon goes away from the system in an irreversible manner. The possibility that after some finite time-interval the emitted photon would be further absorbed by the two-level system, was not considered in Einstein's semiclassical description [10]. However, observation of the quantum (vacuum) Rabi oscillations [1] in the high-Q cavity reveals the fact that, the boundary conditions greatly influence the atomic radiation [11] and consequently, the emitted photon is reabsorbed by the two-level system [2, 3]. The spontaneous emission becomes reversible in an ideal[1] cavity as the two-level system and the field exchange excitation at the rate of Rabi frequency (Ω_R) [2]. The periodicity in the exchange of the excitation leads to the time dependence in the Einstein coefficients.

Three-dimensional multimode Jaynes-Cummings (J-C) Hamiltonian $\hat{H} = \frac{1}{2}\hbar\omega_0\sigma_3 +$

[1] The mode quality factor goes to infinity for an ideal cavity.

$\sum_{\vec{k}s} \hbar\omega_{\vec{k}} \hat{a}^{\dagger}_{\vec{k}s} \hat{a}_{\vec{k}s} - i \sum_{\vec{k}s} \hbar g_{\vec{k}s} [\sigma_+ \hat{a}_{\vec{k}s} - \sigma_- \hat{a}^{\dagger}_{\vec{k}s}]$[2] [12, 13, 14] which was proposed several decades back in this regard, is able to describe the quantum theory of radiation in a resonant cavity beyond (i) Dirac's determination of the Einstein B coefficient [15] within the 1st order time-dependent perturbation theory of quantum mechanics and (ii) Weisskopf-Wigner determination of the Einstein A coefficient [16] within the 1st order time-dependent perturbation theory of quantum field theory (quantum electrodynamics). Perturbation theories, however, can not explain the Rabi oscillation of a two-level system. The quantum Rabi oscillations, on the other hand, is well understood for the J-C model [12] even for a single mode [17, 18]. This model basically offers an understanding of the light-matter interactions in terms of the fundamental processes (spontaneous emission, stimulated emission and absorption) in the light of the cavity-QED. Though there have been an enormous amount of theoretical investigation in the field of the cavity-QED [17, 19], nobody has come up with a cavity-QED theory, except a few quantum master equation approaches with the J-C model (for only the resonant mode) and a phenomenological damping [20, 21] for the quantum Rabi oscillations of a two-level system in a lossy resonant cavity. Loss of the electromagnetic energy from the lossy resonant cavity, however, takes place for the frequency broadening around the resonant mode. This broadening naturally brings multimodes into account.

Now we are coming up with a cavity-QED theory within the J-C model for multimodes around the resonance for explaining the quantum Rabi oscillations in a lossy resonant cavity as observed by Brune *et al* [1]. Multimode J-C model [13] has become quite popular not only for an extension of the single-mode J-C model but also for the multi-photon transitions [22], the dynamics of entanglement [23], *etc*. We are, however, aiming to generalize the Einstein A and B coefficients in connection with the quantum Rabi oscillations under single-photon transitions.

The 3-D multimode J-C model result for the probability of stimulated or spontaneous emission a photon of (angular) frequency $\omega_{\vec{k}}$, wavevector $\vec{k}$ and polarization s over n such photons at time $t = 0$ from a two-level system having the Bohr frequency $\omega_0 = (E_2 - E_1)/\hbar$ found initially ($t = 0$) in the excited state in a cavity, takes

[2]Here we are following the notations [14]: $\sigma_+ = |\psi_2\rangle\langle\psi_1|$, $\sigma_- = |\psi_1\rangle\langle\psi_2|$, $\sigma_1 = [\sigma_+ + \sigma_-]$, $\sigma_2 = -i[\sigma_+ - \sigma_-]$, $\sigma_3 = |\psi_2\rangle\langle\psi_2| - |\psi_1\rangle\langle\psi_1|$, $\hat{a}_{\vec{k}s}$ ($\hat{a}^{\dagger}_{\vec{k}s}$) annihilates (creates) a photon of energy $\hbar\omega_{\vec{k}}$, polarization s and momentum $\hbar\vec{k}$ (having dispersion $\omega = ck$) in the Fock space, $|\psi_1\rangle$ ($|\psi_2\rangle$) is the energy eigenstate for the lower (higher) energy E_1 (E_2) of the two-level system in absence of the light-matter interactions, $g_{\vec{k}s}$ is the coupling constant (assumed real) for the light-matter interaction for the mode $\vec{k}s$, and $\omega_0 = (E_2 - E_1)/\hbar$ is the Borh (angular) frequency of the two-level system.

the form within the dipole approximation[3] as [12, 24, 14]

$$P_{2\to 1}^{n\to n+1}(g_{\vec{k}s},\omega_{\vec{k}},t) = 4g_{\vec{k}s}^2 \times (n+1) \times \frac{\sin^2\left(\frac{\sqrt{(\omega_{\vec{k}}-\omega_0)^2+4g_{\vec{k}s}^2(n+1)}\,t}{2}\right)}{(\omega_{\vec{k}}-\omega_0)^2+4g_{\vec{k}s}^2(n+1)} \tag{3.1}$$

where $g_{\vec{k}s} = \sqrt{\frac{\omega_{\vec{k}}}{2\hbar\epsilon_0 V}}\langle\psi_1|\hat{\vec{d}}\cdot\hat{e}_{\vec{k}s}|\psi_2\rangle$ [13], $\hat{\vec{d}}$ is the electric dipole moment operator for the two-level system, $\hat{e}_{\vec{k}s}$ is the unit-vector for the polarization of the cavity field[4], n is the number of photons of energy $\hbar\omega_{\vec{k}}$ and mode $\vec{k}s$ each present at around the two-level system before it undergoes a spontaneous or stimulated emission resulting $n+1$ photons of energy $\hbar\omega_{\vec{k}}$ and momentum $\hbar\vec{k}$ each after the emission and V is the volume of space occupied by both the two-level system and the photons. It should be mentioned in this regard that Eqn. (3.1) has a similarity with the single-mode J-C model result [12] because we are not considering any coupling between any two modes in the multimode J-C model.

Let us first consider the case of two-level system in a 3-D blackbody cavity [14]. There can be infinitely large number of choices of the modes ($\vec{k}s$) of a photon for a fixed $\omega_{\vec{k}} = \omega$. This causes appearance of the density of states ($\frac{\omega^2 V}{\pi^2 c^3}$[5] for two independent polarizations) once we go to description of the (angular) frequency. Thus averaging over all directions and polarizations for fixed $|\vec{k}| = \omega/c$ we get the net transition (spontaneous emission or stimulated emission) probability

$$\begin{aligned} P_{2\to 1}(t) &= \sum_{n=0}^{\infty}\int_0^{\infty} p_n(\omega) P_{2\to 1}^{n\to n+1}(g_\omega,\omega,t)\frac{\omega^2 V}{\pi^2 c^3}\mathrm{d}\omega \\ &= \sum_{n=0}^{\infty}\int_0^{\infty} p_n(\omega) P_{2\to 1}^{n\to n+1}(g_\omega,\omega,t)\frac{\tilde{u}(\omega)d_{21}^2}{3\epsilon_0 2\hbar^2 g_\omega^2}\mathrm{d}\omega \end{aligned} \tag{3.2}$$

where $g_{\vec{k}s}$ is replaced by the new coupling constant g_ω (such that $g_\omega^2 = \langle g_{\vec{k}s}^2\rangle_{\text{all directions and polarizations}} = \frac{\omega}{2\hbar\epsilon_0 V}\frac{d_{12}^2}{3}$) once the averaging over all the directions is taken, $d_{12} = \langle\psi_1|\hat{\vec{d}}|\psi_2\rangle$ is the transition dipole moment, the factor $1/3$ comes from averaging over the two independent polarization states of the blackbody radiation field, $\tilde{u}(\omega) = \frac{\hbar\omega^3}{\pi^2 c^3}$[6] represents the average energy density per thermal photon per unit (angular) frequency interval, and $p_n(\omega) = (1-\mathrm{e}^{-\frac{\hbar\omega}{k_B T}})\mathrm{e}^{-n\hbar\omega/k_B T}$ [6] is the occupation probability for n thermal photons which take part in spontaneous ($n=0$) or stimulated ($n \geq 1$) emission. While the transition probability $P_{2\to 1}^{n\to n+1}(\omega,t)$ is

[3]If the dimensions of the two-level system are small in comparison with the wavelength of the field and the wave functions of different two-level systems do not overlap, then only we can apply the dipole approximation ($\mathrm{e}^{i\vec{k}\cdot\vec{r}_0} \simeq 1$). Position ($\vec{r}_0$) of the two-level system in the cavity is not important within the dipole approximation

[4]Here $\hat{e}_{\vec{k}s}$ is perpendicular to $\vec{k}$.

[5]It follows from $2\frac{V4\pi k^2}{(2\pi)^3}\mathrm{d}k = 2\frac{V4\pi\omega^2}{(2\pi)^3 c^3}\mathrm{d}\omega$ (for 2 independent polarizations) where c is the speed of light in the space inside the blackbody cavity.

[6]$\tilde{u}(\omega_0)$ often appears in the Planck's distribution formula and is commonly known as the ratio of the Einstein A coefficient and the Einstein B coefficient [25].

sharply peaked at the resonance, the functions $\tilde{u}(\omega)/g_\omega^2$ and $p_n(\omega)$ are smooth in comparison to $P_{2\to1}^{n\to n+1}(\omega,t)$ at around the resonance.

The transition probability $P_{2\to1}^{n\to n+1}(g_\omega,\omega,t)$ in Eqn. (3.2) takes the form $P_{2\to1}^{n\to n+1}(g_\omega,\omega,t) \to 4g_\omega^2(n+1)\pi\frac{t}{2}\delta(\omega-\omega_0)$ in the limiting case of the weak coupling constant and long time exposition ($g_\omega \ll 1/t \ll \omega_0$). This result is compatible with Fermi's golden rule. The net transition probability as in Eqn. (3.2) thus takes the form in this limiting case as $P_{2\to1}(t) \to \sum_{n=0}^{\infty} tp_n(n+1)\frac{\pi\tilde{u}(\omega_0)}{3\epsilon_0}d_{21}^2/\hbar^2$. Here-from one gets the rate of the emission as $|\frac{d}{dt}P_{2\to1}(t)| = R_{2\to1}(t) \to A(0)+u(\omega_0)B_{21}(0)$ where $A(0) = \frac{d_{21}^2\omega_0^3}{3\pi c^3\epsilon_0\hbar}$ is Einstein's A coefficient, $B_{21}(0) = \frac{\pi d_{21}^2}{3\epsilon_0\hbar^2}$ is Einstein's B coefficient and $u(\omega_0) = \tilde{u}(\omega_0)\sum_{n=0}^{\infty} np_n = \frac{\hbar\omega_0^3}{\pi^2c^3}\frac{1}{e^{\hbar\omega_0/k_BT}-1}$ is the average energy density of the thermal photons per unit (angular) frequency interval[7]. This is a common way of deriving Einstein coefficients from the J-C model in the weak coupling limit and long time limit [14]. However, if we don't take these limits, both the time and the coupling constant would enter into the expression of the rates of the spontaneous emission and stimulated emission. Thus one can generalize the Einstein coefficients with time and coupling constant dependences. This chapter is dedicated to explore the time and the coupling constant dependences in the generalized Einstein coefficients and its consequences.

The rest of this chapter deals with the Eqn. (3.2). We calculate the net transition (spontaneous or stimulated emission) probability by integrating the right hand side of the Eqn. (3.2) over the (angular) frequency ω with proper normalization for both the range ($0 < \omega < \infty$) of the frequency and the distribution of the thermal photons at a temperature T. Then we renormalize the coupling constant (g_ω) of the J-C model taking the quantum Rabi oscillations into account and subsequently, we generalize the Einstein coefficients towards time-dependence. Using the renormalized coupling constant we calculate the net transition probability for a lossy resonant cavity with the relevant losses into account, and subsequently we discuss on the 'vacuum' Rabi oscillation. Then we do the similar study of the quantum Rabi oscillations for the injected coherent field. We compare our results with the quantum Rabi oscillations data obtained by Brune *at al* [1] for various situations. Finally, we discuss and conclude.

3.2 Jaynes-Cummings model result for the net transition probability

Since most of the contributions in the net transition probability in Eqn. (3.2) is coming from around the resonance ($\omega \to \omega_0$), we can safely replace $\tilde{u}(\omega)$ by $\tilde{u}(\omega_0)$, $p_n(\omega)$ by $p_n(\omega_0)$ and g_ω by g_{ω_0} while integrating over ω in the domain $0 < \omega < \infty$ or alternatively integrating over the generalized n-photon Rabi frequency $\Omega_n = \pm\sqrt{(\omega-\omega_0)^2+4g_{\omega_0}^2(n+1)}$ from -$\sqrt{\omega_0^2+4g_{\omega_0}^2(n+1)}$ to -$2g_{\omega_0}\sqrt{n+1}$ and $2g_{\omega_0}\sqrt{n+1}$

[7]The expression for $u(\omega_0)$ is often called as Planck's distribution formula.

to ∞ as ω varies from 0 to ω_0 and ω_0 to ∞ respectively with an avoided crossing at $\omega = \omega_0$. The first part of the integrations takes a closed form and becomes equal to the second part if we send the lower limit -$\sqrt{\omega_0^2 + 4g_{\omega_0}^2(n+1)}$ to $-\infty$ within the rotating wave approximation ($\omega_0^2 \gg 4g_{\omega_0}^2$ [8]). Thus we recast Eqn. (3.2) as

$$\begin{aligned} P_{2\to 1}(t) \quad &\simeq \quad A(0)\sum_{n=0}^{\infty} p_n(\omega_0)(n+1) \\ &\quad \times {}_1F_2\left(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-\frac{(\omega_n t)^2}{4}\right)t \end{aligned} \tag{3.3}$$

where ${}_1F_2$ is a generalized hypergeometric function expressed in the usual notation [9] and $\omega_n = 2g_{\omega_0}\sqrt{n+1}$ is the n-photon Rabi frequency. Involvement of the different values of the photon numbers (along with the different values of the n-photon Rabi frequency) in the Rabi oscillation is referred to as the quantum Rabi oscillations. The generalized hyper geometric function reaches 1 exhibiting the expected result $P_{2\to 1}(t) \to [A(0) + u(\omega_0)B_{21}(0)]t$ in the weak coupling limit ($g_{\omega_0}t \ll 1$) and long time limit ($\omega_0 t \gg 1$) [14].

3.2.1 Renormalization of the coupling constant for thermal photons in a blackbody cavity

The requirement that, $P_{2\to 1}(t)$ in Eqn. (3.3) reaches $1/2$ as t goes to infinity (which has also been experimentally observed [1]), renormalizes g_{ω_0} to be the effective (or renormalized) coupling constant as

$$g'_{\omega_0}(\bar{n}) = \frac{A(0)}{\bar{n}}\mathrm{Li}_{-\frac{1}{2}}\left(\frac{\bar{n}}{1+\bar{n}}\right) \tag{3.4}$$

where $\bar{n} = \sum_0^{\infty} n\ p_n(\omega_0) = \frac{1}{e^{\hbar\omega_0/k_BT}-1}$ is the average number of thermal photons in the blackbody cavity at the temperature T and $\mathrm{Li}_j(x) = x + x^2/2^j + x^3/3^j + ...$ is the poly-Logarithmic function of order j. The real function $\mathrm{Li}_j(x)$ though is defined for $x < 1\ \forall\ j$, its special form $\mathrm{Li}_{-\frac{1}{2}}(\frac{\bar{n}}{1+\bar{n}})$ is defined for all finite values of $\bar{n}$. The renormalized coupling constant $g'_{\omega_0}(\bar{n})$, which takes the light-matter coupling for both the thermal photons and no photons (i.e. vacuum) into account, reaches the Einstein A coefficient $A(0)$ at $T \to 0$. We show the same in the inset of figure 3.1. The net transition probability in Eqn. (3.3) is a quasi-periodic function of time and has the quasi (angular) frequency $\omega_\gamma = 2g_{\omega_0}\sqrt{\bar{n}+1}$ which can also be renormalized with the effective coupling constant $g'_{\omega_0}(\bar{n})$, as $\Omega_R(\bar{n}) = 2g'_{\omega_0}(\bar{n})\sqrt{\bar{n}+1}$. This renormalized frequency is the Rabi flopping frequency of the two-level system in

[8] The rotating wave approximation $\omega + \omega_0 \gg |\omega - \omega_0|$ ($\forall\ \omega$) implies $\omega_0^2 \gg 4g_{\omega_0}^2$ to hold near the resonance.

[9] ${}_1F_2(\{1/2\},\{1,3/2\},-(1/4)a^2t^2) = \frac{1}{t}\int_0^t J_0(a\tau)\mathrm{d}\tau$

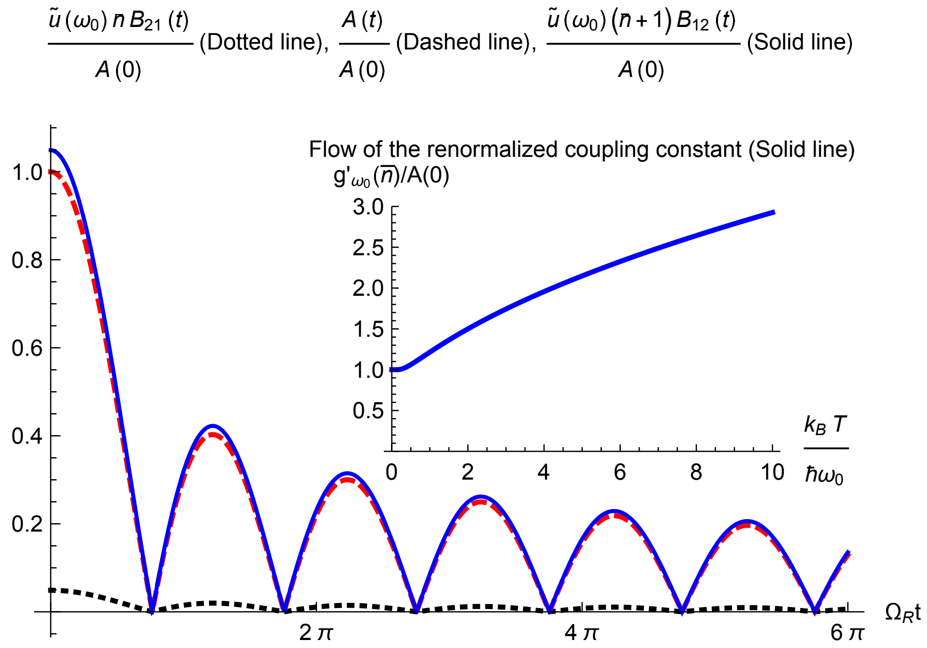

FIGURE 3.1: Multimode J-C model results for generalized Einstein coefficients for $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz and $T = 0.8$ K as taken in Ref.[1] for the circular Rydberg states (with the principal quantum number $n = 50$ and $n = 51$) of a ^{87}Rb atom. While the solid and the dotted lines follow Eqn. (3.5), the dashed line follows Eqn. (3.6). Envelope of the dashed lines follow $A(t)/A(0) \equiv \sqrt{2/\pi\Omega_R t}$ for large t. Solid line of the inset represents temperature dependence of the renormalized coupling constant and follows Eqn. (3.4).

the thermal radiation field[10]. One can, however, determine the value of the A coefficient using the relation $\Omega_R(\bar{n}) = 2g'_{\omega_0}(\bar{n})\sqrt{\bar{n}+1}$ from the experimental data of $\Omega_R = 2\pi \times 47 \times 10^3$ Hz $\simeq 0.295310 \times 10^6$ Hz [1].

3.2.2 Generalization of the Einstein coefficients towards time-dependence under the quantum Rabi oscillations

It is to be mentioned that the rate of transitions ($R_{2\to1}(t) = |\frac{\mathrm{d}}{\mathrm{d}t}P_{2\to1}(t)|$) of the two-level system at $t \to 0$ can be directly obtained from Eqn. (3.3) without referring to the Fermi's golden rule as $R_{2\to1}(0) = A(0)\sum_{n=0}^{\infty} p_n(\omega_0)(n+1) = B_{21}(0)\,u(\omega_0) + A(0)$. If the time-derivative ($\frac{\mathrm{d}}{\mathrm{d}t}P_{2\to1}(t)$) be negative, then it represents the rate of transitions in the reverse order. Thus we have defined the rate of transitions with the absolute value. While the rate $R_{2\to1}(0)$ reaches $u(\omega_0)$ times the Einstein B coefficient in absence of the vacuum fluctuations, it reaches the Einstein A coefficient in absence of the thermal photons. The coupling constant g_{ω_0} in Eqn. (3.3) further has to be replaced by the renormalized coupling constant $g'_{\omega_0}(\bar{n})$ to ensure $P_{2\to1}(\infty) = 1/2$. Eqn. (3.3) with g_{ω_0} replaced by $g'_{\omega_0}(\bar{n})$ thus unifies both the Dirac's theory of stimulated emission and the Weisskopf-Wigner theory of spontaneous emission in a single framework of the J-C model for multimodes. Such a unification was previously done

[10] Connection of this form of the Rabi frequency with the low $\bar{n}$ will be shown below Eqn. (3.6).

only for the resonant frequency ($\omega \to \omega_0$) by the use of the Fermi's golden rule on the time-derivative of the transition probability $P_{2\to1}^{n\to n+1}(\omega,t)$ [14]. Our consideration of the frequency broadening ($\triangle\omega \sim \Omega_R/2$) of the transition probability $P_{2\to1}^{n\to n+1}(\omega,t)$ around the resonant frequency ($\omega = \omega_0$) generalizes the previous unification by bringing time-dependence in the rate of the transitions $R_{2\to1}(t) = |\frac{\mathrm{d}}{\mathrm{d}t}P_{2\to1}(t)|$ as one can expect the same from the experimental observation of $P_{2\to1}(t)$ [1].

It is clear from the Eqn. (3.3) that, the stimulated emission part (n of $(n+1)$) and the spontaneous emission part (1 of $(n+1)$) though are primarily independent in the short time scale, are secondarily dependent on each other through the ${}_1F_2$ part of the net transition probability as time goes on. This is possible because spontaneously emitted photon can also take part in the stimulated emission. Eventually both the spontaneous emission part and the stimulated emission part of the transition rate $R_{2\to1}(t) = |\frac{\mathrm{d}}{\mathrm{d}t}P_{2\to1}(t)|$ become secondarily hybrid. Thus we get the emission rate $R_{2\to1}(t)$ for the renormalized coupling constant $g'_{\omega_0}(\bar{n})$ as $R_{2\to1}(t) = \tilde{u}(\omega_0)\bar{n}B_{21}(t) + A(t) = B_{21}(t)u(\omega_0) + A(t)$ such that

$$\begin{aligned} B_{21}(t) &= B_{21}(0)\sum_{n=0}^{\infty} n\, p_n(\omega_0)|J_0(2g'_{\omega_0}(\bar{n})\sqrt{n+1}t)|/\bar{n} \\ &\simeq B_{21}(0)|J_0(\Omega_R(\bar{n})t)| \end{aligned} \tag{3.5}$$

and

$$\begin{aligned} A(t) &= A(0)\sum_{n=0}^{\infty} p_n(\omega_0)|J_0(2g'_{\omega_0}(\bar{n})\sqrt{n+1}t)| \\ &\simeq A(0)|J_0(\Omega_R(\bar{n})t)| \end{aligned} \tag{3.6}$$

where $\Omega_R(\bar{n}) \simeq 2g'_{\omega_0}(\bar{n})\sqrt{\bar{n}+1}$ is the Rabi flopping frequency as defined before for low photon number fluctuation ($\triangle n = \sqrt{\bar{n}}\sqrt{\bar{n}+1} \lesssim 1$ [26]) for low $\bar{n}$. The transition rate $R_{2\to1}(t)$ becomes time-dependent along with its stimulated emission part $B_{21}(t)u(\omega_0)$ and the spontaneous emission part $A(t)$ only for the nonzero values of the Rabi frequency Ω_R. We can call $B_{21}(t)$ as the generalized Einstein B_{21} coefficient and $A(t)$ as the generalized Einstein A coefficient. We can also have $B_{12}(t)$ as the generalized Einstein B_{12} coefficient. The generalized Einstein coefficients, however, become the original time-independent Einstein coefficients for $\Omega_R \to 0$ i.e. for the case of no Rabi flopping [10, 15, 16]. Eqns. (3.5) and (3.6) represent the multimode J-C model results for the Einstein coefficients. We have got these results published in Ref. [27]. It should also be mentioned that, the generalized Einstein B coefficient as shown in Eqn. (3.5) takes the form similar to that obtained in the semiclassical Rabi model [28]. The semiclassical Rabi model, however, can not generalize the Einstein A coefficient. The generalized Einstein B coefficients obtained within the two models (Rabi model and multimode J-C model) are not exactly same. They significantly differ in the limiting case of $\bar{n} \to 0$.

On the other hand, if $P_{1\to2}^{n+1\to n}(g_\omega,\omega,t)$ be the transition probability counter to

$P_{2\to 1}^{n\to n+1}(g_\omega, \omega, t)$ for the (stimulated) absorption of 1 photon from $n+1$ photons of frequency ω each and $P_{1\to 2}(t)$ be the corresponding net transition probability counter to $P_{2\to 1}(t)$, then we must have $P_{2\to 1}^{n\to n+1}(\omega, t) + P_{1\to 2}^{n+1\to n}(\omega, t) = 1$ and $P_{2\to 1}(t) + P_{1\to 2}(t) = 1$. Here-from we can show that, the rate of the transition probability for the absorption of the two-level system in presence of the average thermal photons $\bar{n}$ and 1 emitted photon at any arbitrary time is $R_{1\to 2}(t) = |\frac{d}{dt}P_{1\to 2}(t)| = \tilde{u}(\omega_0)(\bar{n}+1)B_{12}(t)$. This relation leads to the equality $B_{12}(t) = B_{21}(t)$ as because $\tilde{u}(\omega_0)B_{21}(t) = A(t)$ holds for any arbitrary time according to Eqns. (3.5) and (3.6).

We plot all the generalized Einstein coefficients with proper weightage for stimulated emission rate (dotted line), spontaneous emission rate (dashed line) and absorption rate (solid line) all in units of $A(0)$ in figure 3.1 for a low temperature $T = 0.8$ K so that the background of two-level system is filled with a very small number of average thermal photons ($\bar{n} = 0.0489$ [1]). While on average $\bar{n}$ thermal photons are present in the background of the two-level system for its stimulated emission, on average $\bar{n}$ thermal photons and one emitted photon are present in the background of two-level system for its absorption. This makes significant difference between the two processes corresponding to the observation of the 'vacuum'[11] Rabi oscillation at the low temperature ($T = 0.8$ K) by Brune *et al* [1]. It is clear from figure 3.1 that, the 'vacuum' Rabi oscillation takes place due to subsequent interplay of the spontaneous emission and absorption. Role of the stimulated emission is suppressed in the 'vacuum' Rabi oscillation. On the other hand, role of the spontaneous emission is suppressed at a higher temperature. In that case, the dotted line and the solid line in figure 3.1 would come close to each other.

3.2.3 Renormalization of the coupling constant for photons in a lossy resonant cavity

Let us now consider the spontaneous or stimulated emission from the two-level system in a lossy resonant cavity, say a Fabry-Perot cavity, with z-axis be the cavity axis [1, 3]. Above result for the blackbody cavity is expected to be unaltered if the separation of the two reflecting walls of the resonant cavity is several times larger than the wavelength of the resonant mode. The quantum Rabi oscillations need the emitted photon to have lifetime ($\sim 200\ \mu$s) longer than the light-matter interaction time, so that it can be repeatedly reflected back and forth with the mirrors of the cavity before it leaks out through the holes (of size $\sim$mm^2 each) on the axis of the cavity or becomes absorbed (or scattered) in the walls of the cavity resulting in the "Ohmic" loss [29]. The parameter which ensures the longer lifetime is the higher mode quality factor ($Q = 7 \times 10^7$ [1]) of the cavity. The leakage and the "Ohmic" loss can be attributed to this value ($Q = 7 \times 10^7$ [1]) of the mode quality factor. However, there is additional loss as because the curved surface of the cylindrical geometry of the cavity is open. Thus the probability that the emitted photon escapes from the

[11] We are calling it to be 'vacuum' because the background of the two-level system in the cavity is truly not empty at $T = 0.8$ K.

cavity through the curved surface of the cylindrical shaped open cavity (of circular mirrors of radius r each and separation h) is $p_0 = \frac{2\pi rh}{2\pi rh+2\pi r^2} = \frac{1}{1+\frac{r}{h}}$ which results the net quality factor as $Q' = \frac{1}{\frac{1}{Q}+\frac{p_0 A(0)}{\omega_0}}$ [12]. Derivation of the net quality factor (Q') is shown in Sec. 3.A (Appendix A). Since the individual loss leads to a Lorentzian distribution, convolution of the relevant losses (leakage, "Ohmic" loss, and the loss due the spontaneous emission through the open surface of the cavity) also leads to the Lorentzian distribution $u'(\omega) = u(\omega_0)\frac{2}{\pi}\frac{(\omega_0/Q')^2}{4(\omega-\omega_0)^2+(\omega_0/Q')^2}$ with the net width ω_0/Q' over the Planck's distribution $u(\omega) \simeq u(\omega_0)$. However, above form of the net transition probability (Eqn. (3.3)) would be unaltered if the broadening due to the relevant losses is much higher than that due to the natural decay (i.e. $\omega_0/Q' \gg A(0)$). Thus, we recast Eqn. (3.3) by further renormalizing the coupling constant as shown in Eqn. (3.4) as

$$\begin{aligned} P_{2\to 1}(t) \;&\simeq\; A(0)\sum_{n=0}^{\infty} p_n(\omega_0)(n+1) \\ &\times {}_1F_2\left(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-[g'_{\omega_0}(\bar{n})\sqrt{n+1}t]^2\right)t. \end{aligned} \tag{3.7}$$

Incidentally we have $A(0) \simeq 15.6765$ Hz in free space or in a (very large) black-body cavity for $d_{21} = 1250a_0e$ [3] of the two-level system (^{87}Rb) of our interest and $\omega_0/Q' \simeq 250210$ Hz for the cavity of our interest [1]. Hence the condition $\omega_0/Q' \gg A(0)$ is well met if the value of $A(0)$ remains same (or decreases) in the cavity space. Otherwise, each (angular) frequency in the net transition probability (Eqn. (3.7)) would have to be weighted by the Lorentzian distribution $u'(\omega)$.

3.2.4 Quantum Rabi oscillations for the two-level system in a lossy resonant cavity

However, value of the Einstein A coefficient ($A(0)$) increases enormously in the Fabry-Perot cavity due to the Purcell effect [9]. Broadening due to the relevant losses may not be so large in comparison to $A(0)$ in this situation. Each frequency in the net transition probability (Eqn. (3.7)) should be weighted by the Lorentzian distribution $u'(\omega) = u(\omega_0)\frac{2}{\pi}\frac{(\omega_0/Q')^2}{4(\omega-\omega_0)^2+(\omega_0/Q')^2}$ in this case. Thus Eqn. (3.7) would be further recast in a similar way of reaching Eqn. (3.3) from Eqn. (3.1) as

$$\begin{aligned} P_{2\to 1}(t) \;&=\; A(0)\sum_{n=0}^{\infty} p_n(\omega_0)[n+1]\frac{4}{\pi}\times \\ &\int_{\omega_n}^{\infty}\frac{(\omega_0/Q')^2}{4(\Omega_n^2-\omega_n^2)+(\frac{\omega_0}{Q'})^2}\frac{\sin^2(\Omega_n t/2)}{\Omega_n\sqrt{\Omega_n^2-\omega_n^2}}\mathrm{d}\Omega_n \end{aligned} \tag{3.8}$$

[12] Here $A(0)$ is the frequency broadening ($\triangle\omega$ around the resonance frequency ω_0) for the natural decay in the free space [16]. The natural decay in the free space results in the Q-factor $\frac{\omega_0}{\triangle\omega} = \frac{\omega_0}{A(0)}$.

where $\omega_n = 2g'_{\omega_0}(\bar{n}, Q')\sqrt{n+1}$ is the new renormalized n-photon Rabi frequency and $g'_{\omega_0}(\bar{n}, Q')$ is the new renormalized coupling constant which is to be determined by setting the limit $P_{2\to1}(\infty) = 1/2$.

'Vacuum' Rabi oscillation

Eqn. (3.8) would be approximated by further neglecting the photon-number fluctuation ($\triangle n = \sqrt{\bar{n}}\sqrt{1+\bar{n}}$ [26]) at the higher order of the Taylor expansion of $P_{2\to1}(t)$ about $n = \bar{n}$ for low $\bar{n}$ ($\bar{n} \lesssim 1$) at a low temperature[13], as

$$P_{2\to1}(t) \simeq A(0)[\bar{n}+1]\frac{4}{\pi} \times \int_{\omega_{\bar{n}}}^{\infty} \frac{(\omega_0/Q')^2}{4(\Omega_{\bar{n}}^2 - \omega_{\bar{n}}^2) + (\frac{\omega_0}{Q'})^2} \frac{\sin^2(\Omega_{\bar{n}}t/2)}{\Omega_{\bar{n}}\sqrt{\Omega_{\bar{n}}^2 - \omega_{\bar{n}}^2}} d\Omega_{\bar{n}}. \quad (3.9)$$

The number 1 next to $\bar{n}$ in Eqn. (3.9) arises purely from the quantum fluctuations. Effect of the quantum fluctuations are suppressed in the classical regime ($k_B T/\hbar\omega_0 \gg 1$). Thus Eqn. (3.9) corresponds to the classical Rabi oscillation for $\bar{n} \gg 1$. However, the new renormalized coupling constant used in Eqn. (3.9) can be determined by setting the limit $P_{2\to1}(\infty) = 1/2$ as $g'_{\omega_0}(\bar{n}, Q') = \frac{A(0)\sqrt{\bar{n}+1}}{1+4g'_{\omega_0}(\bar{n},Q')\sqrt{\bar{n}+1}Q'/\omega_0}$ which further determines the Rabi frequency for a lossy resonant cavity at a low temperature as $\Omega_R(\bar{n}, Q') = 2g'_{\omega_0}(\bar{n}, Q')\sqrt{\bar{n}+1} = \frac{-1+\sqrt{1+16A(0)[\bar{n}+1]Q'/\omega_0}}{4Q'/\omega_0}$. This relation further determines the Einstein A coefficient to be as $A(0) = \frac{\Omega_R}{2(\bar{n}+1)} + \frac{\Omega_R^2 Q'}{\omega_0(\bar{n}+1)}$. While the 1st term of $A(0)$ represents the Einstein A coefficient in the free space, the 2nd term represents enhancement of the A coefficient due to the Purcell effect in the resonant cavity. Now we get enhanced value of the A coefficient as $A(0) \simeq 0.473053 \times 10^6$Hz for the ^{87}Rb atom in the resonant cavity of our interest [1]. The net quality factor corresponding to this $A(0)$ now takes the value $Q' = 1.28318 \times 10^6$.

We plot the right hand side of the Eqn. (3.9) in the figure 3.2-a for the ^{87}Rb atom in the resonant cavity [1]. The solid line in the figure 3.2-a represents the 'vacuum' Rabi oscillation in the resonant cavity for the parameters as mentioned in the figure-caption. The cavity is truly not empty rather has on the average $\bar{n} = 0.0489$ thermal photons in it [1]. For this reason we are not calling the Rabi oscillation to be as the vacuum Rabi oscillation, rather we are calling it to be as the 'vacuum' Rabi oscillation. Note that only $n = 0$ photons are involved in the case of the (true) vacuum Rabi oscillation. The dotted line represents a fit with the same equation but for a lower value ($Q = 7 \times 10^5$) of the Q-factor. Damping of the Rabi oscillation even in the high Q cavity, as shown in figure 3.2-a, is caused due to the finite width ($\sim \Omega_R/2$) of the frequency distribution around the resonance. We have got this result published in Ref. [27]. Better matching for the lower Q-factor can be attributed to the substantial losses from the resonant cavity corresponding to the frequency broadening due to

[13] Here $\bar{n}$ is a small quantity at a low temperature. Thus $\sqrt{n+1}$ at the argument of the generalized hypergeometric function in Eqn. (3.7) is approximated as $\sqrt{\bar{n}+1}\ \forall\ n$.

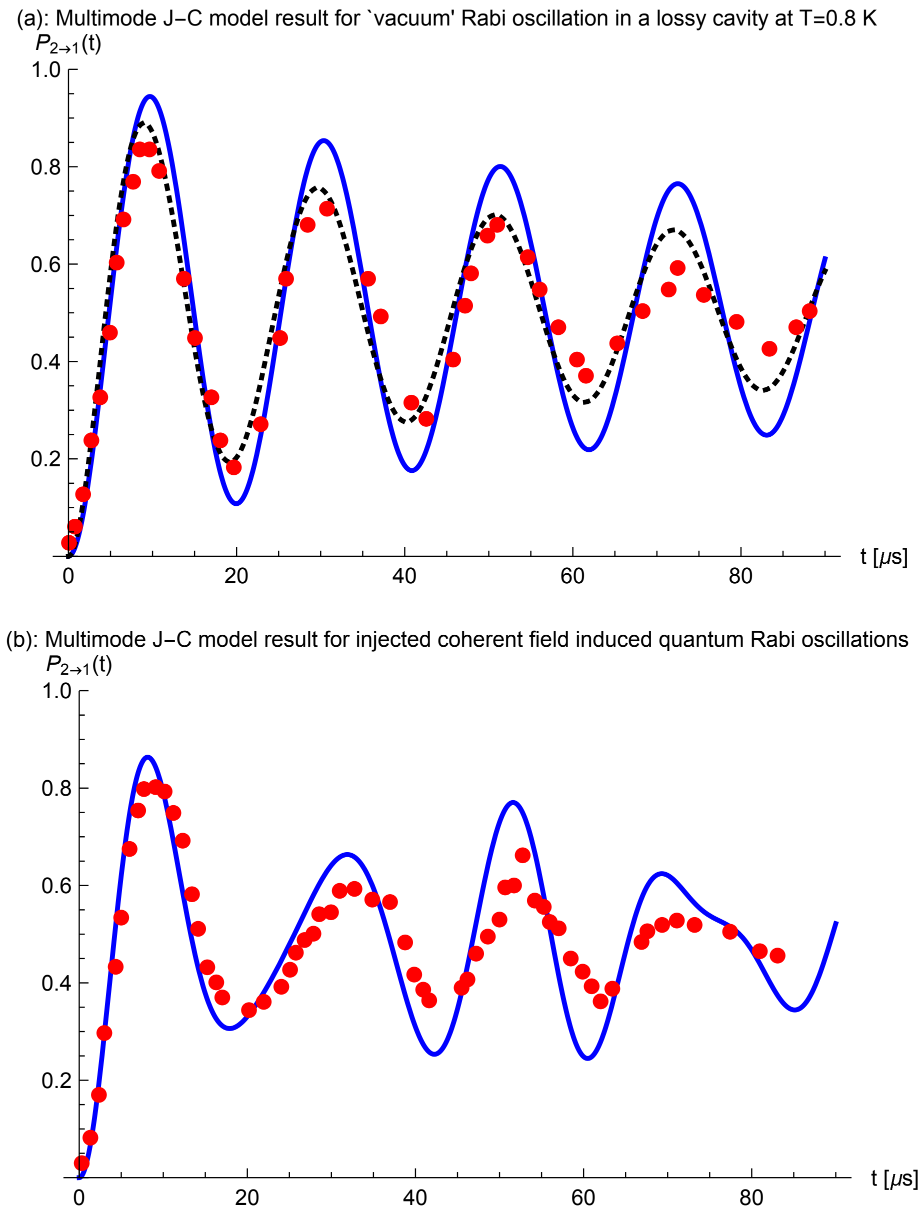

FIGURE 3.2: (a): Solid line represents the 'vacuum' Rabi oscillation, and follows Eqn. (3.9) for the Bohr frequency $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz, Rabi frequency $\Omega_R = 2\pi \times 47 \times 10^3$ Hz and average number of thermal photons $\bar{n} = 0.0489$. Circles represent corresponding experimental data [1] adapted for the circular Rydberg states (with the principal quantum number $n = 50$ and $n = 51$) of ^{87}Rb atoms in an open resonant cavity of the Q-factor $Q = 7 \times 10^7$ and size $\pi(50/2)^2 \times 27$ mm^3 at the temperature $T = 0.8$ K. The dotted line represents the same for $Q = 7 \times 10^5$.
(b): Solid line represents injected coherent field induced quantum Rabi oscillations, and follows Eqn. (3.8) for the same parameters as mentioned above except for Rabi frequency $\Omega_R = 2\pi \times 55.6949 \times 10^3$ Hz and the average number of injected photons $\bar{n} = 0.4$ in the lossy cavity. Circles represent corresponding experimental data [1] adapted for the same two-level system.

inhomogeneous light-matter coupling along the cavity axis [1], Doppler broadening due to the speed distribution of ^{87}Rb atoms in the cavity, thermal broadening, *etc*.

Quantum Rabi oscillations for injected coherent field

Quantum Rabi oscillation was not only observed in the form of 'vacuum' Rabi oscillation but also in the form of coherent field induced Rabi oscillation [1]. Let us also do similar study of injected coherent field induced quantum Rabi oscillations. The main difference in this respect comes from the probability distribution of photons. While the probability distribution follows exponential law for the thermal photons, it follows Poisson distribution $p_n(\omega_0) = \frac{\bar{n}^n}{n!}\mathrm{e}^{-\bar{n}}$ for $\bar{n}$ average number photons of frequency ω_0 in the coherent field. Averaging of the transition probability $P_{2\to1}(t)$ over the polarization is no longer needed. Thus the factor $\frac{1}{3}$ is no longer needed in Eqn. (3.2). All values of the frequencies as mentioned in Eqn. (3.2) are also not welcome in the injected coherent field as because the coherent field has finite maximum detuning, say $(\omega - \omega_0)_{max} = 10^6$ Hz in Brune *et al*s' experiment [1]. However, significant contribution in the integrals in Eqns. (3.2) and (3.8) are coming from the domain $\omega_0 - \Omega_R$ to $\omega_0 + \Omega_R$ of the frequency ω. Incidentally, the maximum detuning is about 3 times of Ω_R [1]. Thus Eqn. (3.8) with $p_n(\omega_0) = \frac{\bar{n}^n}{n!}\mathrm{e}^{-\bar{n}}$ would still be okay for the net transition probability of the spontaneous emission or the stimulated emission induced by the injected coherent field. Injected coherent field increases the light-matter coupling constant so as the Rabi frequency. The renormalized coupling constant ($g'_{\omega_0}(\bar{n}, Q')$) can be determined from the limiting value $P_{2\to1}(\infty) = 1/2$ for the previous values of $A(0)$ ($= 0.473053 \times 10^6$ Hz) and Q' ($= 1.28318 \times 10^6$ Hz)). We determine the new renormalized coupling constant to the second order in experimental value $\bar{n} = 0.4$ ($\lesssim 1$) [1] as $g_{\omega_0}(0.40, Q') \simeq 0.147877 \times 10^6$ Hz. Here-from we get the Rabi frequency as $\Omega_R = 2g_{\omega_0}(0.40, Q')\sqrt{0.4+1} \simeq 55.6949 \times 2\pi \times 10^3$ Hz.

The solid line in the figure 3.2-b represents the injected coherent field induced quantum Rabi oscillations in the resonant cavity for the parameters as mentioned in the figure-caption. Amplitude of the quantum Rabi oscillations in the figure 3.2-b is observed to be less than that in the figure 3.2-a because of the larger photon number fluctuation in the case of the figure 3.2-b. Photon number fluctuation kills the quantum Rabi oscillations for large values of $\bar{n}$ ($\gg 1$). Damping of the Rabi oscillations even for the coherent field in the cavity, as shown in figure 3.2-b, is caused due to the finite width of the frequency broadening at around the resonance. However, we see good agreement of our theoretical result with the experimental data in the figure 3.2-b. We have got this result published in Ref. [27]. The matching would have been better had we considered additional losses from the cavity, inhomogeneous light-matter coupling, Doppler broadening, finite detuning of the injected coherent field, thermal broadening, higher order effect of $\bar{n}$ in the light-matter coupling constant, *etc*.

3.3 Conclusion

We have obtained multimode Jaynes-Cummings model results for all the generalized Einstein coefficients for a two-level system in the thermal radiation field and have shown that all the generalized Einstein coefficients depend on time and Rabi frequency in a similar manner for low photon number fluctuation. These results are accurate for fairly large Bohr frequency ($\omega_0 \gg \Omega_R$) and are significantly different from the results obtained within the first order time-dependent perturbation theory which considers $\Omega_R \rightarrow 0$. Renormalization of the light-matter coupling done for both the vacuum field and thermal photons together leads to such a difference from the previous theories. We have obtained analytical results within the multimode Jaynes-Cummings model for the quantum Rabi oscillations for both the thermal photons and the photons of an injected coherent field. Our results on the quantum Rabi oscillations match well with the experimental data [1]. Our generalization of the Einstein coefficients is an invitation to the experimentalists for direct measurement of the Einstein coefficients for the two-level system(s) in a blackbody cavity.

While the Einstein coefficients deal with the fundamental processes (e.g. spontaneous emission, stimulated emission and absorption), they don't directly deal with the Rabi oscillation. On the other hand, the quantum Rabi oscillations deal with the fundamental processes. Hence it is possible to derive Einstein coefficients from the analyses of the quantum Rabi oscillations and we have done that for a nonzero Rabi frequency. Thus we have generalized the Einstein coefficients towards time and Rabi frequency dependences.

Drexhage observed alterations in the rate of spontaneous emission, regarding the influence the atomic radiation, while working on the fluorescence of organic dyes deposited on dielectric films over a metallic mirror [11, 2]. Once a two-level system in a resonant cavity emits a photon it is periodically reabsorbed in the cavity exhibiting the quantum Rabi oscillation. Thus time-dependence of the Einstein coefficients is not a surprise at least for a two-level system in a resonant cavity. However, probability of the reabsorption is negligibly small for the same system in the free space[14]. Thus the Einstein coefficients are not found to be time-dependent in the free space.

We are not able to compare the result on the generalized Einstein B coefficient with the existing experimental data as because they have not been obtained by any direct measurement; rather, experimentalists apply time-dependent perturbation theory ($A/B = \frac{\hbar\omega_0^3}{\pi^2 c^3}$) for the indirect measurement of the Einstein B coefficient from the experimental value of the Einstein A coefficient [30]. Time-dependence of the generalized Einstein A coefficient could have been caught by the experimentalists had they measured it for longer time scale i.e. the time scale of the quantum Rabi oscillations. Measurement of the absorption coefficient [25] for a two-level system doesn't also serve the purpose of capturing the time-dependence of the generalized

[14]Here, free space refers to a large blockbody cavity.

Einstein B coefficient, as because, averaging of the absorption of photons of all possible frequencies and of all possible directions of incidence is not considered in the measurement [31].

Throughout the chapter by "frequency" we have meant "angular frequency".

We have dealt with a single two-level system (qubit) in the 3-D multimode J-C model. Transition probability calculated for this system would not have changed if we had taken non-interacting and distinguishable identical two-level systems.

While the quantum Rabi oscillation is studied for strong light-matter interactions ($g_{\omega_0} \gg \sup\{\gamma,\ \kappa\}$[15] [32]), the Einstein rate equations are often applied for weak light-matter interactions ($g_{\omega_0} \ll \sup\{\gamma,\ \kappa\}$ [32]). Incidentally, the multimode J-C model gives results in both the weak coupling regime and the strong coupling regime as far as the rotating wave approximation ($2g_{\omega_0} \ll \omega_0$) is applicable. Thus we have been interested in bridging the quantum Rabi oscillation and the phenomenological rate equations by the multimode J-C model. The experimental data [1], which we have compared with our results in the figure 3.2, satisfy both the strong coupling condition and the rotating wave approximation. The J-C model, however, is not applicable in the ultrastrong coupling ($g_{\omega_0} \sim \omega_0$) and deep strong coupling ($g_{\omega_0} \gg \omega_0$) regimes [33, 34]. The quantum Rabi model which generalizes the J-C model is applicable in these regimes [35, 36].

The generalized Einstein A and B coefficients obtained in Eqns. (3.5) and (3.6) may take place in the time-dependent rate coefficients $R_{1\to 2}(t)$, $R_{2\to 1}(t)$, and $A(t)$ in the Einstein rate equations if the time-independent rate coefficients are replaced by the time-dependent rate coefficients. Population dynamics would have to be studied in terms of the time-dependent rate coefficients in such a case. Such a population dynamics with the time-dependent rate coefficients would be interesting for analysing the entropy production of a two-level system in the thermal radiation field. In Chapter-5 we will study the population dynamics and entropy production for the time-dependent forms of the generalized Einstein coefficients obtained in Eqns. (3.5) and (3.6).

3.A Appendix A: Derivation of the net quality factor

The quality factor Q of a resonant optical cavity is defined as follows [37]

$$Q = w_0 \frac{W}{P_{loss}} \tag{3.A.10}$$

where w_0 is the (angular) resonance frequency, W is the electromagnetic energy stored in the resonant mode of the cavity, and P_{loss} is the electromagnetic energy lost per optical cycle to the walls of the cavity.

The "Ohmic" loss, apart from the loss due to the surface a.c. current flow in the (conducting) cavity walls, also includes the losses due to the host-crystal absorption,

[15] Here γ is the non-resonant decay rate and $\kappa = \omega_0/Q$ is the photon decay rate of the cavity [32].

impurities, scattering loss, excited-state absorption, and other effects [29]. We are also considering the loss of the stored electromagnetic energy due to the leakage from the holes on the cavity axis in addition to the "Ohmic" loss. If Q_1 be the quality factor of the cavity corresponding to the "Ohmic" loss and Q_2 be the quality factor of the cavity corresponding to the leakage, then the inverse of quality factor (Q) of the cavity corresponding to both the losses is given by [37]

$$\frac{1}{Q} = \frac{1}{Q_1} + \frac{1}{Q_2}. \tag{3.A.11}$$

The "Ohmic" loss, however, would be very low for a superconducting cavity and the quality factor for such a cavity would be very high ($Q = 7 \times 10^7$ [1]). The differential equation for the variation of the stored electromagnetic energy over the time t follows [37, 38]

$$\frac{\mathrm{d}W}{\mathrm{d}t} = -\frac{\omega_0}{Q} W \tag{3.A.12}$$

for both the "Ohmic" loss and the leakage together. A solution to this equation is

$$W(t) = W(0)\mathrm{e}^{-\frac{\omega_0 t}{Q}}. \tag{3.A.13}$$

Here-from we can write the temporal part of the electric field associated with the resonant mode as [38]

$$E(t) = E_0 \mathrm{e}^{-\frac{\omega_0 t}{2Q}} \mathrm{e}^{-i\omega_0 t}. \tag{3.A.14}$$

It is clear from the above equation that the oscillation of the electric field dies as $E(t) \propto E_0 \mathrm{e}^{-\frac{\omega_0 t}{2Q}}$ in the resonant optical cavity. Fourier transform of the above temporal part of the electric field becomes

$$\tilde{E}(\omega) = E_0 \int_0^\infty \mathrm{e}^{-\frac{\omega_0 t}{2Q}} \mathrm{e}^{i[\omega-\omega_0]t} \mathrm{d}t = \frac{E_0 2iQ}{2Q[\omega-\omega_0] + i\omega_0} \tag{3.A.15}$$

for the time $t > 0$ and the (angular) frequency $\omega > 0$. Here-from we get the spectral distribution of the (angular) frequencies as [38]

$$|\tilde{E}(\omega)|^2 = E_0^2 \frac{1}{[\omega-\omega_0]^2 + \frac{\omega_0^2}{4Q^2}}. \tag{3.A.16}$$

The shape of this spectral distribution is Lorentzian and the full width at half maximum (FWHM) of the distribution is given by $\triangle\omega = \frac{\omega_0}{Q}$ [38].

Another Lorentzian broadening of the (angular) frequencies, similar to the one in Eqn. (3.A.16), is also obtained for the spontaneous emission from a two-level system (atom/molecule) in the free space within the Weisskopf-Wigner approximation as

[16, 39]

$$|\tilde{E}(\omega)|^2 = E_0^2 \frac{1}{[\omega - \omega_0]^2 + \frac{A(0)^2}{4}} \tag{3.A.17}$$

where $A(0)$ is the Einstein A coefficient. The width (FWHM) of this Lorentzian broadening is given by $\triangle\omega = A(0)$ [38]. However, if the two-level system be kept in the cavity, then only a fraction of the total spontaneous emission can escape from the cavity resulting in an additional loss through the open surface of the cavity. Let the cavity be of cylindrical shape and its curved surface is open. The probability that an emitted photon escapes from the cavity through the curved surface is $p_0 = \frac{2\pi rh}{2\pi rh + 2\pi r^2} = \frac{1}{1+\frac{r}{h}}$ where r is the radius of each of the mirrors of the cavity and h is the separation of the two mirrors. Thus Eqn. (3.A.17) would be modified for the spontaneous emission through the curved surface of the cavity as

$$|\tilde{E}(\omega)|^2 = E_0^2 \frac{1}{[\omega - \omega_0]^2 + \frac{p_0^2 A(0)^2}{4}}. \tag{3.A.18}$$

Convolution of the two Lorentzian distributions of Eqns. (3.A.16) and (3.A.18) is also another Lorentzian distribution with the net width (FWHM) $\triangle\omega' = \frac{\omega_0}{Q} + p_0 A(0)$ which is the addition of the widths (FWHM) of the two distributions [40]. If we compare Eqn. (3.A.18) with Eqn. (3.A.16) then we can assign a quality factor for the loss associated with the spontaneous emission through the curved surface of the cavity as

$$Q_3 = \frac{\omega_0}{p_0 A(0)}. \tag{3.A.19}$$

The inverse of the net quality factor corresponding to the broadenings of Eqns. (3.A.16) and (3.A.18), on the other hand, would be an addition of the inverse of the individual quality factors as mentioned in Eqn. (3.A.11) [37]. Thus the inverse of the net quality factor of the lossy resonant optical cavity of our interest would be $\frac{1}{Q'} = \frac{1}{Q} + \frac{1}{Q_3} = \frac{1}{Q} + \frac{p_0 A(0)}{\omega_0}$. Here-from we get the desired net quality factor as [27]

$$Q' = \frac{1}{\frac{1}{Q} + \frac{p_0 A(0)}{\omega_0}}. \tag{3.A.20}$$

This form of the net quality factor has been used in Eqn. (3.8).

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Chapter 4

Multimode Jaynes-Cummings model results for the collapse and revival of the quantum Rabi oscillations in a lossy cavity

4.1 Introduction

Collapse and revival of the quantum Rabi oscillations of a two-level system (atom / molecule) is an interesting area of research in the field of cavity quantum electrodynamics [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. Eberly *et al* first predicted the phenomena of the collapse and the revival within the single-mode Jaynes-Cummings (J-C) model [12] for the quantum Rabi oscillations of a two-level system interacting with coherent photons in a cavity [1]. The collapse and the revival were subsequently observed by investigating the dynamics of the interaction of a single Rydberg atom with the resonant mode of an electromagnetic field in a superconducting cavity [5]. While the existing theories [1, 2, 3, 4, 10, 11] for the collapse and the revival usually require a large average number of photons ($\bar{n} \gg 1$) in the coherent field, a seminal experiment [7] on the same was carried out by Brune *et al* for a low average number of photons ($\bar{n} \gtrsim 1$) in the coherent field. In fact, as far as we know, all the experiments on the collapse and the revival were carried out for low average number of photons [5, 13, 7] except the one [14] carried out for $\bar{n} = 13.4$. Hence we theoretically investigate the collapse and the revival for a low average number of photons in a coherent field. Theory for the collapse and the revival is also available for low average number of injected coherent photons as well as for all values of the average number of the injected coherent photons [15, 6, 16, 7, 14, 17, 10]. This theory takes only the resonant mode into account for the light-matter interactions. We are, however, interested in considering multimodes into account.

J-C model takes only the resonant cavity mode into account for the explanation of the collapse and the revival of the quantum Rabi oscillations of a two-level system in a loss-less cavity [12, 1]. However, the cavities are not loss-less in reality [7]. This brings a frequency broadening as well as the appearance of multimodes around the

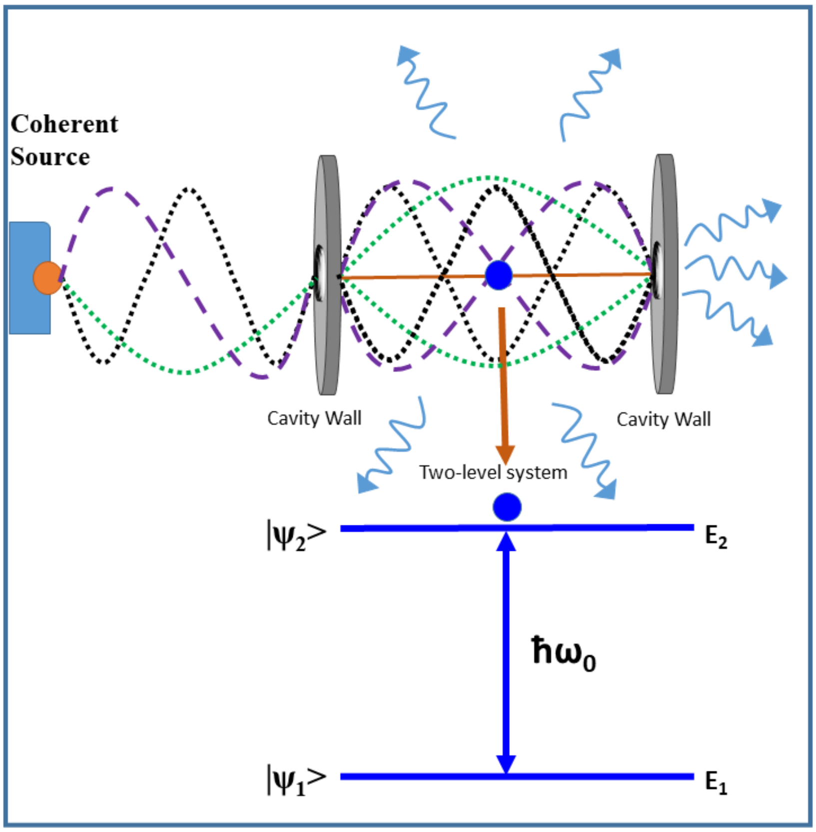


FIGURE 4.1: Schematic diagram for a two-level system interacting with injected coherent photons in a lossy resonant cavity.

resonant mode into account. Brune *et al*'s experiment on the collapse and the revival were carried out in a lossy resonant cavity of the mode quality factor $Q = 7 \times 10^7$ [7]. The schematic diagram for the two-level system interacting with the injected coherent photons in the lossy resonant cavity is shown in figure 4.1. It is clear from figure 4.1 how the injected coherent photons are introduced into the cavity and how the two-level system is interacting with the multi-modes of the injected coherent photons in the cavity. Losses from the cavity are shown by the wavy arrows in the same figure. The frequency broadening in Brune *et al*'s experiment can be attributed to the multimode J-C model, $\hat{H} = \frac{1}{2}\hbar\omega_0\sigma_3 + \sum_{\vec{k}s}\hbar\omega_{\vec{k}}\hat{a}^\dagger_{\vec{k}s}\hat{a}_{\vec{k}s} - i\sum_{\vec{k}s}\hbar g_{\vec{k}s}[\sigma_+\hat{a}_{\vec{k}s} - \sigma_-\hat{a}^\dagger_{\vec{k}s}]$ [1] [18, 19], rather than the single-mode Jaynes-Cummings model [20]. Thus the theoretical explanation of the collapse and revival of the quantum Rabi oscillations in a lossy resonant cavity needs a novel approach with the multimode Jaynes-Cummings model. The novel approach must take relevant losses from the cavity into account for the explanation of Brune *et al*'s experimental data [7]. Here we provide such a

[1]Here $\hat{H}$ is the Hamiltonian operator for a two-level system interacting with photons in a lossy resonant cavity. We are following the notations [19]: $\sigma_+ = |\psi_2\rangle\langle\psi_1|$, $\sigma_- = |\psi_1\rangle\langle\psi_2|$, $\sigma_1 = [\sigma_+ + \sigma_-]$, $\sigma_2 = -i[\sigma_+ - \sigma_-]$, $\sigma_3 = |\psi_2\rangle\langle\psi_2| - |\psi_1\rangle\langle\psi_1|$, $\hat{a}_{\vec{k}s}$ ($\hat{a}^\dagger_{\vec{k}s}$) annihilates (creates) a photon of energy $\hbar\omega_{\vec{k}}$, polarization s and momentum $\hbar\vec{k}$ (having dispersion $\omega = ck$) in the Fock space, $|\psi_1\rangle$ ($|\psi_2\rangle$) is the energy eigenstate for the lower (higher) energy E_1 (E_2) of the two-level system in absence of the light-matter interactions, $g_{\vec{k}s}$ is the coupling constant (assumed real) for the light-matter interaction for the mode $\vec{k}s$, and $\omega_0 = (E_2 - E_1)/\hbar$ is the Borh (angular) frequency of the two-level system.

novel theory for the collapse and the revival by considering relevant losses from the cavity especially the "Ohmic" loss [21] to the walls of the cavity, the leakage from the cavity, and the loss due to the spontaneous emission through the open surface of the cavity.

Multimode J-C model [18] is popular not only for an extension of the single-mode J-C model [12] but also for the multi-photon transitions [22], the dynamics of entanglement [23], *etc* [20]. Multimode J-C model has been successfully used by us [20] to explore the quantum Rabi oscillations of a two-level system interacting with a very low average number of injected coherent photons ($\bar{n} = 0.4$) in a lossy resonant cavity as described in Brune *et al*'s experiment [7]. Such a very low average number of photons was treated perturbatively (up to the second order in $\bar{n}^2 \ll 1$) in Ref. [20]. However, Brune *et al* [7] obtained two more sets of data for low average number of injected coherent photon numbers $\bar{n} = 0.85 \pm 0.04$ and $\bar{n} = 1.77 \pm 0.15$ in the same cavity showing the collapse and the revival of the quantum Rabi oscillations of a two-level system (^{87}Rb atom). A non-perturbative method is needed for the theoretical explanation of these two sets of data for the collapse and the revival. Hence we extend our method described in Ref. [20] for this purpose. The collapse and the revival of the quantum Rabi oscillations, of course, were not discussed in Ref. [20].

Calculation in this chapter begins with Eqn. (3.8) of the previous chapter. This equation is an outcome of the multimode J-C model and it is nothing but the net transition probability ($P_{2\to1}(t)$) which describes the quantum Rabi oscillations in time (t) domain for a two-level system interacting with coherent photons in a lossy resonant cavity. This transition probability is a function of time and a number of parameters including the renormalized coupling constant which can be determined by the mode quality factor of the cavity and the average number of coherent photons incident on the two-level system. We determine the transition probabilities for the average numbers of coherent photons $\bar{n} = 0.85$ and $\bar{n} = 1.77$ and the mode quality factor $Q = 7 \times 10^7$ after determining the renormalized coupling constants within a graphical method. We compare our theoretical results with the experimental data obtained by Brune *et al* [7] and the existing theoretical results obtained within the single-mode J-C model [7, 8, 14]. We also estimate the collapse time and the revival time for $\bar{n} = 0.85$ and 1.77. Finally, we conclude.

4.2 Collapse and Revival

Let us consider the two-level system in a lossy resonant Fabry-Perot cavity of the resonance frequency ω_0 and the mode quality factor Q. The two-level system is interacting with the coherent photons which are injected through a hole on the cavity axis. Let the average number of coherent photons injected on the two-level system be $\bar{n}$. We consider the quantum Rabi oscillations of the two-level system in the processes of the spontaneous emission, the stimulated emission and the stimulated absorption. The quantum Rabi oscillations need the two-level system to strongly interact with the injected photons of the cavity field. The high mode quality factor of the cavity ensures strong light-matter coupling. The photon emitted from the two-level has a long life-time ($\sim 200\ \mu$s) in such a situation. The emitted photon repeatedly reflects back and forth with the mirrors of the cavity before it leaks out through the holes on the axis of the cavity or becomes absorbed (or scattered) in the walls of the cavity resulting in the "Ohmic" loss [21]. However, the curved surface of the cylindrical geometry of the cavity is also kept open. This causes additional loss from the cavity. This loss is associated with the spontaneous emission from the two-level system through the curved surface of the cavity [20]. The probability that an emitted photon escapes from the cavity through the curved surface is $p_0 = \frac{2\pi r h}{2\pi r h + 2\pi r^2} = \frac{1}{1+\frac{r}{h}}$ where r is the radius of each of the mirrors of the cavity and h is the separation of the two mirrors [20]. All these losses result in the net quality factor as $Q' = \frac{1}{\frac{1}{Q}+\frac{p_0 A(0)}{\omega_0}}$ [20] where $A(0)$ is the frequency broadening due to the natural decay in the space inside the cavity. Here $A(0)$ is nothing but the enhanced value of the Einstein A coefficient due to the Purcell effect [24]. Let us consider that initially ($t = 0$) the two-level system was at the excited state. Thus we get the net transition probability of two-level system from the excited state ($|\psi_2\rangle$) to the ground state ($|\psi_1\rangle$) at time t, as [20]

$$\begin{aligned} P_{2\to 1}(t) &= A(0) \sum_{n=0}^{\infty} \frac{4}{\pi} p_n [n+1] \times \\ & \int_{\omega_n}^{\infty} \frac{(\omega_0/Q')^2}{4(\Omega_n^2 - \omega_n^2) + (\frac{\omega_0}{Q'})^2} \frac{\sin^2(\Omega_n t/2)}{\Omega_n \sqrt{\Omega_n^2 - \omega_n^2}} \mathrm{d}\Omega_n \end{aligned} \tag{4.1}$$

where $\omega_n = 2g'_{\omega_0}(\bar{n}, Q')\sqrt{n+1}$ is the renormalized n-photon Rabi frequency, $p_n = \frac{\bar{n}^n}{n!}\mathrm{e}^{-\bar{n}}$ represents the probability of occupation of n coherent photons, and $g'_{\omega_0}(\bar{n}, Q')$ is the renormalized light-matter coupling constant.

Eqn. (4.1) was already there in the previous chapter (Eqn. (3.8)) and it is able to describe the collapse and revival of the quantum Rabi oscillations of the two-level system in the lossy resonant cavity for the low average number of coherent photons ($\bar{n} \gtrsim 1$). The descriptions of the collapse and the revival require determination of the renormalized coupling constant which, however, was not done in any literature for the low average number of coherent photons ($\bar{n} \gtrsim 1$). We determine renormalized coupling constants for low average numbers of coherent photons as follows. We also describe the collapse and the revival below.

For $t \to \infty$, we have $\sin^2(\Omega_n t/2) \to \frac{1}{2}$ in Eqn. (4.1). Experimental results suggest us to take the limiting value $\lim_{t\to\infty} P_{2\to 1}(t) = 1/2$ [7, 13]. Now by setting $P_{2\to 1}(\infty) = 1/2$ and integrating over Ω_n in Eqn. (4.1), we get

$$f(g'_{\omega_0}) = \frac{1}{2} \tag{4.2}$$

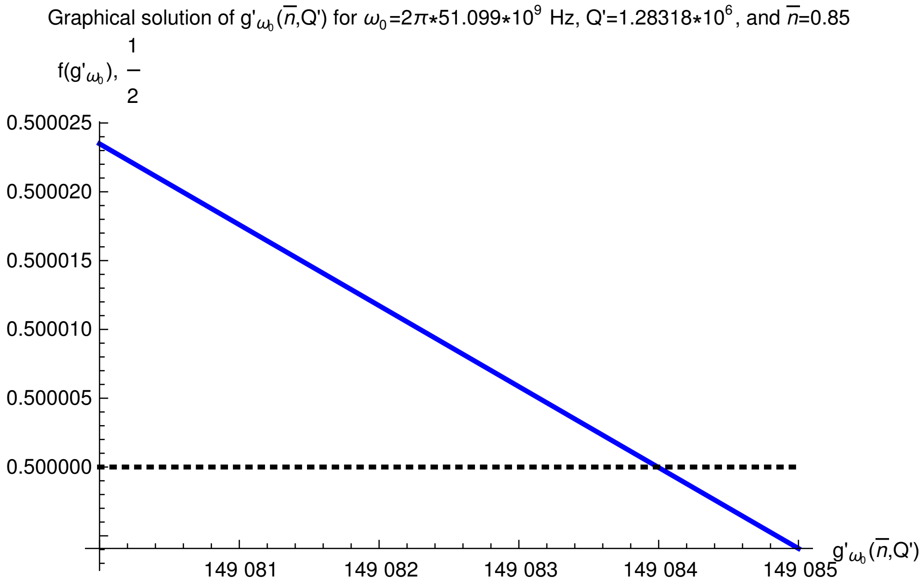


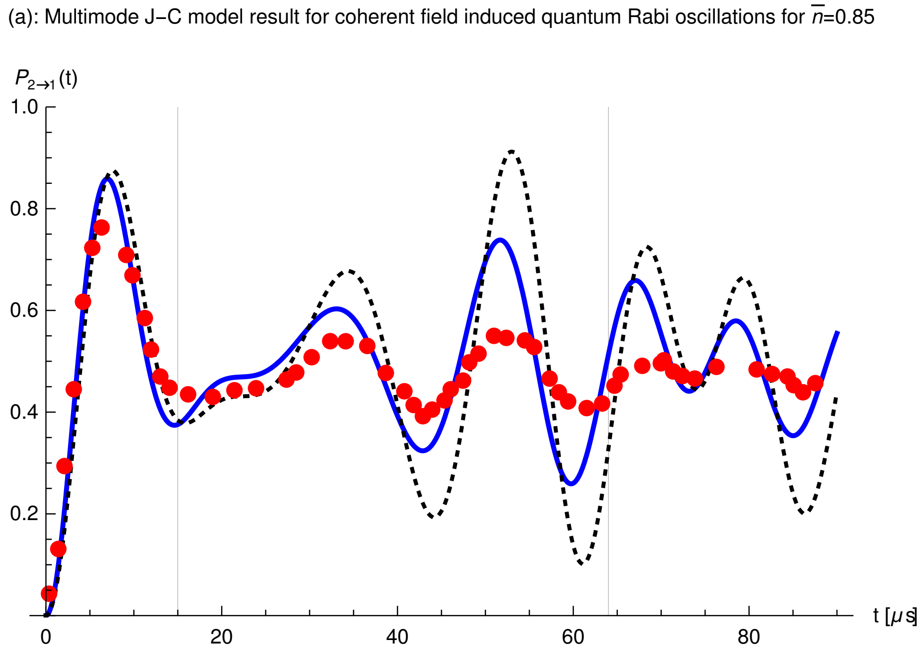
FIGURE 4.2: Solid line represents the left-hand side of Eqn. (4.2) for the parameters as mentioned in the plot label and for $A(0) = 0.473053 \times 10^6$ Hz. Dotted line represents the right-hand side of the same equation for the same parameters.

where

$$f(g'_{\omega_0}) = \sum_{n=0}^{\infty} \frac{A(0)[\omega_0/Q']\mathrm{e}^{-\bar{n}}\bar{n}^n}{2g'_{\omega_0}(\bar{n},Q')[4g'_{\omega_0}(\bar{n},Q') + \frac{\omega_0}{Q'\sqrt{1+n}}]n!}. \tag{4.3}$$

The Einstein A coefficient was calculated from the experimental data [7] for the 'vacuum' Rabi oscillation of the two-level system (^{87}Rb atom) for the Rabi frequency $\Omega_R = 2\pi \times 47 \times 10^3$Hz, the Bohr frequency as well as resonance frequency $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz, the average number of thermal photons $\bar{n}_{th} = 0.0489$, and the cavity specification $r = 25$ mm, $h = 27$ mm and $Q = 7 \times 10^7$, as $A(0) = 0.473053 \times 10^6$ Hz [20]. The net quality factor is resulted in as $Q' = 1.28318 \times 10^6$ out of all these parameters [20]. The experiment on the collapse and revival of the quantum Rabi oscillations of the same two-level system was done in the same setup except the thermal photons replaced with the injected coherent photons. Such a replacement does not change the Einstein A coefficient rather changes the renormalized coupling constant $g'_{\omega_0}(\bar{n},Q')$ as well as the Rabi frequency $\Omega_R = 2g'_{\omega_0}(\bar{n},Q')\sqrt{\bar{n}+1}$ [20].

Let us now determine the renormalized coupling constant $g'_{\omega_0}(\bar{n},Q')$ from Eqn. (4.2) for fixed $\bar{n}$ and for $A(0) = 0.473053 \times 10^6$ Hz, $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz and $Q' = 1.28318 \times 10^6$ as mentioned above. We already have mentioned that Brune *et al* took $\bar{n} = 0.85 \pm 0.04$ and 1.77 ± 0.15 for the observations of the collapse and revival [7]. We employ the graphical method for the determination of the renormalized coupling constant from Eqn. (4.2) for the above parameters. This method is considered to be a non-perturbative method. We plot both the left-hand side (solid line) and the right-hand side (dotted line) of Eqn. (4.2) with respect to the renormalized coupling constant in figure 4.2 for the above parameters and for $\bar{n} = 0.85$. The intersection



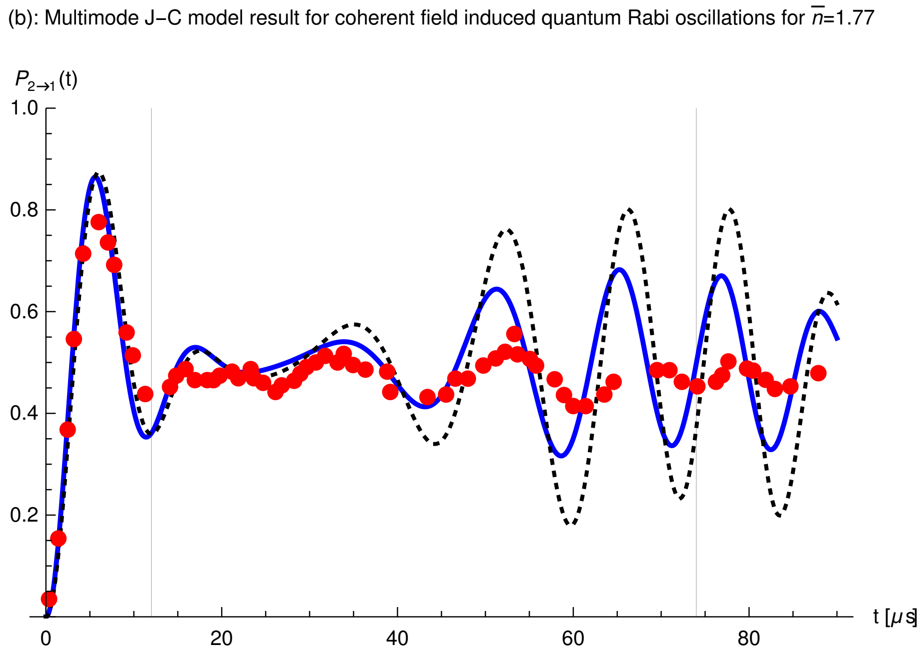


FIGURE 4.3: (a): Solid line represents coherent field induced quantum Rabi oscillations, and it follows Eqn. (4.1) for the Bohr frequency $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz, renormalized coupling constant $g'_{\omega_0} = 149084$ Hz, Rabi frequency $\Omega_R = 2\pi \times 64.5457 \times 10^3$ Hz and average number of coherent photons $\bar{n} = 0.85$ in a lossy resonant cavity of specification $r = 25$ mm, $h = 27$ mm and $Q = 7 \times 10^7$. Circles represent corresponding experimental data [7] adapted for the circular Rydberg states (with the principal quantum number $n = 50$ and $n = 51$) of ^{87}Rb atoms in the lossy resonant cavity. Dotted line follows Eqn. (4.4) for $g = g'_{\omega_0} = 149084$ Hz and $\bar{n} = 0.85$.
(b): Solid line represents coherent field induced quantum Rabi oscillations, and it follows Eqn. (4.1) for the Bohr frequency $\omega_0 = 2\pi \times 51.099 \times 10^9$ Hz, renormalized coupling constant $g'_{\omega_0} = 152852$ Hz, Rabi frequency $\Omega_R = 2\pi \times 80.9769 \times 10^3$ Hz and average number of coherent photons $\bar{n} = 1.77$ in the same lossy resonant cavity. Circles represent corresponding experimental data for the same system. Dotted line follows Eqn. (4.4) for $g = g'_{\omega_0} = 152852$ Hz and $\bar{n} = 1.77$.

of these two plots solves the renormalized coupling constant on the horizontal axis of figure 4.2. Thus for $\bar{n} = 0.85$ we get the renormalized coupling constant to be as $g'_{\omega_0} = 149084$ Hz. This graphical solution can be called as a numerical solution because the left-hand side of Eqn. (4.2) is evaluated numerically before being plotted in figure 4.2. Similarly, for the same parameters except for $\bar{n} = 1.77$ we get the renormalized coupling constant to be as $g'_{\omega_0} = 152852$ Hz. Value of the renormalized coupling constant enables us to plot Eqn. (4.1) where periodic dephasing over the time for various photon numbers (n) results in the collapse in the quantum Rabi oscillations and periodic rephasing over the time for various photon numbers results in revival in the quantum Rabi oscillations. Multimodes further result in dephasing in the quantum Rabi oscillations. We plot Eqn. (4.1) in figure 4.3-a for $\bar{n} = 0.85$ and in figure 4.3-b for $\bar{n} = 1.77$. We compare our results (solid lines) with the corresponding sets of experimental data in figures 4.3-a and 4.3-b. We also need to compare our results with the existing theoretical result [7, 14, 8]

$$P_{2\to 1}(t) = \sum_{n=0}^{\infty} \frac{e^{-\bar{n}}\bar{n}^n}{n!} \sin^2\left(gt\sqrt{n+1}\right) \tag{4.4}$$

obtained for similar purpose under the consideration of the light-matter coupling with only the resonant mode ($\omega = \omega_0$) and no loss from the cavity. Dotted lines in figure 4.3-a and 4.3-b follow Eqn. (4.4) and represent single-mode J-C model results for the coherent field-induced quantum Rabi oscillations for the same coupling constants $\{g = g'_{\omega_0}\}$ and the same average photon numbers $\{\bar{n}\}$ taken for the solid lines. We have got these results published in Ref. [25].

The collapse happens at a point ($t = t_c$) when different quantum Rabi oscillations take place in π amount of out of phase. This causes destructive interference in the quantum Rabi oscillations. For low average number of coherent photons ($\bar{n} \gtrsim 1$) too, the maximum of the Poisson distribution $p_n = \frac{\bar{n}^n}{n!}e^{-\bar{n}}$ in Eqn. (4.1) occurs at around $n = \bar{n}$. The standard deviation for the distribution is $\triangle n = \sqrt{\bar{n}}$. Thus we apply the condition ($[\omega_{\bar{n}+\triangle n} - \omega_{\bar{n}-\triangle n}]t_c = \pi$ [26]) for the collapse in Eqn. (4.1), as

$$2g'_{\omega_0}(\bar{n}, Q')\left[\sqrt{\bar{n}+\sqrt{\bar{n}}+1} - \sqrt{\bar{n}-\sqrt{\bar{n}}+1}\right]t_c = \pi. \tag{4.5}$$

Here-from we estimate the collapse times as $t_c \simeq 15$ μs for the first set of data (for figure 4.3-a) and $t_c \simeq 12$ μs for the second set of data (for figure 4.3-b). Vertical lines at $t = 15$ μs and 12 μs indicate the collapse of the quantum Rabi oscillations in figures 4.3-a and 4.3-b, respectively. We have got these results published in Ref. [25].

The revival takes place at a point ($t = t_r$) when all the neighbouring quantum Rabi oscillations come in phase again and add up for constructive interference. Thus we apply the condition ($[\omega_{\bar{n}+1} - \omega_{\bar{n}}]t_r = 2\pi$ [14, 10, 26]) for the revival in Eqn. (4.1), as

$$2g'_{\omega_0}(\bar{n}, Q')\left[\sqrt{\bar{n}+2} - \sqrt{\bar{n}+1}\right]t_r = 2\pi. \tag{4.6}$$

Here-from we estimate the revival times as $t_r \simeq 64$ μs for the first set of data (for figure 4.3-a) and $t_r \simeq 74$ μs for the second set of data (for figure 4.3-b). Vertical lines at $t = 64$ μs and 74 μs indicate the revival of the quantum Rabi oscillations in figures 4.3-a and 4.3-b, respectively. We have got these results published in Ref. [25].

4.3 Conclusion

We have theoretically obtained multimode Jaynes-Cummings model results for the collapse and the revival of the quantum Rabi oscillations of a two-level system interacting with injected coherent photons in a lossy resonant cavity. We have extended our previous theory [20] within a non-perturbative method in this regard. We have compared our results with two sets of experimental data [7] for low average number of coherent photons ($\bar{n} = 0.85$ and 1.77) incident on a two-level system in the lossy resonant cavity. Our results match reasonably well with the experimental data, at least, better than the theoretical one (Eqn. (4.4) [7, 14, 8] obtained for only the resonant mode and no loss from the cavity under consideration.

We had to cite Ref. [20] quite often in this chapter because it is an extension of the previous work [20] on the quantum Rabi oscillations. This extension is necessary because the collapse and the revival of the quantum Rabi oscillations of a two-level system, however, have separate existence [1, 2, 3, 4, 5, 10, 11, 14, 15, 17] over the usual discussion on the quantum Rabi oscillations.

The solid line, which represents the function $f(g'_{\omega_0})$ of the renormalized coupling constant in figure 4.2, appears to be straight in the figure for the small range of the renormalized coupling constant. It would not have appeared to be a straight line if we had taken a large range of the renormalized coupling constant in the figure.

It is clear from figure 4.3 that the multimode Jaynes-Cummings model result is almost the same as the single-mode Jaynes-Cummings model result for short time-evolution of the net transition probability. These two results significantly differ at a large time. This implies that the non-resonant modes are significant at large times.

It appears that we have developed a theory for the collapse and the revival for the low average number of injected coherent photons. Eqns. (4.1) and (4.2) are our key results in this regard. However, nowhere in these two equations, even in the subsequent equations, we have considered $\bar{n}$ to be small. Hence our theory is applicable for all values of the average number of injected coherent photons.

Our results are significantly different from the previous theoretical results [7, 14, 8] from (i) the consideration of the multimodes around the resonant mode into account, and (ii) the consideration of the frequency broadening due to the "Ohmic" loss to the walls of the cavity, the leakage from the cavity, and the loss due to the spontaneous emission through the open surface of the cavity. Further consideration of substantial losses corresponding to the frequency broadening due to inhomogeneous light-matter coupling along the cavity axis, Doppler broadening due to the speed distribution of ^{87}Rb atoms in the cavity, thermal broadening, *etc* may improve our theory. Such an improvement is kept as an open problem.

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Chapter 5

Population dynamics of two-level systems for the generalized Einstein coefficients

5.1 Introduction

While the stimulated transition-rates are time-independent in the (quantum) Markovian master equations and are solvable for the case of the time-dependent perturbation on the system [1, 2, 3], they are time-dependent in the (quantum) non-Markovian master equations and are usually hard to solve [4, 5]. The explicit time-dependent terms in the stimulated transition rates render the non-Markovian master equations analytically intractable. Some simplified versions of the non-Markovian master equations can, of course, be solved either in the limiting cases of weak [6] and linear [7] interactions between the system and the bath or in the limiting case of structured bath even for strong interactions [8].

Nonequilibrium statistical mechanics is often modelled with the semiclassical or the quantum master equations which to some extent are generalizations of Einstein's rate equation, such as the Pauli master equation [9], the Boltzmann–Uehling–Uhlenbeck equation or the quantum statistical Boltzmann equation [9, 10], the Gorini–Kossakowski–Sudarshan–Lindblad equation [1], the Bloch–Redfield master equation [3, 2], the Caldeira–Leggett master equation [11, 2], the quantum Fokker–Planck equation [12], the adiabatic/nonadiabatic master equation [13], Van Hove master equation [14], and the Nakajima–Zwanzig master equation [4]. These equations are either of Markovian master equation or non-Markovian master equation [1] type irrespective of the strength of the system-bath (i.e., light-matter or matter-matter) coupling. None of these equations can be derived fully from either the Schrodinger equation or the Liouville–von Neumann equation or even the Heisenberg equation of motion because the system can not be found in a pure state in the thermal radiation field. These equations (so as Einstein's rate equations) are arrived purely from phenomenological point of view because (i) the bath is assumed to be not affected

[1] Here only Van Hove and Nakajima-Zwanzig master equations are listed to be of non-Markovian type.

by the (much smaller) system, and (ii) the effects of the bath-variables are averaged out with heuristically structured spectral line-shapes of the bath or in turn temporal correlations in the system within various approximations such as the Markov approximation, the Born approximation, *etc.*) [15]. The stimulated rate coefficients in the master equations, in general, are time-dependent within the finite time interval after commencement of the light-matter interactions. However, the fundamental processes remain phenomenologically same in both the weak coupling regime and the strong coupling regime even if the rate coefficients are time-dependent. Thus, we are generalizing Einstein's rate equations for the time-dependent rate coefficients for the two-level systems (atoms/molecules) in the thermal radiation field because the time-dependent coefficients are not bringing any other (new) fundamental processes into the consideration. Application of the generalized rate equations would be a useful model for studying nonequilibrium statistical mechanics for both the weak and the strong light-matter couplings. The generalized rate equations may be classified as a time-dependent semiclassical Markovian master equation as (i) the rates of the stimulated transitions are essentially derived from a semi-classical (Rabi) model and are found to be time-dependent, and (ii) all the occupation probabilities (P_1 and P_2) in the generalized rate equations are employed at the same time with no memory kernels in the equations.

In this chapter we describe the population dynamics of identical two-level systems (atoms/molecules) for the generalized Einstein coefficients. We are also aiming to study the entropy production of a two-level system from the population dynamics. The two-level systems are assumed to be distinguishable so that the systems obey classical statistics. The population dynamics of the two-level systems can be understood from the population dynamics of only one two-level system in such a situation. The two level systems, of course, are interacting with either the thermal radiation field or the monochromatic radiation field. We are considering two cases for the population dynamics. We are considering the (semicalssical) Rabi model, as described in Chapter-2, for case-I and the multimode Jaynes-Cummings model, as described in Chapter-3, for case-II.

For case-I, the generalized Einstein B coefficient obtained in Eqn. (2.11) within the Rabi model takes place in the time-dependent rate coefficients $R_{1\rightarrow 2}(t)$ and $R_{2\rightarrow 1}(t)$ in the Einstein rate equations if the time-independent rate coefficients are replaced by the time-dependent rate coefficients. The Einstein A coefficient can not be touched within the Rabi model. It is kept unaltered for this case. We study the population dynamics and entropy production for both the time-dependent forms of the B coefficient obtained in Eqn. (2.11) for the thermal radiation field and in Eqn. (2.13) for the monochromatic radiation field.

For case-II, the generalized Einstein A and B coefficients obtained in Eqns. (3.5) and (3.6) take place in the time-dependent rate coefficients $R_{1\rightarrow 2}(t)$, $R_{2\rightarrow 1}(t)$, and $A(t)$ in the Einstein rate equations if the time-independent rate coefficients are replaced by the time-dependent rate coefficients. We study the population dynamics and entropy production for the time-dependent forms of the generalized Einstein coefficients obtained in Eqns. (3.5) and (3.6).

5.2 Case-I: Population dynamics and entropy production of a two-level system within the Rabi model

In this section we aim (i) to generalize Einstein's rate equations with the generalized Einstein B coefficient for the two-level system in the thermal radiation field and (ii) to describe population dynamics of the system by finding exact analytic solutions of the rate equations. It should be mentioned in this regard that, the thermal radiation field is not coherent. The time-dependence of the transition probability, which is related to the population dynamics, is also of high interest, and has been experimentally investigated by Brune *et al* for a two-level system (atom) in the coherent radiation field (and 'vacuum' field) in a high-Q cavity [16]. Theoretical explanation of the population dynamics has been found numerically by Escher and Ankerhold [8] in connection with the dissipative quantum dynamics of the two-level system interacting with a structured reservoir consisting of damped harmonic modes. However, we are considering the light-matter interactions at the semi-classical level, so that, the population dynamics can be analytically described in a fairly accurately manner, at least for $\omega_0 \gg \Omega_R$ and $k_B T \gtrsim \hbar\Omega_R$.

5.2.1 Population dynamics within the Rabi model

We already have mentioned in Chapters 2 and 3 that the bare energy eigenstates of the two-level system (atom/molecule) of our interest are given by $|\psi_1\rangle$ and $|\psi_2\rangle$. While $|\psi_1\rangle$ corresponds to the energy eigenvalue E_1, $|\psi_2\rangle$ corresponds to the energy eigenvalue E_2 (such that $E_2 > E_1$). The Bohr frequency of the two-level system is given by $\omega_0 = [E_2 - E_1]/\hbar$. The rate of stimulated emission from the state $|\psi_2\rangle$ to the state $|\psi_1\rangle$ at time t is given by $R_{2\to1}(t)$. The rate of (stimulated) absorption from the state $|\psi_1\rangle$ to the state $|\psi_2\rangle$ at time t is given by $R_{1\to2}(t)$. The Rabi frequency of the two-level system, of course, is given by Ω_R. Occupation probabilities of the two-level systems are given by $P_1(t)$ for the state $|\psi_1\rangle$ and $P_2(t)$ for the state $|\psi_2\rangle$.

It is to be noted that Ω_R, however small, can greatly influence the time-evolution of the statistical mechanical occupation probabilities $P_1(t)$ and $P_2(t)$ of the states $|\psi_1\rangle$ and $|\psi_2\rangle$ respectively even for the case of thermal radiation field. Time-evolution of the occupation probabilities are to be determined from Einstein's rate (master) equations [17, 18, 19] which are now revised with the time-dependent stimulated transition rates as

$$\frac{\mathrm{d}P_2}{\mathrm{d}t} = -AP_2(t) - R_{2\to1}(t)P_2(t) + R_{1\to2}(t)P_1(t) \tag{5.1}$$

and

$$\frac{\mathrm{d}P_1}{\mathrm{d}t} = AP_2(t) + R_{2\to1}(t)P_2(t) - R_{1\to2}(t)P_1(t) \tag{5.2}$$

where $A = \frac{\omega_0^3 d_{12}^2}{3\pi\epsilon_0\hbar c^3}$ [20, 18] is the original (time-independent) Einstein's A coefficient which represents the rate of spontaneous emission from the upper level to the lower level due to the quantum (vacuum) fluctuations. The time-evolution of these probabilities, because of the constraint $P_1(t) + P_2(t) = 1$, can be solely determined from any one of the above two equations, say Eqn. (5.1), with $P_1(t)$ replaced by $1 - P_2(t)$. Thus, we recast Eqn. (5.1) with the time-dependent rates of stimulated absorption and stimulated emission, $R(t) = R_{1\to2}(t) = R_{2\to1}(t)$ as

$$\frac{\mathrm{d}P_2}{\mathrm{d}t} = R(t) - (A + 2R(t))P_2(t). \tag{5.3}$$

The rate of stimulated transitions was found in Ref. [21] to be time-independent: $R(t) = R(0) = B_{12}u(\omega_0) = B_{21}u(\omega_0) \geq 0$, within the first order time-dependent perturbation theory of quantum mechanics [18]. Here $B_{12} = B_{21}$ is the original Einstein B coefficient and $u(\omega_0)$ is the average energy density of the thermal photons per unit (angular) frequency interval around ω_0. Weisskopf and Wigner determined the rate coefficient A within the domain of quantum field theory [21, 20]. Eqn. (5.3), in such a case, has a physical solution with the initial condition: $P_2(0) = 0$, as [17, 18]

$$P_2(t) = \frac{R(0)}{A + 2R(0)}[1 - \mathrm{e}^{-[A+2R(0)]t}] \tag{5.4}$$

which is often equated with the (time-independent) Boltzmann probability, $P_2(\infty) = \frac{\mathrm{e}^{-E_2/k_BT}}{\mathrm{e}^{-E_1/k_BT}+\mathrm{e}^{-E_2/k_BT}}$, in thermodynamic equilibrium for $t \to \infty$ [17, 18]. Occupation probability of the lower level, on the other hand, can be given by $P_1(t) = 1 - P_2(t)$. Eqn. (5.4) is Einstein's semiclassical result for the occupation probability [17, 18]. Let us call the time-dependent probabilities, $P_1(t)$ and $P_2(t)$, which follow from Eqn. (5.4), as Einstein probabilities. Dotted lines in figures 5.1 and 5.2 represent the Einstein probabilities. Our aim for the rest of the subsection is to modify the Einstein probabilities due to the presence of the Rabi flopping in the same system within the semiclassical description.

For the two-level system in the thermal radiation field

We solve Eqn. (5.3) with the initial condition $P_2(0) = 0$, for the stimulated transition rate $R(t)$ in Eqn. (2.10) as

$$\begin{aligned} P_2(t) &= |R(0)|\mathrm{e}^{-At-2R(0)f_{\Omega_R}(t)} \\ &\quad \times \int_0^t \mathrm{e}^{A\tau+2R(0)f_{\Omega_R}(\tau)}|J_0(\Omega_R\tau)|\mathrm{d}\tau \end{aligned} \tag{5.5}$$

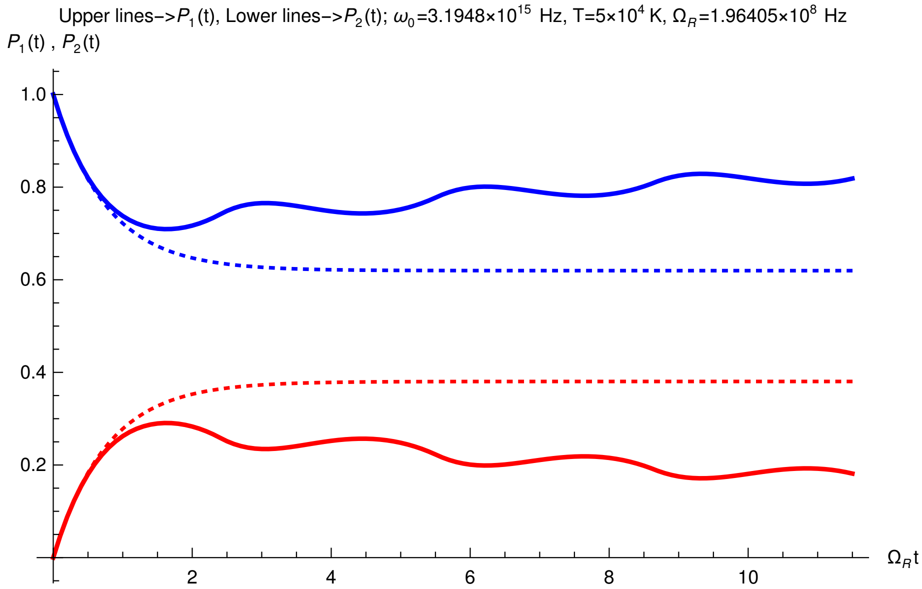

FIGURE 5.1: Occupation probabilities for the $3s_{\frac{1}{2}}$ and $3p_{\frac{1}{2}}$ states of an ^{23}Na atom in the thermal radiation field with the condition that the system initially was in the lower level. Lower and upper solid lines follow Eqns. (5.5) and (5.7), respectively, for the parameters as mentioned in the figure corresponding to $d_{12} = 2.5ea_0 = 2.1196 \times 10^{-29}$Cm [22]. Lower and upper dotted lines represent Einstein probabilities for the same system, and follow Eqn. (5.4) and its follow-up respectively.

where $f_{\Omega_R}(t)$ is given by

$$\begin{aligned} f_{\Omega_R}(t) &= {}_1F_2\left(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-\frac{\Omega_R^2 t^2}{4}\right)\left[2\mathrm{U}(J_0(\Omega_R t))-1\right]t \\ &\quad -\sum_{j=1}^{\lfloor \Omega_R t\rfloor}\left[(-1)^j \gamma_{0,j}\, {}_1F_2\left(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-\frac{\gamma_{0,j}^2}{4}\right)\right. \\ &\quad \left.\times \mathrm{U}(\Omega_R t-\gamma_{0,j})\right]\frac{2}{\Omega_R} \end{aligned} \tag{5.6}$$

where $\gamma_{0,j}$ is the jth zero of J_0 and U is the unit step function. Now, we get the occupation probability of the lower level from Eqn. (5.5) as

$$P_1(t) = 1 - P_2(t). \tag{5.7}$$

Eqns. (5.5) and (5.7) are our semiclassical results for the occupation probabilities of the two states of the two-level system in the thermal radiation field. We have got these results published in Ref. [23]. We plot these probabilities in figure 5.1 for the relevant values of the parameters for the $3s_{\frac{1}{2}}$ and $3p_{\frac{1}{2}}$ states of an ^{23}Na atom. For this plot, we have purposefully considered the temperature to be very high ($T = 5 \times 10^4$ K) so that both the rates of spontaneous ones ($A/\Omega_R = 0.314566$) and stimulated ones ($R(0)/\Omega_R = 1/2$) are comparable to the Rabi flopping frequency to show oscillations in the occupation probabilities. An ^{23}Na atom is not expected to be ionized even in such a high temperature, as its first ionization potential is 5.1 eV=59183 k_BK. While the occupation probability ($P_2(t)$) of the upper level asymptotically (i.e., for $\Omega_R t \gg 1$) vanishes as $\frac{R(0)}{A}\sqrt{\frac{2}{\pi\Omega_R t}}$, the occupation probability ($P_1(t)$) of the lower level asymptotically reaches unity as $1 - \frac{R(0)}{A}\sqrt{\frac{2}{\pi\Omega_R t}}$. It is clear from figure 5.1 that the occupation probabilities of the two-level system are significantly deviating from the Einstein probabilities (as well as the Boltzmann probabilities) as time evolves and the system goes away from thermodynamic equilibrium as a consequence of the Rabi flopping with non-zero frequency. Our results, of course, match with Einstein probabilities if Rabi flopping is turned off, i.e., if $\Omega_R \to 0$.

For the two-level system in the monochromatic radiation field

On the other hand, for the case of the monochromatic wave, we solve Eqn. (5.3) with the initial condition $P_2(0) = 0$ for $R(t)$ in Eqn. (2.12) as

$$\begin{aligned} P_2(t) &= R(0)\mathrm{e}^{-At-2R(0)g_{\Omega_R}(t)} \\ &\times \int_0^t \mathrm{e}^{A\tau+2R(0)g_{\Omega_R}(\tau)}|\sin(\Omega_R\tau)|\mathrm{d}\tau \end{aligned} \tag{5.8}$$

where $g_{\Omega_R}(t)$ is given by

$$\begin{aligned} g_{\Omega_R}(t) &= \frac{1-\cos(\Omega_R t)}{\Omega_R}[2\mathrm{U}(\sin(\Omega_R t)) - 1] - \frac{2}{\Omega_R} \\ &\times \sum_{j=1}^{\lfloor \Omega_R t \rfloor} (-1)^j[1-\cos(j\pi)]\mathrm{U}(\Omega_R t - j\pi). \end{aligned} \tag{5.9}$$

For this case of monochromatic radiation field too, we have $P_1(t) = 1 - P_2(t)$. These occupation probabilities are semiclassical (but not statistical mechanical) for the study of the single frequency in the monochromatic wave. We have got these results published in Ref. [23]. We plot these semiclassical probabilities in figure 5.2 for the relevant values of the parameters for the same system. It is clear from this figure that, the semiclassical probabilities oscillate near the corresponding Einstein probabilities without decay of their amplitudes. Thus, the two-level system (atom/molecule) neither in thermal radiation field nor in the monochromatic radiation field equilibrate with the surroundings as long as the Rabi flopping frequency is non-zero.

5.2.2 Entropy production within the Rabi model

Although the two-level system in the rapidly oscillating electromagnetic field makes transitions (if frequency of the oscillations is close to the Bohr frequency of the two levels), the transitions occur over a much larger time scale ($t \sim 1/\Omega_R$). Thus, it is not exactly known when the system would make a transition. Instead, we know the probability of the transition and consequently, the occupancy of the two states

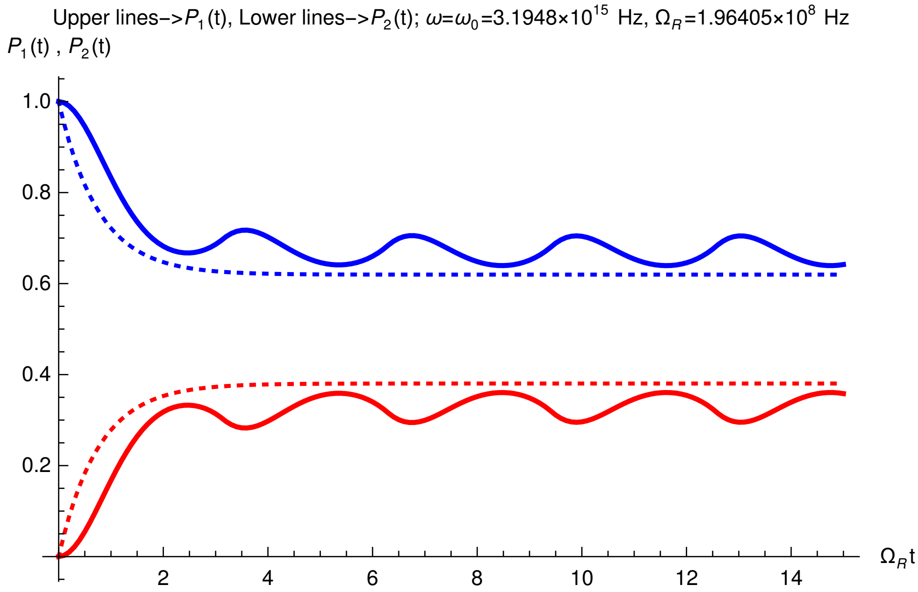

FIGURE 5.2: Lower and upper solid lines represent occupation probabilities, and follow Eqn. (5.8) and its follow-up for the same parameters of the two-level system at the resonance in the monochromatic radiation field as mentioned in figure 5.1. Adjacent dotted lines represent corresponding Einstein probabilities, and follow Eqn. (5.4) and its follow-up respectively.

becomes probabilistic. This loss of information can be quantified by the entropy production of the system. The entropy production of the two-level system either in the thermal radiation field or in the monochromatic radiation field can be written, by following the Pauli–von Neumann formalism of nonequilibrium statistical mechanics as [9, 24, 19]

$$S(t) = -k_B[P_1(t)\ln(P_1(t)) + P_2(t)\ln(P_2(t))]. \tag{5.10}$$

We illustrate the time-dependence of the entropy in figure 5.3 for both the cases. The result corresponding to the monochromatic case is shown in the inset of figure 5.3. Pauli proved the quantum mechanical H-theorem (i.e., $\frac{dS(t)}{dt} \geq 0$) even for a single atom/molecule (say, a two-level system) in the radiation field by introducing the Pauli master equation (which is analogous to Einstein's rate equation for $A = 0$) [9, 10]. Here the rates of stimulated transitions to be time-independent for this purpose [9].

However, the two-level system in thermal (or monochromatic) radiation field does not fully evolve spontaneously. The stimulated transitions have control over the evolution of the system. Moreover, the spontaneous emission favour the lower level, as clear from figure 5.1, once control of the thermal (broad band) radiation to the stimulated transitions is damped (as $\sim 1/\sqrt{\Omega_R t}$) after sufficiently long time ($\Omega_R t \gg 1$). Such a damping of the Rabi flopping in free space (large blackbody cavity) is caused due to the finite width ($\sim \Omega_R$) of the frequency distribution around the resonance because all the frequency components of the thermal radiation field

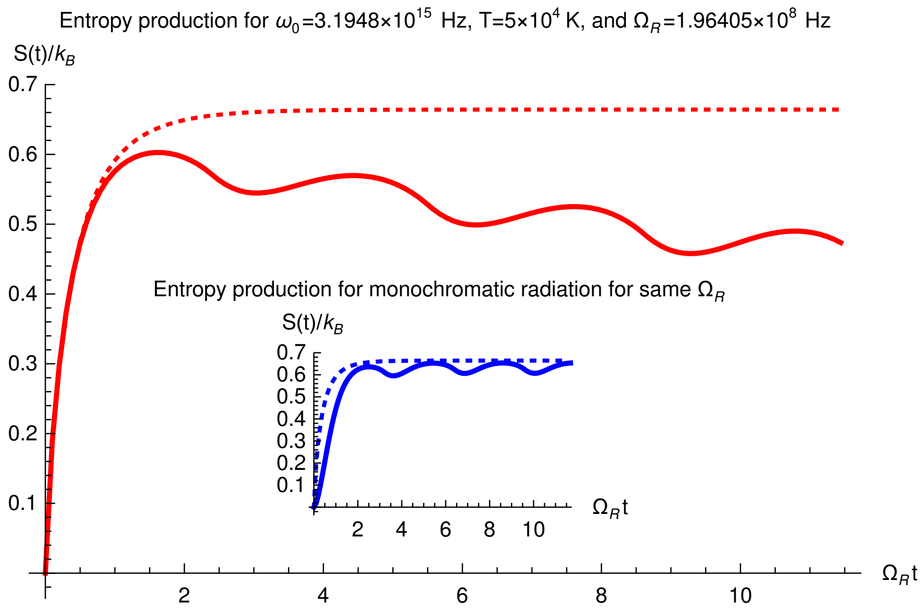

FIGURE 5.3: Entropy production for the $3s_{\frac{1}{2}}$ and $3p_{\frac{1}{2}}$ states of an ^{23}Na atom in the thermal radiation field. Plots follow from Eqn. (5.10) for the parameters as mentioned in figure 5.1. Dotted lines represent the same obtained from Einstein probabilities (Eqn. (5.4) and its follow-up).

incoherently contribute to the resultant Rabi flopping. Thus, a nonzero finite value of the Rabi flopping frequency causes extraordinary favour on top of the spontaneous transitions to the lower level after sufficiently long time, and consequently, the entropy ($S(t)$) of the two-level system, instead of always increasing with time, asymptotically (i.e., for $\Omega_R t \gg 1$) vanishes as $k_B \frac{R(0)}{A}\sqrt{\frac{2}{\pi\Omega_R t}}\left[1 - \ln\left[\frac{R(0)}{A}\sqrt{\frac{2}{\pi\Omega_R t}}\right]\right]$. This feature is apparent in figure 5.3 where we also have plotted the entropy production based on the Einstein probabilities. Such a damping, however, is not possible for the monochromatic wave, as clear from figure 5.2, as there is no frequency distribution of the incident waves which causes damping to the Rabi flopping. Thus the Rabi flopping causes oscillations of the entropy near the non-decreasing semi-classical result (having the saturation value $k_B \ln(2)$) as clear in the inset of figure 5.3. We have got these results published in Ref. [23]. All the oscillations or the damping are caused for nonzero finite value of the Rabi flopping frequency. Thus, if $\Omega_R \to 0$, we again get back Einstein's semiclassical result. It is clear from figure 5.3 that the entropy of the system is not always increasing. This is not a surprise because the two-level system we are considering is no longer a thermodynamically isolated system rather it is open to interact with the radiation field. The second law of thermodynamics is not applicable to such a system.

A question naturally arises: whether there would be any change in the occupation probabilities if we take an alternative initial condition such that initially the two-level system is at the upper level (i.e., $P_1(0) = 0, P_2(0) = 1$) like that in Eqn. (2.2). The Rabi flopping frequency would not certainly change under this alternation. The rate of transitions $R(t)$ would not also change. However, some of the results would change, e.g., an additional term $e^{-At-2R(0)f_{\Omega_R}(t)}$ would have to be added to the r.h.s. of Eqn. (5.5) [2]. The two solid lines both in figure 5.1 and figure 5.2 would intersect keeping their individual tails unaltered.

5.3 Case-II: Population dynamics and entropy production of a two-level system within the multimode Jaynes-Cummings model

While the generalized Einstein B coefficients are same and time-dependent within the (semiclassical) Rabi model, the Einstein A coefficient remains the original time-independent A coefficient in the same model [23]. This causes even a small Ω_R to greatly influence the time-evolution of the statistical mechanical occupation probabilities $P_1(t)$ and $P_2(t)$ of the two states $|\psi_1\rangle$ and $|\psi_2\rangle$, respectively [3]. However, in Section 3.3.2 we have all the generalized Einstein coefficients to be time-dependent within the (quantum mechanical) multimode Jaynes-Cummings (J-C) model. Let us now investigate how the occupation probabilities ($P_1(t)$ and $P_2(t)$) of the two-level system in the thermal radiation field evolve with time for the multimode J-C model results of the generalized Einstein coefficients obtained in Eqns. (3.5) and (3.6).

5.3.1 Population dynamics within the multimode Jaynes-Cummings model

Time-evolution of the occupation probabilities ($P_1(t)$ and $P_2(t)$) of the two-level system in the thermal radiation field are to be determined from Einstein's rate (master) equations [17, 18, 19] which are now revised with the generalized Einstein coefficients in Eqns. (3.5) and (3.6) as

$$\begin{aligned}\frac{\mathrm{d}P_2}{\mathrm{d}t} &= -A(t)P_2(t) - u(\omega_0)B_{21}(t)P_2(t)\\ &\quad +u(\omega_0)B_{12}(t)P_1(t) \end{aligned} \tag{5.11}$$

and

$$\begin{aligned}\frac{\mathrm{d}P_1}{\mathrm{d}t} &= A(t)P_2(t) + u(\omega_0)B_{21}(t)P_2(t)\\ &\quad -u(\omega_0)B_{12}(t)P_1(t) \end{aligned} \tag{5.12}$$

where $P_1(t)$ is the occupation probability of the bare state $|\psi_1\rangle$ of the two-level system at time t, $P_2(t)$ is the occupation probability of the bare state $|\psi_2\rangle$ of the two-level system at time t, ω_0 is the Bohr frequency of the two-level system, $A(t)$ is the generalized Einstein A coefficient as obtained in Eqn. (3.6), $B_{21}(t)$ is the generalized Einstein B coefficient as obtained in Eqn. (3.5), and $u(\omega_0)$ is the average energy density of the thermal photons per unit (angular) frequency interval around ω_0. We also

[2] An additional term $e^{-At-2R(0)g_{\Omega_R}(t)}$ would have to be added to the r.h.s. of $P_2(t)$ in Eqn. (5.8).

[3] See the previous section for the same.

have $B_{12}(t) = B_{21}(t)$ as discussed below Eqn. (3.6). Time-evolution of the occupation probabilities, because of the constraint $P_1(t) + P_2(t) = 1$, can be solely determined from any one of the above two equations, say Eqn. (5.11), with $P_1(t)$ be replaced by $1 - P_2(t)$. Thus, we recast Eqn. (5.11) with the spontaneous emission rate $A(t)$ and the stimulated emission rate $R(t) = B_{21}(t)u(\omega_0) = B_{12}(t)u(\omega_0)$ as

$$\frac{dP_2}{dt} = R(t) - [A(t) + 2R(t)]P_2(t). \tag{5.13}$$

While $R(0)$ is the rate-coefficient for stimulated emission/absorption found within the first order time-dependent perturbation theory of quantum mechanics [21], $A(0)$ is the rate-coefficient for the spontaneous emission found within the first order perturbation theory[4] of quantum electrodynamics [20]. Eqn. (5.13) has a physical solution for $R(t) = R(0)$ and $A(t) = A(0)$ with the initial condition $P_2(0) = 1$ as [17, 18]

$$\begin{aligned} P_2(t) &= \frac{R(0)}{A(0) + 2R(0)} \\ &\quad + \left[1 - \frac{R(0)}{A(0) + 2R(0)}\right] \mathrm{e}^{-[A(0)+2R(0)]t} \end{aligned} \tag{5.14}$$

which is often equated with the (time-independent) Boltzmann probability $P_2(\infty) = \frac{\mathrm{e}^{-E_2/k_B T}}{\mathrm{e}^{-E_1/k_B T} + \mathrm{e}^{-E_2/k_B T}}$ in thermal equilibrium for $t \to \infty$ [17, 18]. Occupation probability of the lower level, on the other hand, can be given by $P_1(t) = 1 - P_2(t)$. Eqn. (5.14) is Einstein's semiclassical result for the occupation probability [17, 18]. Let us call the time-dependent probabilities $P_1(t)$ and $P_2(t)$ which follow from Eqn. (5.14), as Einstein probabilities [17, 18]. Dotted lines in figure 5.4 represent the Einstein probabilities. It is clear from Eqns. (3.5) and (3.6) that, $R(t) = R(0)$ and $A(t) = A(0)$ are possible only when $\Omega_R \to 0$ i.e. when there is no Rabi oscillation. Our aim for the rest of the subsection is to modify the Einstein probabilities due to the presence of the Rabi flopping in the same system within the quantum field theoretic description of the multimode J-C model.

We solve Eqn. (5.13) with the initial condition $P_2(0) = 1$ for $B_{21}(t) = B_{12}(t) = R(t)/u(\omega_0)$ of Eqn. (3.5) and $A(t)$ of Eqn. (3.6) as

$$\begin{aligned} P_2(t) &= \mathrm{e}^{-[A(0)+2R(0)]f_{\Omega_R}(t)} \Bigg[1 + R(0) \times \\ &\quad \int_0^t \mathrm{e}^{[A(0)+2R(0)]f_{\Omega_R}(\tau)} |J_0(\Omega_R \tau)| \mathrm{d}\tau \Bigg] \end{aligned} \tag{5.15}$$

[4] The first order perturbation theory is compatible with Fermi's golden rule.

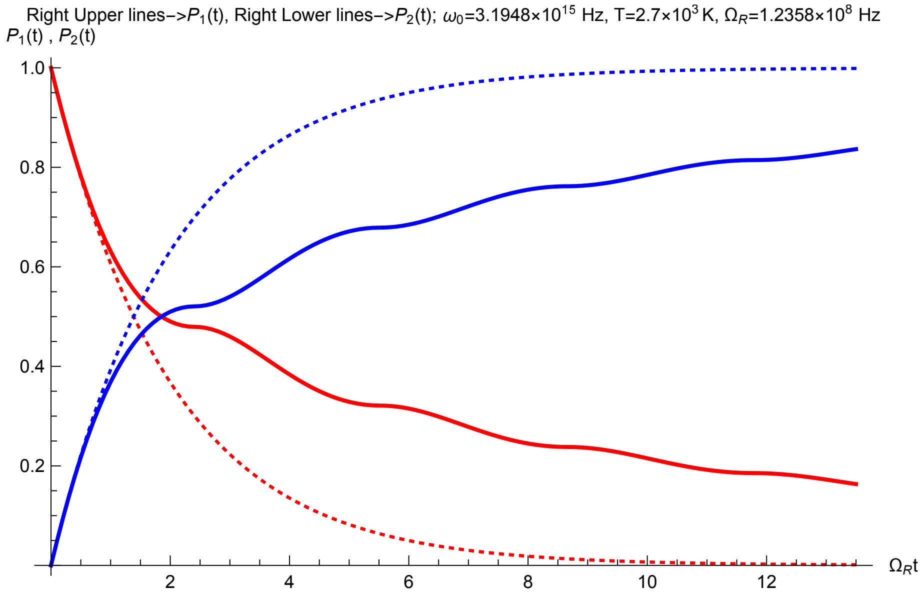

FIGURE 5.4: Occupation probabilities for the $3s_{\frac{1}{2}}$ and $3p_{\frac{1}{2}}$ states of an ^{23}Na atom in the thermal radiation field with the condition that the system initially was in the upper level. Right lower and right upper solid lines follow Eqns. (5.15) and (5.17), respectively for the parameters as mentioned in the figure corresponding to $d_{21} = 2.5ea_0 = 2.1196 \times 10^{-29}$Cm [22]. Lower and upper dotted lines represent Einstein probabilities for the same system, and follow Eqn. (5.14) and its complementary, respectively.

where $f_{\Omega_R}(t)$ is given by

$$\begin{aligned} f_{\Omega_R}(t) \quad = \quad & {}_1F_2\left(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-\frac{\Omega_R^2 t^2}{4}\right)\left[2\mathrm{U}(J_0(\Omega_R t))-1\right]t \\ & -\frac{2}{\Omega_R}\sum_{j=1}^{\lfloor \Omega_R t \rfloor}\left[(-1)^j \gamma_{0,j}\,{}_1F_2\left(\{\frac{1}{2}\},\{1,\frac{3}{2}\},-\frac{\gamma_{0,j}^2}{4}\right)\right. \\ & \left.\times \mathrm{U}(\Omega_R t-\gamma_{0,j})\right] \end{aligned} \tag{5.16}$$

where $\gamma_{0,j}$ is the jth zero of the Bessel function (J_0) of the first kind of order 0 and U is the unit step function. Now we get the occupation probability of the lower level from Eqn. (5.15) as

$$P_1(t) = 1 - P_2(t). \tag{5.17}$$

Eqns. (5.15) and (5.17) are our quantum mechanical results for the occupation probabilities of the two states of the two-level system in the thermal radiation field. We have got these results published in Ref. [25]. We plot these probabilities in the figure 5.4 for the relevant values of the parameters for the $3s_{\frac{1}{2}}$ and $3p_{\frac{1}{2}}$ states of an ^{23}Na atom. We have profusely considered the temperature to be equal to 2700 K which is the usual temperature of the sodium vapour lamp and the usual temperature for the excitement of the two states. Rate of the stimulated emission

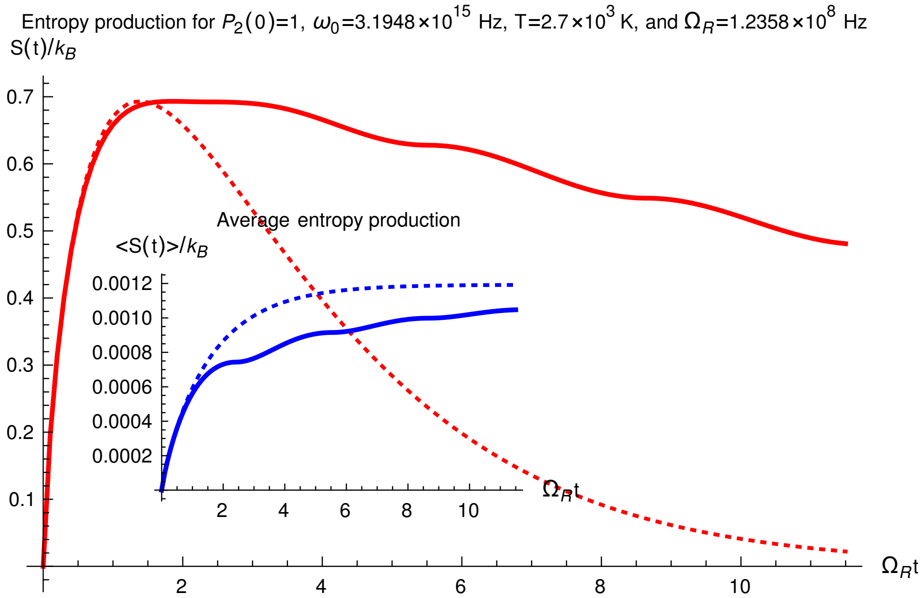

FIGURE 5.5: Entropy production for the $3s_{\frac{1}{2}}$ and $3p_{\frac{1}{2}}$ states of an ^{23}Na atom in the thermal radiation field. The plot (solid line) follows from Eqn. (5.18) for the parameters as mentioned in figure 5.4. The dotted line represents the same obtained from Einstein probabilities (Eqn. (5.14) and its complementary). The solid line in the inset represents the average entropy production for same parameters except the initial condition. The dotted line in the inset represents the same based on the Einstein probabilities.

($R(0) = 0.000059409\ \Omega_R$) is much less than that of the spontaneous emission ($A(0) = 0.499941\ \Omega_R$) at such a temperature. This causes significant deviation of the occupation probability from the Einstein probability. Amplitude of the partial oscillation having quasi-frequency Ω_R/π in the occupation probability would have increased if we had taken even a lower value of $R(0)/A(0)$ ($\ll 1$) at a lower temperature. Occupation probability, in contrary to that of the semiclassical Rabi model [23], asymptotically ($t \to \infty$) approaches the Einstein probability so as the Boltzmann probability. This is possible for low average number of thermal photons ($\bar{n}$) because all the generalized Einstein coefficients ($A(t)$, $B_{21}(t)$, and $B_{12}(t)$) vary with time in a similar fashion for low average number of thermal photons [5]. It is clear from the figure 5.4 that, the quantum Rabi oscillation slows down the occupation probability reaching the Boltzmann probability. The deviation of the occupation probability from the Einstein probability as well as the amplitude of the partial oscillation would decrease had the ratio $R(0)/A(0)$ been taken large ($\gtrsim 1$) at a higher temperature. Our result, of course, exactly matches with the Einstein probability if the Rabi flopping is completely turned off, i.e., if we take $\Omega_R \to 0$.

5.3.2 Entropy production within the multimode Jaynes-Cummings model

Though the light-matter interactions take place in short time scale ($t \sim 1/\omega_0$), dipole-transitions take place in longer time scale ($t \sim 1/\Omega_R$). Occupancy of the two levels of the system becomes probabilistic as because it is not known exactly when the two-level system makes a transition. This loss of information leads to the entropy production of the two-level system as [19, 23]

$$S(t) = -k_B[P_1(t)\ln(P_1(t)) + P_2(t)\ln(P_2(t))]. \tag{5.18}$$

We show the time-dependence of the entropy production in the figure 5.5 for the occupation probabilities (Eqns. (5.15) and (5.17)) and the Einstein probabilities (Eqn. (5.14) and its complementary) for the fixed temperature $T = 2700$ K and initial condition $P_2(0) = 1$. We already have mentioned that, the quantum Rabi oscillation slows down the occupation probability reaching the Boltzmann probability. Similar feature is also apparent in the entropy production (solid line) in the figure 5.5 where we also have plotted the entropy production (dotted line) which has been calculated based on the Einstein probabilities. The entropy of the two-level system is always less than equal to $k_B \ln(2)$ as expected. The two forms of the entropy production eventually meet the equilibrium entropy at $t \to \infty$. It is clear from the figure 5.5 that, entropy of the two-level system is not always an ever non-decreasing function of time at least for the initial condition $P_2(0) = 1$. This is, however, not an example of the violation of the second law of stochastic thermodynamics. Jarzynski equality rather allows non-increase of the entropy for some (not all) realizations of the initial conditions [26]. We also show average entropy production of two-level system in the inset of the figure 5.5 for all the realizations of the initial conditions ($P_2(0) = 1$ & $P_1(0) = 1$) with their proper statistical weights (Boltzmann probabilities). We have got these results published in Ref. [25].

It is clear from the inset of the figure 5.5 that, the average entropy productions which are calculated based on both the occupation probabilities (solid line) and the Einstein probabilities (dotted line), however, are always non-decreasing function of time at least for $R(0)/A(0) \ll 1$ as well as for low photon number fluctuation ($\triangle n = \sqrt{\bar{n}}\sqrt{\bar{n}+1} \lesssim 1$). Thus we validate the second law stochastic thermodynamics for a two-level system in the thermal radiation field for a low average number of thermal photons. Hence we can safely say that, a two-level system or a gas of two-level systems in the thermal radiation field is a practical example of a thermodynamically isolated system for $R(0)/A(0) \ll 1$. However, the two-level system in the thermal radiation field would behave like an open system for a large average number of incident photons. The second law of thermodynamic would not be applicable for such a case.

[5] See Section 3.3.2 for the same.

5.4 Conclusion

5.4.1 Conclusion for the population dynamics and entropy production within the Rabi model

Although the limit $\Omega_R \to 0$ retrieves the original B coefficient, yet the time-dependence plays a significant role in the population dynamics. The oscillations in the B coefficient, even for very small Ω_R, drives the system away from the thermodynamic equilibrium at any finite temperature. This is at odds with the Einstein's assumption about the thermodynamic equilibrium of a two-level system with the thermal radiation field [17]. The predicted equilibrium, however, can be ensured for the case $\Omega_R \to 0$, i.e., in absence of the Rabi flopping, as is expected. Nonzero finite value of the light-matter coupling ($\sim \Omega_R$) quasi-periodically drives the two-level system for the multi-frequency modes of the thermal radiation field. We have also obtained results for the same system in the monochromatic radiation field. The drive would be periodic for this case.

While the Rabi flopping usually is studied for strong light-matter interactions ($\Omega_R/A \gg 1$), Einstein's rate equations are usually applied for weak light-matter interactions ($\Omega_R/A \ll 1$). Incidentally, the Rabi model, which gives exact results in both the weak coupling regime and the strong coupling regime, is not phenomenologically different from the fundamental processes' point of view. We have been interested in bringing the Rabi flopping and the rate equation together in a single footing for this reason. We have been specially interested in the intermediate regime ($0 \lnapprox \Omega_R/A \lesssim 1$) where the partial oscillations, as shown in figure 5.1 for $\Omega_R/A = 3.17898$, are expected to be damped for the broadband excitations [27]. These partial oscillations, of course, are not periodic [6] for the nonzero width ($\triangle\omega$) of the frequency broadening around the resonance. The partial oscillations as shown in figure (5.2), however, would neither be damped nor be aperiodic for monochromatic wave, i.e., for extremely narrow band ($\triangle\omega \to 0$).

Roles of the fundamental processes (the spontaneous emission, the stimulated emission, and the stimulated absorption) in the evolution of the entropy of a system are exemplified by considering the Rabi model as a toy model for the two-level system in the thermal radiation field. We are, however, not shaking the usual notion of the thermal equilibrium between atoms (or molecules) and black body radiation and Einstein's conclusions, as they are all correct for memory-less transitions under no external drive due to the light–matter interactions. The second law of thermodynamics is not applicable for such an external drive because the two-level system is no longer a thermodynamically isolated system, rather becomes an open system, under the influence of the external drive.

Before concluding this subsection, we take this opportunity to point out that, our work opens avenue of one interesting research possibility: how to calculate entropy

[6] This aperiodicity can be linked to the non-regular intervals of the zeros of the Bessel function (J_0) in Eqn. (2.9).

productions of the laser trapped ultra-cold Bose and Fermi systems by generalizing the toy model.

5.4.2 Conclusion for the population dynamics and entropy production within the multimode Jaynes-Cummings model

We have studied the population dynamics for the two-level system by generalizing the Einstein rate equations with the generalized Einstein coefficients. The population dynamics obtained by us differs significantly from that obtained in the semi-classical theory [17, 18] for a low temperature. While the quantum Rabi oscillation is studied for strong light-matter interactions ($g_{\omega_0} \gg \sup\{\gamma, \kappa\}$[7] [28]), the Einstein rate equations are often applied for weak light-matter interactions ($g_{\omega_0} \ll \sup\{\gamma, \kappa\}$ [28]). Incidentally, the multimode J-C model gives results for both the weak coupling regime and the strong coupling regime as far as the rotating wave approximation ($2g_{\omega_0} \ll \omega_0$) is applicable. Thus we have been interested in bridging the quantum Rabi oscillation and the phenomenological rate equations by the multimode J-C model. The partial oscillations, as shown in figure (5.4), are expected to be damped for the broadband excitations in the intermediate regime [27].

Although the limit $\Omega_R \to 0$ gives all the Einstein coefficients back, yet the time-dependence of the generalized Einstein coefficients plays a significant role in the dynamics of probabilities of the two states of the system either at a low temperature or at a low average number of thermal photons. The two-level system approaches thermal equilibrium in the thermal radiation field as because the temporal parts of all the generalized Einstein coefficients are same as far as low photon number fluctuation ($\triangle n = \sqrt{\bar{n}}\sqrt{\bar{n}+1} \lessapprox 1$) is concerned [25]. The second law of (stochastic) thermodynamics is applicable in such a situation because the two-level system behaves like a thermodynamically isolated system for low average number of photons. Had the generalized A coefficient been a constant, the oscillations in the generalized B coefficient even for very small Ω_R, would have driven the system away from the thermal equilibrium at any finite temperature [25]. The temporal parts are expected to be differed for large photon number fluctuation, and the two-level system is expected to go away from thermal equilibrium. The second law of (stochastic) thermodynamics is not applicable in such a situation because the two-level system behaves like an open system for large average number of photons. Study of the generalized Einstein coefficients and population dynamics of the two-level system in the thermal radiation field having large photon number fluctuation is kept as an open problem.

Role of the fundamental processes in the time-evolution of entropy of a system are shown by considering the multimode J-C model as a toy model for the two-level system in the thermal radiation field. Time-dependence of the generalized Einstein coefficients opens a path to go beyond Pauli-von Neumann formalism of the non-equilibrium statistical mechanics [25]. The population dynamics studied by us

[7] Here γ is the non-resonant decay rate and $\kappa = \omega_0/Q$ is the photon decay rate of the cavity [28].

would be useful for studying non-perturbative quantum nonequilibrium statistical mechanics for the time-dependent Markovian process undergone on a cold gas of atoms or molecules.

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Chapter 6

Conclusions and future scope

6.1 Conclusions

Chapter-1 contains the basic elements of the works done in this thesis. Conclusions have already been mentioned at the end of the other chapters. However, brief conclusions of the Ph.D. works, especially the summaries, can be described chapter-wise as follows.

In Chapter-2, we have obtained Rabi model [1] result for the Einstein B coefficient [2]. The system of interest for this chapter is mainly a two-level system (atom/molecule) in the thermal radiation field. Starting from the Rabi Hamiltonian, which is useful in arriving at non-perturbative results within the rotating wave approximation, we have found Einstein's B coefficient to be time-dependent: $B(t) = B_0|J_0(\Omega_R t)|$ for a two-level system in thermal radiation field [2]. Here B_0 is the original Einstein B coefficient, Ω_R is the Rabi flopping (angular) frequency of the two level system, and J_0 is the zeroth order Bessel function of the first kind. Here the light-matter interaction is treated classically but the two-level system is treated quantum mechanically, and our result can be considered as a semiclassical result. We, of course, get back the original B coefficient in the limiting case of $\Omega_R \to 0$. Our result for the generalized B coefficient is fairly accurate for large Bohr frequency ($\omega_0 \gg \Omega_R$) and fairly high temperature ($k_B T \gtrsim \hbar\Omega_R$), and is significantly different from the perturbation result [3] which is not reliable near the resonance ($\omega \to \omega_0$). Our analytical result regarding the B coefficient is an invitation for the experimentalists to do direct measurement of the B coefficient. We also have obtained Rabi model result for the Einstein B coefficient for a monochromatic light incident on the two-level system [2].

In Chapter-3, we have generalized the Einstein A and B coefficients from quantum field theoretic point of view by bringing the fundamental processes and the quantum Rabi oscillation in a single footing for the light-matter interactions for nonzero Rabi frequency [4]. The generalized Einstein coefficients are found to be as $A(t) = A(0)|J_0(\Omega_R(\bar{n})t)|$ and $B_{12}(t) = B_{21}(t) = B_0|J_0(\Omega_R(\bar{n})t)|$ where $A(0)$ is the original Einstein A coefficient, B_0 is the original Einstein B coefficient, $\Omega_R(\bar{n})$ is the Rabi frequency, $\bar{n}$ is the average number of thermal photons incident on a two-level system (atom/molecule) [4], J_0 is the zeroth order Bessel function of the first kind, and t is the time. Our results for the generalized Einstein coefficients are fairly accurate for large Bohr frequency ($\omega_0 \gg \Omega_R(\bar{n})$) and low average number of thermal photons ($\bar{n} \lesssim 1$). Our results are significantly different from the results of the previous theories for the Einstein A coefficient [5] and the Einstein B coefficient [3]. We also have analytically obtained multimode Jaynes-Cummings model results for the quantum Rabi oscillations of a two-level system in a lossy resonant cavity containing (i) thermal photons and (ii) injected photons of a coherent field [4]. We have renormalized the coupling constants for the light-matter interactions for these cases. The net transition probability calculated [4] for 'vacuum' Rabi oscillation of a two-level system in a lossy Resonant cavity matches well with the seminal experimental data obtained by Brune *et al* [6]. The net transition probability calculated [4] for the quantum Rabi oscillations of the two-level system interacting with an injected coherent field in a lossy Resonant cavity also matches well with the seminal experimental data obtained by Brune *et al* [6].

In Chapter-4, we have numerically obtained theoretical results for the collapse and the revival of the quantum Rabi oscillations for low average number of coherent photons injected on a two-level system in a lossy resonant cavity [7]. We have adopted the multimode Jaynes-Cummings model for the same and especially treated the Ohmic losses from the cavity. We have compared our results with two sets of experimental data [6] for low average number of coherent photons ($\bar{n} = 0.85$ and 1.77) incident on a two-level system in the lossy resonant cavity. Our results match reasonably well with the experimental data, at least, better than the theoretical one [6, 8, 9] obtained for only the resonant mode and no loss from the cavity under consideration.

In Chapter-5, we have studied population dynamics of two-level systems interacting with both the thermal radiation field and the monochromatic light. While the interactions of the two-level systems and the monochromatic light have been treated classically (with the Rabi model) [2], the interactions of the two-level systems and the thermal radiation field have been treated both classically (with the Rabi model) [2] and quantum mechanically (with the multimode Jaynes-Cummings model) [4]. For the semiclassical cases we already have obtained the generalized (time-dependent) Einstein B coefficient in Chapter-2 [2]. For the quantum mechanical case too we already have obtained the generalized (time-dependent) Einstein A and B coefficients in Chapter-3 [4]. We have studied the population dynamics for all these cases with the help of Einstein rate equations where the original Einstein coefficients are replaced by the generalized Einstein coefficients [2, 4]. The A coefficient is, of course, kept unaltered for the semiclassical cases [2]. Time-dependence of the generalized Einstein coefficients opens a path to go beyond Pauli-von Neumann formalism of the non-equilibrium statistical mechanics [2, 4]. The population dynamics allows us to further study the entropy production of a a two-level system [2, 4]. For the semiclassical cases, we have shown that the Rabi oscillation can drive the two-level system away from the thermodynamic equilibrium [2]. On the other hand, for the quantum mechanical case, we have shown that the Rabi oscillation of a small Rabi frequency (Ω_R) can not drive the two-level system away from the thermodynamic equilibrium [4]. However, reaching the thermodynamic equilibrium is prolonged due to the quantum Rabi oscillations in the two-level system [4].

Throughout the thesis by “frequency" we have meant “angular frequency".

6.2 Future scope

The future scopes of the Ph.D. works can be described chapter-wise as follows.

In Chapter-2, we have theoretically obtained (semiclassical) Rabi model result for the Einstein B coefficient for a two-level system (atom/molecule). This B coefficient depends on time and Rabi frequency. It is a generalization of the original B coefficient. Experimentalists may verify our theory by directly measuring the Einstein B coefficient in the time scale of the time-period of the Rabi oscillation of the two-level system. This generalized Einstein B coefficient would be useful to study the population dynamics of a gas of two-level systems (atoms/molecules) at a large temperature. Such a population dynamics would be useful to study nonequilibrium statistical mechanics beyond the Pauli-von Neumann formalism [10, 11] at a large temperature.

In Chapter-3, we have theoretically obtained (quantum mechanical) multimode Jaynes-Cummings model results for the Einstein A and B coefficients for a two-level system (atom/molecule). These coefficients depend on time and Rabi frequency. These are generalizations of the original Einstein coefficients. Experimentalists may verify our theory by directly measuring the Einstein coefficients in the time scale of the time-period of the Rabi oscillation of the two-level system. These generalized Einstein coefficients would be useful to study the population dynamics of a gas of two-level systems (atoms/molecules) at a low temperature. Such a population dynamics would be useful to study nonequilibrium statistical mechanics beyond the Pauli-von Neumann formalism [10, 11] at a low temperature. All these exercises can be further extended for a three-level system by adopting the mutimode three-level Jaynes-Cummings model. We also have studied the quantum Rabi oscillations of the two-level system in a lossy resonant cavity by adopting the multimode Jaynes-Cummings model. Such a study can be further extended by adopting the multimode quantum Rabi model for the ultra-strong coupling regime and the deep-strong coupling regime [12, 13].

In Chapter-4, we have numerically obtained theoretical results for the collapse and the revival of the quantum Rabi oscillations for low average number of coherent photons injected on a two-level system (^{87}Rb atom) in a lossy resonant cavity. We have adopted the multimode Jaynes-Cummings model for the same and especially treated the Ohmic losses from the cavity. Our results match reasonably well with the experimental data [6], at least, better than the theoretical one [6, 8, 9] obtained for only the resonant mode and no loss from the cavity under consideration. However, our results can be further improved by considering the losses due to the frequency broadening of inhomogeneous light-matter coupling along the cavity axis, Doppler broadening due to the speed distribution of ^{87}Rb atoms in the cavity, thermal broadening, *etc*.

In Chapter-5, we have theoretically described the population dynamics of ideal distinguishable two-level systems (atoms/molecule) in the thermal radiation field by adopting the multimode Jaynes-Cummings model. These results are applicable for the low average number of thermal photons incident on the two-level systems. Our results can be further extended for the large number of thermal photons incident on the two-level systems. Our results can also be further extended for the laser trapped ultra-cold Bose or Fermi gas of two-level systems.

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