Modelling integrated photonic quantum technologies

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Contents

1 Abstract

The purpose of this report is to explore the light-matter interaction of single photons and two level emitters. This will be done by deriving the Fock state master equations for a two level system coupled to the electromagnetic field, before solving them to find state excitation probabilities and output flux expectations. This model is then extended to model single photon interactions with a molecule coupled to a localised phonon mode.

2 Introduction

Quantum technology is a cross-disciplinary field of applied research underpinned by advances in quantum theory. The field is rapidly growing in the areas of quantum communications, quantum simulation, quantum computing, and quantum metrology [1]. Each of these areas rely on the properties of quantum states — such as entanglement, coherence, and superposition — to gain an advantage over classical networks, computers, and sensors.

A promising platform on which to develop quantum technology, particularly for communications and information processing, is integrated photonics [2]. Analogously to the microchips produced using very large scale integrated electronics, integrated photonics allows the creation of photonic integrated circuits (PICs) containing many photonic devices on a single chip. Many materials are being studied for their suitability in integrated photonics; however, semiconductor materials are of particular interest due to existing fabrication techniques and possible integration with digital electronics [3].

A necessary component of a PIC is a waveguide which supports transmission of light around the chip. This is achieved by forming channels of contrasting refractive index. Two techniques to do so include layering materials of differing refractive index [4], or engineering the photonic band structure with periodic dielectric materials known as photonic crystals [5]. Both of these techniques can be implemented in a semiconductor PIC.

Another critical component for integrated photonic quantum technology is an on-demand single photon source [6]. Such sources have been implemented using semiconductor quantum dots [7]. Therefore, it is important that single photon interactions with such systems are understood.

Furthermore, in order to enable information processing, photon interactions must occur. However, photons at optical frequencies do not interact easily. Therefore, coupling to a solid state system is used to mediate information exchange. This can be done through linear [8] or non-linear processes [9][10].

Therefore, the aim of this report will be to explain the light-matter interaction between single photons and two level systems when confined to one dimension. This will be achieved by giving an overview of the underlying physics before moving on to create a model which will be extended to a molecular two level system.

3 Background theory

3.1 Quantisation of the electromagnetic field

Maxwell's equations for the electric field, $\mathbf{E}(\mathbf{r},t)$, and magnetic field, $\mathbf{B}(\mathbf{r},t)$, in free space can be written

$$\boldsymbol{\nabla}\cdot\mathbf{E}=\boldsymbol{\nabla}\cdot\mathbf{B}=0, \tag{1}$$

$$\boldsymbol{\nabla}\times\mathbf{E}=-\frac{\partial\mathbf{B}}{\partial t}, \tag{2}$$

$$\boldsymbol{\nabla}\times\mathbf{B}=\mu_o\epsilon_0\frac{\partial\mathbf{E}}{\partial t}, \tag{3}$$

where μ_0 and ϵ_0 are the permeability and permittivity of free space, respectively [11]. From these, one obtains the wave equations

$$\boldsymbol{\nabla}^2\mathbf{E}=\frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2}, \tag{4}$$

$$\boldsymbol{\nabla}^2\mathbf{B}=\frac{1}{c^2}\frac{\partial^2\mathbf{B}}{\partial t^2}, \tag{5}$$

where $c=\sqrt{1/(\mu_o\epsilon_0)}$ is the speed of light in a vacuum. The solutions to Equations 4 and 5 are plane waves with a phase velocity equal to the speed of light and correspond to electromagnetic radiation. From here, the simple example of a single mode will be used to illustrate the quantisation procedure before extending to the many mode case. If confined to a one-dimensional cavity in the z-direction and the electric field is polarised in the x-direction, the solutions are standing waves written as

$$E_x(z,t)=\sqrt{\frac{2\omega^2}{V\epsilon_0}}q(t)\sin(kz), \tag{6}$$

$$B_y(z,t)=\frac{1}{\omega c}\sqrt{\frac{2\omega^2}{V\epsilon_0}}\dot{q}(t)\cos(kz), \tag{7}$$

where V is the volume of the cavity mode, ω is the frequency of a mode with associated wave number, k, and $q(t)$ is a function of time with the dimension of length. The energy of the electromagnetic field is given by

$$H=\frac{1}{2}\int dV[\epsilon_0\mathbf{E}^2+\frac{1}{\mu_0}\mathbf{B}^2]. \tag{8}$$

Thus, by substituting Equations 6 and 7 into Equation 8, one obtains

$$H=\frac{1}{2}(p^2+\omega^2q^2), \tag{9}$$

where $p=\dot{q}(t)$. This is equivalent to the Hamiltonian for a one dimensional harmonic oscillator where q takes the place of canonical position and p takes the place of canonical

momentum [12]. To describe this system quantum mechanically, one can use the correspondence principle to replace p, q, and H with the operators $\hat{p}$, $\hat{q}$, and $\hat{H}$, while $\hat{p}$ and $\hat{q}$ obey the canonical momentum-position commutation relation [13]. Thus, we can write the Hamiltonian as

$$\hat{H} = \hbar\omega(\hat{a}^\dagger\hat{a} + \frac{1}{2}), \tag{10}$$

where $\hbar$ is Planck's constant and the annihilation operator has been defined as

$$\hat{a} = \frac{1}{\sqrt{2\hbar\omega}}(\omega\hat{q} + i\hat{p}) \tag{11}$$

and its Hermitian conjugate, $\hat{a}^\dagger$, is the creation operator. The eigenvalues of $\hat{H}$ are $E_n = \hbar\omega(n+1/2)$, where n are eigenvalues of the operator $\hat{n} = \hat{a}^\dagger\hat{a}$, and has corresponding eigenstates $|n\rangle$. Using the commutation relation $[\hat{a}, \hat{a}^\dagger] = 1$, and defining the ground state as $|0\rangle$, it can be shown that the actions of $\hat{a}$ and $\hat{a}^\dagger$ are [13]

$$\hat{a}\,|n\rangle = \sqrt{n}\,|n-1\rangle\,, \tag{12}$$

$$\hat{a}^\dagger\,|n\rangle = \sqrt{n+1}\,|n+1\rangle\,. \tag{13}$$

Therefore, one can see that the creation (annihilation) operator increases (decreases) the energy of the system by one quantum of energy $\hbar\omega$. As such, we can think of $|n\rangle$ as the state with n photons in the cavity. If one considers the single mode electric field of an optical cavity, then from Equation 6 and the definition of the creation operator,

$$\hat{\mathbf{E}} = \mathbf{e}\sqrt{\frac{\hbar\omega}{\epsilon_0 V}}(\hat{a} + \hat{a}^\dagger)\sin(kz) \tag{14}$$

where $\mathbf{e}$ is the unit vector of the electric field [12].

In the Heisenberg picture, operators evolve according to the Heisenberg equation of motion:

$$\frac{d\hat{O}}{dt} = \frac{1}{i\hbar}[\hat{O}, \hat{H}] \tag{15}$$

where $\hat{O}$ is the operator of interest [14]. By substituting $\hat{a}$, and the Hamiltonian in Equation 10, into the Heisenberg equation of motion, one arrives at

$$\frac{d\hat{a}}{dt} = -i\omega\hat{a}, \tag{16}$$

which has the solution $\hat{a}(t) = \hat{a}(0)e^{-i\omega t}$. Similarly, $\hat{a}^\dagger(t) = \hat{a}^\dagger(0)e^{i\omega t}$ [12].

In the above we have only considered single modes in a cavity of finite length. To calculate the field in a wave guide capable of supporting multiple modes, we must sum over all i modes such that Equation 14 becomes

$$\hat{\mathbf{E}} = \sum_i \mathbf{e}\sqrt{\frac{\hbar\omega}{\epsilon_0 V}}(\hat{a}_i + \hat{a}_i{}^\dagger)\sin(k_i z), \tag{17}$$

where $[\hat{a}_i, \hat{a}_j^\dagger] = \delta_{ij}$. Furthermore, by only considering single modes in a cavity of finite length, one imposes boundary conditions such that $k = 2m\pi/L$. This results in possible

modes being spaced by $\Delta\omega = 2\pi c/L$. As $L \to \infty$, the mode spectrum becomes continuous. The procedure for transforming to continuous mode operators is as follows [15]:

$$\hat{a_i} \to \sqrt{\Delta\omega}\hat{a}(\omega), \tag{18}$$

$$\delta_{ij} \to \Delta\omega\delta(\omega - \omega'), \tag{19}$$

$$\sum_i \to \frac{1}{\Delta\omega}\int d\omega. \tag{20}$$

Following this procedure produces

$$\hat{\mathbf{E}} = -\frac{i}{2}\mathbf{e}\int_0^\infty d\omega\sqrt{\frac{\hbar\omega}{\epsilon_0 V\Delta\omega}}(\hat{a}(\omega) + \hat{a}^\dagger(\omega))(e^{i\frac{i\omega z}{c}} - e^{-i\frac{i\omega z}{c}}), \tag{21}$$

for the electric field operator in the Schrödinger picture. In the Heisenberg picture, this procedure will produce terms in $e^{-i\omega(t-\frac{z}{c})}$ and $e^{-i\omega(t+\frac{z}{c})}$; these correspond to forward and backward propagation, respectively. Neglecting the backward propagating terms results in

$$\hat{\mathbf{E}} = i\mathbf{e}\int_0^\infty \sqrt{\frac{\hbar\omega}{4\pi\epsilon_0 cA}}\hat{a}(\omega)e^{-i\omega(t-\frac{z}{c})} + h.c., \tag{22}$$

where $A = V/L$ is the effective cross-sectional area of the wave guide mode, and $h.c.$ is the Hermitian conjugate of the previous term [15].

3.1.1 Fock states

In a wave guide which supports a continuum of frequency modes, as discussed in Section 3.1, the single photon state can be defined by

$$|1\rangle = \int d\omega\ \tilde{\xi}(\omega)b^\dagger(\omega)\,|0\rangle\,, \tag{23}$$

where the integral is over all frequencies supported by the wave guide, $b^\dagger(\omega)$ is the quantum noise operator defined in Equation 49, and $|0\rangle$ is the vacuum state. This can be be interpreted as a single photon superimposed over many frequencies weighted by the spectral density function, $\xi(\omega)$ [16]. The spectral density function must be normalised such that

$$\int d\omega|\xi(\omega)|^2 = 1. \tag{24}$$

Making the assumption that $\xi(\omega)$ is quasimonocrhomatic, such that it has a spread much smaller than the carrier frequency, the spectral density function can be described by a slowly-varying envelope, $\xi'(\omega)$, oscillating at the carrier frequency, ω_c [17]:

$$\tilde{\xi}(\omega) = \xi'(\omega)e^{i\omega_c t}. \tag{25}$$

Therefore, by defining the operator $B^\dagger(\xi)$ as

$$B^\dagger(\xi) \equiv \int dt\ \xi(t)b^\dagger(t), \tag{26}$$

Equation 23 can be written $|1\rangle = B^\dagger \xi |0\rangle$ [18]. This can then be extended to N-photon states for which [15]

$$|N\rangle = \frac{1}{\sqrt{N!}}[B^\dagger(\xi)]^N |0\rangle . \tag{27}$$

3.2 The Jaynes-Cummings model

The Jaynes-Cummings model [19] is a quantum mechanical model of the interaction between light and a two level emitter, such as an atom or quantum dot, typically used for single optical modes. The Hamiltonian of the total system is written as

$$\hat{H} = \hat{H}_{\text{emitter}} + \hat{H}_{\text{field}} + \hat{H}_{\text{int}}, \tag{28}$$

where $\hat{H}_{\text{emitter}}$ is the emitter Hamiltonian, $\hat{H}_{\text{field}}$ is the field Hamiltonian, and $\hat{H}_{\text{int}}$ is the interaction Hamiltonian.

The basis states of the emitter system are its ground state, $|g\rangle$, and its excited state, $|e\rangle$. The raising operator is $\hat{\sigma}_+ = |e\rangle\langle g|$, and takes the atom from its ground to its excited state; and, conversely, the lowering operator is $\hat{\sigma}_- = |g\rangle\langle e|$. The two states are eigenstates of the emitter Hamiltonian, but if we take the groundstate to have zero energy, then the emitter Hamiltonian can be written as

$$\hat{H}_{\text{emitter}} = \hbar\omega_0 \hat{\sigma}_+, \tag{29}$$

where the energy splitting between the two states is $\hbar\omega_0$ [20].

The Hamiltonian of the field is given by Equation 8 but if one takes the energy for $n = 0$, the zero point energy energy, to be zero then the field Hamiltonian becomes

$$\hat{H}_{\text{field}} = \hbar\omega \hat{a}^\dagger \hat{a}. \tag{30}$$

To consider the interaction Hamiltonian, the approximation is made that the coupling between the electrons and electromagnetic field is through a dipole interaction

$$\hat{H}_{\text{int}} = \hat{\mathbf{d}} \cdot \hat{\mathbf{E}}, \tag{31}$$

where $\hat{\mathbf{d}} = -e\hat{\mathbf{r}}$ is the electric dipole moment of an electron at position $\mathbf{r}$. This approximation is appropriate when the electric field is spatially uniform across the emitter, such as when the wavelength of light is much larger than the size of the emitter [21]. By inserting Equation 14, the interaction Hamiltonian becomes

$$\hat{H}_{\text{int}} = \hat{d} g(\hat{a} + \hat{a}^\dagger), \tag{32}$$

where $\hat{d} = \hat{\mathbf{d}} \cdot \mathbf{e}$ and $g = -\sqrt{\hbar\omega/\epsilon_0 V}\sin(kz)$. The expectation of $\hat{d}$ when the emitter is in the ground or excited state is zero due to the selection rule arising from $\hat{\mathbf{r}}$ only connecting states of opposite parity [14]. Therefore, $\langle g|\hat{d}|g\rangle = \langle e|\hat{d}|e\rangle = 0$. The off-diagonal elements can be written as $d|g\rangle\langle e|$ and $d^*|e\rangle\langle g|$. Hence,

$$\hat{d} = d(\hat{\sigma}_+ + \hat{\sigma}_-), \tag{33}$$

where d is real [12], and

$$\hat{H}_{\text{int}} = dg(\hat{\sigma}_+ + \hat{\sigma}_-)(\hat{a} + \hat{a}^\dagger) = dg(\hat{\sigma}_+\hat{a} + \hat{\sigma}_+\hat{a}^\dagger + \hat{\sigma}_-\hat{a}^\dagger + \hat{\sigma}_+\hat{a}^\dagger). \tag{34}$$

In the interaction picture, known as the rotating frame, $\hat{H}_{\text{int}}(t)$ evolves as

$$\begin{aligned}\hat{H}_{\text{int}}(t) = dg(&\hat{\sigma}_+(0)\hat{a}(0)e^{i(\omega_0-\omega)t} \\ &+\hat{\sigma}_+(0)\hat{a}^\dagger(0)e^{i(\omega_0+\omega)t} \\ &+\hat{\sigma}_-(0)\hat{a}(0)e^{-i(\omega_0-\omega)t} \\ &+\hat{\sigma}_-(0)\hat{a}^\dagger(0)e^{-i(\omega_0+\omega)t}).\end{aligned} \tag{35}$$

By inspecting the arguments of the exponential terms, and considering the case where $\omega_0 \approx \omega$, one notices that the terms involving $\hat{\sigma}_-\hat{a}$ and $\hat{\sigma}_+\hat{a}^\dagger$ oscillate at a much greater frequency than the other terms. These non-number conserving terms are neglected under the rotating wave approximation [12]. Therefore, moving back to the Schrödinger frame, the total Hamiltonian is approximated by

$$\hat{H} \approx \hbar\omega_0\hat{\sigma}_+ + \hbar\omega\hat{a}\hat{a}^\dagger + dg(\hat{\sigma}_+\hat{a} + \hat{\sigma}_-\hat{a}^\dagger). \tag{36}$$

For continuous modes, as discussed in Section 3.1, H_{int} in the interaction picture is given by

$$\hat{H}_{\text{int}}(t) = -i\hbar\hat{\sigma}_+ \int d\omega\ \kappa(\omega)\hat{a}(\omega)e^{-i(\omega-\omega_0)t} + h.c., \tag{37}$$

where $\kappa(\omega) = |\langle e|d|g\rangle|\sqrt{\omega/4\pi\epsilon_0\hbar cA}$ [22].

3.3 Stochastic calculus

The theoretical study of quantum optics can be considered as a subset of the the study of open systems. Open systems are systems which are coupled to an environment, such as a heat bath, which leads to the interchange of information between the system and the environment [23]. For quantum optics, this environment is the infinite number of modes in the electromagnetic field. The system's dynamics will therefore be affected by damping and fluctuations [24]. In closed systems there is no information transfer between the system and environment. In contrast, open systems require a form of calculus which accommodates stochastic processes to describe their dynamics, in order to deal with the infinite environmental degrees of freedom, which will be presented in this section.

3.3.1 Stratonovich calculus

A Stratonovich stochastic differential equation (SDE) may be written in the form

$$\dot{x} = \alpha(x) + \beta(x)\chi(t), \tag{38}$$

where $\dot{x}$ is the time derivative of x, α and β are general functions, and $\chi(t)$ is a rapidly varying continuous stochastic function of time. $\chi(t)$ can be idealised by Gaussian static noise such that its expectation is zero,

$$E[\chi(t)] = 0, \tag{39}$$

and its correlation time is zero,

$$E[\chi(t)\chi(t')] = \delta(t - t'), \tag{40}$$

where $\delta(t - t')$ is the Dirac delta function. The time derivative of a function, $f(x)$, is

$$\dot{f}(x) = f'(x)[\alpha(x) + \beta(x)\chi(t)] \tag{41}$$

where $f'(x)$ is the derivative with respect to x. Under the normal rules of calculus it is possible to assume:

1. The chain rule applies.
2. $dx = \dot{x}dt$.
3. The noise and system are independent.

However, by doing so it can be shown that the variance of $x(t + dt)$ is zero [25]. This is problematic as, from Equation 38, one expects $\chi(t)$ to introduce noise into x. Therefore, the previous assumptions are inconsistent. Stratonovich calculus asserts that assumptions (1) and (2) are true but (3) is false. Therefore, the normal rules of calculus may be used but cannot be applied to systems where (3) is true. On the other hand, Itô calculus asserts that (1) is false but (2) and (3) are true. Therefore, Itô calculus can be useful when the source of noise is independent from the system of interest. However, we must use new rules when manipulating differential equations.

3.3.2 Itô calculus

According to the usual rules of calculus, the infinitesimal increment of $f(x)$ can be written as

$$df = \frac{df}{dx}dx = \frac{df}{dt}dt. \tag{42}$$

However, in Itô calculus this rule no longer applies. Instead, to find df, one takes the Taylor expansion of $f(x)$ up to second order:

$$df = f'(x)dx + \frac{1}{2}f''(x)(dx)^2. \tag{43}$$

This is known as Itô's lemma [25]. An Itô SDE may be written in the form

$$dx = a(x)dt + b(x)dW(t), \tag{44}$$

where $dW = \chi(t)dt$ is known as the Wiener increment. The reason that one must take the expansion in Equation 43 to second order is that in Itô calculus the noise infinitesimal increments, such as dW, are not necessarily zero in the second-order as is usually the case. For the case of the Wiener increment, $(dW)^2 = dt$. This also results in a different product rule:

$$d(X(t)Y(t)) = dX(t)Y(t) + X(t)dY(t) + dX(t)dY(t), \tag{45}$$

where the third term is in addition to the usual product rule [26]. By using these rules, it can be shown that the variance of $x + dt$ is non-zero and related to $b(x)$ as expected [25]. It is often convenient to construct a SDE using the Stratonovich form and then convert it to the Itô form for solution [24]. For instance, the Itô form of a general Stratonovich SDE as shown in Equation 38 is written as

$$dx = [\alpha(x) + \frac{1}{2}\beta(x)\beta'(x)]dt + \beta(x)dW(t), \tag{46}$$

and can be generalised to a greater number of variables [27].

4 Quantum stochastic calculus

In the Schrödinger picture, states evolve via the unitary time-evolution operator, $U(t)$, which obeys the equation

$$i\hbar\frac{\partial U(t)}{\partial t} = H(t)U(t), \tag{47}$$

where H is the Hamiltonian, and the hat operator notation has been dropped [14]. To understand the evolution of a stochastic quantum system, one can use the Jaynes-Cummings model as an example and take the the Hamiltonian in Equation 37 in the quantum white-noise limit.

4.1 Quantum white noise limit

In order to take the quantum white noise limit one must first assume weak-coupling, such that $|\kappa(\omega)|^2 << \omega_0$ and $\kappa(\omega)$ is centered around ω_0 [17]. Secondly, one assumes that the correlation time of the field is short compared to the interaction time, known as taking the Markov approximation. Thus, the spectral response can be assumed to be flat: $\kappa(\omega) \approx \kappa(\omega_0) \equiv \sqrt{\gamma/2\pi}$ [28]. Therefore, by setting $\hbar = 1$, the interaction Hamiltonian can be written as

$$H_{\text{int}}(t) = i\sqrt{\gamma}(\sigma_- b^\dagger(t) - \sigma_+ b(t)), \tag{48}$$

where

$$b(t) = \frac{1}{\sqrt{2\pi}}\int d\omega\ a(\omega)e^{-i(\omega-\omega_0)t}. \tag{49}$$

The field operators $b(t)$ and $b^\dagger(t)$ have the commutation relation

$$[b(t), b^\dagger(t)] = \delta(t - t'), \tag{50}$$

when the limits in Equation 49 extend between $\pm\infty$, which can be approximated to be true when the spectrum of the coupling is flat. Therefore, the correlation time is zero, analogously to the classical white noise described in Section 3.3.1, and are known as the quantum white-noise operators. Hence, by substituting Equation 48 into Equation 51,

$$\frac{dU(t)}{dt} = \sqrt{\gamma}(\sigma_- b^\dagger(t) - \sigma_+ b(t))U(t). \tag{51}$$

However, due to the singular nature of the commutation relation in Equation 50, this differential equation is not rigorously defined [17]. Therefore, one must use the tools of stochastic calculus. This is done by defining the quantum stochastic process

$$B(t) = \int_0^t dt' b(t'), \tag{52}$$

and its Hermitian conjugate, $B^\dagger(t)$. One can then find their differential increments,

$$\int_t^{t+dt} dt' b(t') \to dB_t \quad \text{and} \quad \int_t^{t+dt} dt' b^\dagger(t') \to dB_t^\dagger, \tag{53}$$

which are the quantum analogue of the classical Wiener process in Section 3.3.1, and are called the quantum noise increments [17]. Note the subscripted t represents time dependence. Now it is possible to rewrite Equation 51 as a quantum stochastic differential equation (QSDE) in Stratonovich form:

$$dU_t = \sqrt{\gamma}(\sigma_- dB_t^\dagger - \sigma_+ dB_t) \circ U_t. \tag{54}$$

Here, $\circ$ indicates that the order of operations matters due to the operators being non-commuting. As discussed, this is because in Stratonovich calculus the noise and system are not assumed to be independent. Alternatively, one can transform Equation 54 into Itô form:

$$dU_t = (\sqrt{\gamma}\sigma_- dB_t^\dagger - \sqrt{\gamma}\sigma_+ dB_t - \frac{1}{2}\gamma\sigma_+\sigma_- dt)U_t. \tag{55}$$

Here, we no longer see $\circ$ due to the fact that we can treat dB_t and $dB_t^\dagger$ as commuting with the system operators in the integrand. This is because, in the Itô calculus, we assume the system and noise are indpendent. Equivalently, this can be thought of as the integrand and differential operator acting in indpendent time intervals [25]. The first two terms match the Stratonovich term and come from the dipole coupling to the quantum noise. The third term is the Itô correction.

Equation 55 can be extended to create a general QSDE for the time evolution operator. To do so, another quantum noise, Λ_t, which can drive the system must be introduced:

$$\int_t^{t+dt} dt' \; b^\dagger(t')b(t') \to d\Lambda_t. \tag{56}$$

Such a noise operator arises when considering an interaction in which a system operator, $\tilde{S}$, couples to the number of photons in the field through the Hamiltonian:

$$H_{\text{number}}(t) = \tilde{S}b^\dagger(t)b(t). \tag{57}$$

By also considering this interaction and the Hamiltonian of the system, H, it can be shown that

$$dU_t = (-(\frac{1}{2}L^\dagger L + iH)dt - L^\dagger S dB_t + L dB_t^\dagger + (S - I)d\Lambda_t))U_t \tag{58}$$

where L is a jump operator, such as the dipole transition operator, I is the identity operator, and S is a unitary operator which can be calculated from $\tilde{S}$ [29][30].

4.2 Itô-Langevin equations

In the Heisenberg picture, an operator O at time t is given by [14]

$$O_t = U_t^\dagger O U_t. \tag{59}$$

Thus, the differential increment of O_t is $dO_t = d(U_t^\dagger O U_t)$. Using the Itô product rule in Equation 45,

$$dO_t = d(U_t^\dagger O U_t) = dU_t^\dagger O U_t + U_t^\dagger O dU_t + dU_t^\dagger O dU_t, \tag{60}$$

where the third term contains a second order increment. As dU_t is given by Equation 58, and $dU_t^\dagger$ can be found by taking its Hermitian conjugate, one is able to construct the equation of motion for a general system operator. These equations are known as Itô-Langevin equations. However, in order to do so, one must deal with second-order quantum noise increments such as $dB_t^\dagger dB_t$. These can be handled using the set of rules listed in Table 1 [26]. The resulting equation of motion for a system operator, O, is

$$dO = (i[H, O)] + \mathcal{L}^\dagger[L]O)dt + [L^\dagger, O]S dB + S^\dagger[O, L]dB^\dagger + (S^\dagger O S - O)d\Lambda \tag{61}$$

where

$$\mathcal{L}^\dagger[L]O = L^\dagger O L - \frac{1}{2}(L^\dagger L O + O L^\dagger L) \tag{62}$$

is the adjoint of the Lindblad superoperator [17]. The adjoint is defined in such that

$$\mathrm{Tr}_{\mathrm{sys}}[O\mathcal{L}(t)\rho_{\mathrm{sys}}] = \mathrm{Tr}_{\mathrm{sys}}[(\mathcal{L}^\dagger(t)O)\rho_{\mathrm{sys}}], \tag{63}$$

where $\mathcal{L}$ is the Lindblad superoperator [31]. The number of photons in the field between time t and $t + dt$, the flux, is given by $d\Lambda$ [32]. Using the same method as above, an equation of motion for the output flux has been found to be [17]

$$d\Lambda_t^{\mathrm{out}} = L^\dagger L dt + L^\dagger S dB_t + S^\dagger L dB_t^\dagger + S^\dagger S d\Lambda_t. \tag{64}$$

4.3 Fock states and quantum noise operators

The action of the quantum noise increments, dB_t and $d\Lambda_t$, on the Fock states described in Section 3.1.1 are as follows:

$$dB_t \left|n\right\rangle = dt\sqrt{n}\xi(t) \left|n-1\right\rangle \tag{65}$$

$$d\Lambda_t \left|n\right\rangle = dB_t^\dagger \sqrt{n}\xi(t) \left|n-1\right\rangle. \tag{66}$$

These results are derived in [17] and will be useful in the following section.

$\times$	dB_t	$d\Lambda_t$	$dB_t^\dagger$	dt
dB_t	0	dB_t	dt	0
$d\Lambda_t$	0	$d\Lambda_t$	$dB_t^\dagger$	0
$dB_t^\dagger$	0	0	0	0
dt	0	0	0	0

Table 1: Itô product table for the quantum noise increments under vacuum. To find the product, take the row and multiply by the column. Adapted from [26].

5 Master equations

The following section will examine how to model the interactions of photon wave packets with a two level emitter. For notational convenience, the hat operator notation will be dropped, such that $\hat{H} \to H$ and Planck's constant will be set to $\hbar = 1$.

5.1 Vacuum master equation

To find the equation of motion for a system of interest, the master equation for the system must be derived. In a vacuum, one starts by assuming the initial total state, at time t_0, of the the system and field can be written

$$\rho(t_0) = \rho_{\text{sys}} \otimes \rho_{\text{vac}}, \tag{67}$$

which is the product state of the initial system state, ρ_{sys}, and the vacuum field field state, ρ_{field}. One then chooses the the interaction picture where operators evolve according to the Itô-Langevin equation in Equation 61. In the case of the field being in a vacuum state, one can neglect the quantum noise terms which leads to the vacuum Itô-Langevin equation:

$$dO = (i[H, O] + \mathcal{L}^\dagger[L]O)dt. \tag{68}$$

Now if one considers the system state density, $\rho_{00}(t)$, where $\rho_{00}(t_0) = \rho_{\text{sys}}$, then it will evolve as an operator in the vacuum Itô-Langevin equation. Therefore,

$$\frac{d}{dt}\rho_{0,0}(t) = -i[H, \rho_{0,0}] + \mathcal{L}[L]\rho_{0,0}, \tag{69}$$

where the Hemiticity of the density matrix has been used [33], and $\mathcal{L}[L]\rho_{0,0}$ is as defined in Equation 62. This is the master equation for the case where the field is in a vacuum state. The first term is simply the evolution of the system as if it were closed. The second term is a Lindbladian dissipator which reflects the coupling of the system to the field [17].

5.2 Fock state master equations

Now one can consider the case when the field is not in the vacuum state. When the field is prepared with N input photons, the initial state of the system and field can be written

$$\rho(t_0) = \rho_{\text{sys}} \otimes |N\rangle\langle N|, \tag{70}$$

where N is a Fock-state of the field [17]. It should be noted that this is meaningful only in the case where the wavepacket starts a long distance from the system [34]. As will be shown, the system can also couple to Fock-states $|n\rangle$ where $n = 0, 1, \ldots, N$. In the vector space of Hermitian operators, the Hilbert-Schmidt inner product is defined as

$$\langle A|B\rangle \equiv \mathrm{Tr}[A^\dagger B], \tag{71}$$

where A and B are operators [35]. Thus, it is possible to define the asymmetric expectation of a joint operator, O, as

$$\mathbb{E}_{m,n}[O] \equiv \mathrm{Tr}_{\mathrm{sys+field}}[(\rho_{\mathrm{sys}} \otimes |m\rangle\langle n|)^\dagger O]. \tag{72}$$

By tracing over only the field in Equation 72, we can define reduced density operators, $\rho_{m,n}$, by

$$\mathbb{E}_{m,n}[O] = \mathrm{Tr}_{\mathrm{sys}}[\rho_{m,n}^\dagger O]. \tag{73}$$

When $m = n$, $\rho_{m,n}$ are density matrices describing the state of the system when interacting with n-photon Fock states [17]. Off diagonal operators are not valid state matrices as they are non-Hermitian [34]; however, it will be shown that they are involved in the system dynamics. One can define another asymmetric expectation of O which only traces over the Fock states, and is therefore an operator:

$$\varpi_{m,n}[O] \equiv \mathrm{Tr}_{\mathrm{field}}[(I_{\mathrm{sys}} \otimes |m\rangle\langle n|)^\dagger O)]. \tag{74}$$

If one considers an operator acting on the system, such that $O = X \otimes I_{\mathrm{field}}$, then the time derivative is given by

$$\frac{d}{dt}\varpi_{m,n}[X] = \mathrm{Tr}_{\mathrm{field}}[(I_{\mathrm{sys}} \otimes |m\rangle\langle n|)^\dagger \frac{dX}{dt})], \tag{75}$$

where dX is given by the Itô-Langevin equation in Equation 61. It should be noted that while X is a system operator, dX is a joint operator. Therefore, the actions of the quantum white-noise operators on the Fock-states, as discussed in Section 4.3, are required. The result of this derivative is the master equation in the Heisenberg picture:

$$\begin{aligned}\frac{d}{dt}\varpi_{m,n}[X] = {} & \varpi_{m,n}(i[H, X]) + \varpi_{m,n}(\mathcal{L}[L]X) \\ & + \sqrt{m}\xi^*(t)\varpi_{m-1,n}(S^\dagger[X, L]) \\ & + \sqrt{n}\xi(t)\varpi_{m,n-1}([L^\dagger, X]S) \\ & + \sqrt{mn}|\xi(t)|^2\varpi_{m-1,n-1}(S^\dagger X S - X).\end{aligned} \tag{76}$$

To find the master equation in the Schrödinger picture, one needs to consider the time derivative of $\mathbb{E}_{m,n}(X)$, and the definition of $\varpi_{m,n(X)}$, to find

$$\frac{d}{dt}\mathbb{E}_{m,n}[X(t)] = \mathrm{Tr}_{\mathrm{sys}}\left[\rho_{\mathrm{sys}}\frac{d}{dt}\varpi_{m,n}[X(t)]\right], \tag{77}$$

which can be expanded by substituting the result in Equation 76:

$$\frac{d}{dt}\mathbb{E}_{m,n}[X(t)] = \mathrm{Tr}_{\mathrm{sys}}[\rho_{\mathrm{sys}}\mathrm{Tr}_{\mathrm{field}}[(I_{\mathrm{sys}} \otimes |m\rangle\langle n|)^\dagger i[H(t), X(t)]]] + \ldots \tag{78}$$

In order to move the time dependence from the operators to the states, one can consider the first term:

$$\mathrm{Tr}_{\mathrm{sys}}[\rho_{\mathrm{sys}}\mathrm{Tr}_{\mathrm{field}}[(I_{\mathrm{sys}}\otimes|m\rangle\langle n|)^{\dagger}i[H(t),X(t)]]] \tag{79}$$

$$= i\mathrm{Tr}_{\mathrm{sys+field}}[\rho_{\mathrm{sys}}(I_{\mathrm{sys}}\otimes|m\rangle\langle n|)^{\dagger}U(t)^{\dagger}HXU(t)] + h.c. \tag{80}$$

$$= i\mathrm{Tr}_{\mathrm{sys+field}}[U(t)\rho_{\mathrm{sys}}(I_{\mathrm{sys}}\otimes|m\rangle\langle n|)^{\dagger}U(t)^{\dagger}HX] + h.c. \tag{81}$$

$$= i\mathrm{Tr}_{\mathrm{sys}}[\rho^{\dagger}_{m,n}(t)HX] + h.c. \tag{82}$$

$$= i\mathrm{Tr}_{\mathrm{sys}}[[\rho^{\dagger}_{m,n}(t),H]X] \tag{83}$$

where the time evolution of an operator in the Heisenberg picture has been used as in Equation 59, the cycylic property of the trace has been used, $O(t_0)\equiv O$, and $\rho_{m,n}(t)$ is defined by

$$\rho_{m,n}(t) = \mathrm{Tr}_{\mathrm{field}}[U(t)(\rho_{\mathrm{sys}}(I_{\mathrm{sys}}\otimes|m\rangle\langle n|)^{\dagger}U(t)^{\dagger}]. \tag{84}$$

Therefore,

$$\mathrm{Tr}_{\mathrm{sys}}[\frac{d\rho^{\dagger}_{m,n}(t)}{dt}X] = \mathrm{Tr}_{\mathrm{sys}}[(-i[H,\rho^{\dagger}_{m,n}(t)]+\dots)X]. \tag{85}$$

By comparing terms in the trace, and following the same method for the other terms, one obtains the Fock state master equations in the Schrödinger picture

$$\begin{aligned}\frac{d}{dt}\rho_{m,n}(t) =& -i[H,\rho_{m,n}(t)] + \mathcal{L}[L]\rho_{m,n} \\ &+ \sqrt{m}\xi(t)[S\rho_{m-1,n},L^{\dagger}] \\ &+ \sqrt{n}\xi^{*}(t)[L,\rho_{m,n-1}S^{\dagger}] \\ &+ \sqrt{mn}|\xi(t)|^{2}(S\rho_{m-1,n-1}S^{\dagger}-\rho_{m-1,n-1}).\end{aligned} \tag{86}$$

From this, one can see that each of the reduced density matrices, $\rho_{m,n}$ couple to all of the other reduced density matrices of smaller m and n down to the reduced density matrix for the vacuum, $\rho_{0,0}$, which is equal to the result found in Section 5.1. Therefore, the first two terms in Equation 86 represent the system dynamics in a vacuum and the other terms represent the interaction with the wavepacket $\xi(t)$ [17].

5.3 Photon flux

The integrated mean photon number up to time t is given by $\mathbb{E}[\Lambda^{\mathrm{out}}_t]$ [17]. From Equation 64, the Itô-Langevin equation for the Λ^{out}_t, taking the expectation using Equation 72, and applying the quantum noise operators, one attains

$$\begin{aligned}\frac{d}{dt}\mathbb{E}_{m,n}[\Lambda^{\mathrm{out}}_t] =& \ \mathbb{E}_{m,n}[L^{\dagger}L] \\ &+\sqrt{m}\xi^{*}(t)\mathbb{E}_{m-1,n}[SL^{\dagger}] \\ &+\sqrt{n}\xi(t)\mathbb{E}_{m-1,n}[L^{\dagger}S] \\ &+\sqrt{mn}|\xi(t)|^{2}\mathbb{E}_{m-1,n-1}[S^{\dagger}S]\end{aligned} \tag{87}$$

By solving this equation, the integrated photon flux may be found [17].

5.4 Solving the master equations

Solving the master equations involves solving a set of coupled differential equations. Fortunately, the master equations couple downwards so the number of equations is finite and can be readily solved by numerical integration, as was done in this project. The code used in this project is available on GitHub [36]. Firstly, initial field state must be decided. The maximum number of photons in the input wavepacket, N, must be decided as this will dictate the number of reduced density matrices, $\rho_{m,n}$ required, and the number of equations to be solved. Hence, the field may be prepared in a superposition of Fock states such that

$$\rho_{\text{field}} = \sum_{m,n=0}^{N} c_{m,n} \left|n\right\rangle \left\langle m\right| . \tag{88}$$

The total system state is then

$$\rho_{\text{total}}(t) = \sum_{m,n} c^*_{m,n} \rho_{m,n}(t), \tag{89}$$

and the expectation of a system operator X is given by [17]

$$\mathbb{E}_{\text{total}}[X] = \text{Tr}_{\text{sys + field}}[\rho_{\text{total}}(t)^\dagger X] = \sum_{m,n} c^*_{m,n} \mathbb{E}_{m,n}[X]. \tag{90}$$

Additionally, the form of the temporal wavepacket, $\xi(t)$, must be chosen. In this project, a square-normalised Gaussian profile originating far from the system was selected, given by

$$\xi(t) = \left(\frac{\Omega^2}{2\pi}\right)^{1/4} e^{-\frac{\Omega^2}{2}(t-t_a)^2}, \tag{91}$$

where t_a is the time when the peak passes the emitter and must be chosen to be sufficiently large such that the wavepacket does not have a large overlap with the emitter at $t = 0$, and Ω determines the width of the wavepacket. Secondly, the system must be defined in terms of its basis states and its operators H, L, and S. From these, the master equations may be implemented in a vectorised form. Then, the diagonal density matrices of $\rho_{m,n}(t = 0)$ are set to the initial system state, $\rho_{n,n}(t = 0) = \rho_{\text{sys}}$, and off-diagonal matrices set to zero $\rho_{m,n} = 0$. Hence, the master equations may be integrated numerically to attain $\rho_{m,n}(t)$. Then, using Equations 89 and 90, the system state and operator expectations can be found.

6 Results

6.1 Scattering from a two level emitter

For the master equation of a two level emitter coupled to a dipole interaction as described in Section 3.2, one sets $H = 0$ (in the rotating frame); $L = \sqrt{\Gamma}\left|g\right\rangle\left\langle e\right|$, where Γ is the

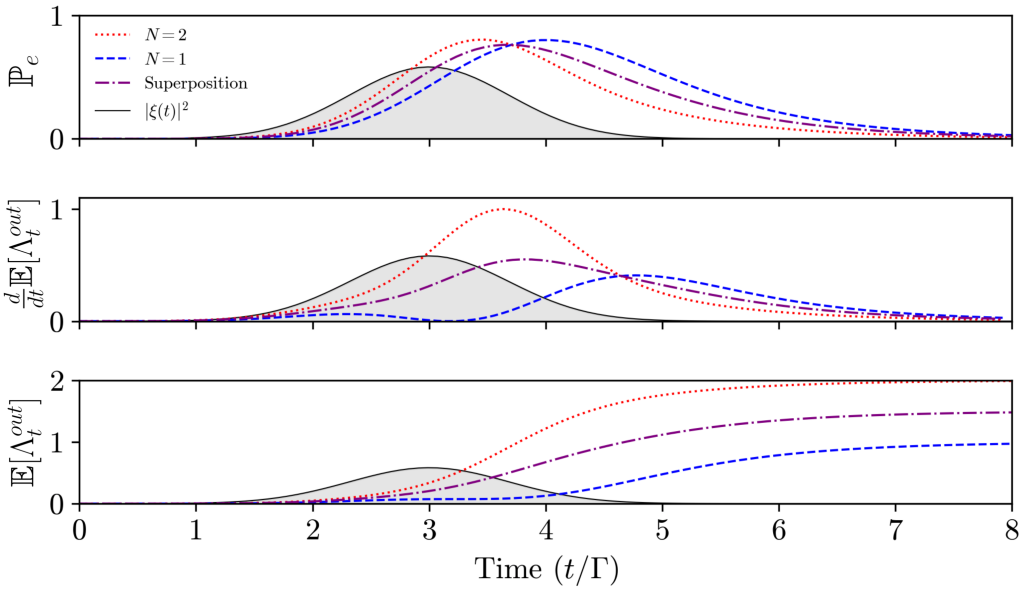

Figure 1: Interaction of a two level system with a Gaussian wave packet, $\xi(t)$, for $N = 2$ photons, $N = 1$ photon, and a superposition of one and two photons. The coupling rate was set to $\Gamma = 1$. Top: Probability of the emitter being in the excited state. Middle: Expectation of the photon flux at the emitter. Bottom: Expectation of the integrated photon flux.

coupling rate; and $S = I$ [17]. By substituting these operators into Equation 86, the Schrödinger master equations for up to two photons were found. The wavepacket, $\xi(t)$, was set as in Equation 91 with $t_a = 3/\Gamma$ and $\Omega = 1.46\Gamma$. This choice of Ω was chosen as it has been found to induce the greatest peak excitation for single photons [37]. The initial state of the emitter was set to the ground state, and the coupling rate was set by $\Gamma = 1$. Then the master equation was integrated numerically to find $\rho_{m,n}(t)$. From the solutions, the probability of the emitter being in the excited state, $\mathbb{P}_e$, was calculated using Equation 90, with X set as $|e\rangle\langle e|$. Similarly, Equation 87 was solved to find the expected photon flux, $\frac{d}{dt}\mathbb{E}_{m,n}[\Lambda_t^{\text{out}}]$, and the expected integrated photon flux, $E_{m,n}[\Lambda_t^{\text{out}}]$. This was done for the case where the field was prepared in the state with two ($N = 2$) photons: $\rho_{\text{field}} = |2\rangle\langle 2|$; one ($N = 1$) photon: $\rho_{\text{field}} = |1\rangle\langle 1|$; and a superposition: $\rho_{\text{field}} = \frac{1}{2}(|1\rangle\langle 1| + |2\rangle\langle 2|)$. These results are plotted in Figure 1.

The single photon excitation shown in Figure 1 is in agreements with the analytic solution found in [38]. One can see that the one and two photon cases result in similar excitation profiles but with distinctly different output fluxes. In particular, one can see a double peak in the single photon case. This is due to the fact that the emitter can absorb at most one photon and so much of the wavepacket is undisturbed [17] in the two photon case.

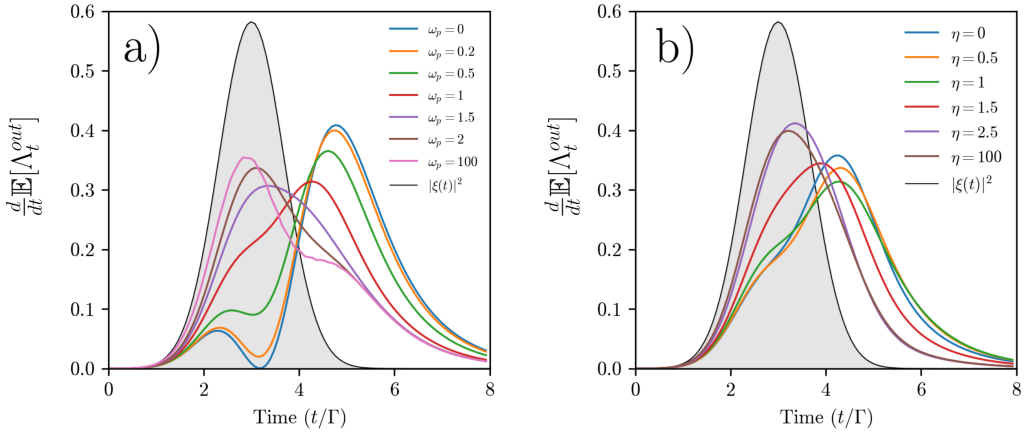

Figure 2: Interaction of a two level molecular system, including localised phonons, with a single photon Gaussian wave packet, $\xi(t)$. a) Expected photon flux is plotted for different phonon frequencies, ω_p, in units of Γ when $\eta = 1$. b) Expected photon flux is plotted for different values of η, in units of $\hbar\Gamma$, when $\omega_p = \Gamma$.

6.2 Scattering from a molecule

The master equation for a two level emitter was extended to model a molecular system with vibrational degrees of freedom. This was modelled by assuming that the two level emitter is also coupled to a bath of localised phonon modes. This may be done by treating the vibrational modes as part of the system's degree of freedom [39]. This results in the system Hamiltonian, H, becoming

$$H = \sum_{i=1}^{M} (M\omega_{p_i} a_i^\dagger a_i + \eta_i \sigma_+ \sigma (a_i^\dagger + a_i), \tag{92}$$

where there are M localised harmonic modes described by creation and annihilation operators $a_i^\dagger$ and $a_i^\dagger$, with frequency splittings ω_{p_i}, and are coupled to the two level emitter with strength η_i [39]. In the course of this project, only a single phonon mode was considered, $M = 1$, and thus H becomes

$$H = \omega_p a^\dagger a + \eta \sigma_+ \sigma_- (a_i^\dagger + a_i). \tag{93}$$

The results for the expected output photon flux during the interaction of the molecular system with a single photon Gaussian wave packet is shown in Figure 2. A range of different values for η and ω_p were investigated. By increasing the phonon frequency splitting, ω_p, one can see that the flux matches more closely the wave packet, which is correlated to the photon flux in the absence of the molecule. Additionally, the double peak seen with no phonon contribution is lost and the peaks are broadened.

Figure 2, shows the result of adjusting the ratio between η and ω_p, which is related to the Huang-Rhys parameter [40], and is therefore an indication of the fraction of light

emitted through a phonon sideband. However, this was not investigated in the course of this project.

Another key limitation of this analysis is that only a single phonon mode was considered. This may result in unrealistic results as the Hamiltonian in Equation 93 is non-number conserving and therefore coupling to higher vibrational modes was not incorporated into the master equations. Therefore, future work should look at how the solutions converge with an increasing number of available phonon modes.

7 Conclusion

In the course of this project, the underlying physics required to understand the interaction between two level systems and light was explored. Additionally, an introduction to stochastic calculus and its application to open quantum systems was made. From this, the master equations, describing the dynamics of a two level system interacting with few photon wave packets, were derived. These equations were then solved numerically, allowing the excitation of the two level system and flux to be investigated. This model was then extended to consider a molecule coupled to both the electromagnetic field and to a single local phonon mode. Although more work is required to apply this theory to realistic scenarios, the work carried out in this project lays a useful foundation for looking at how integrated quantum photonic technologies can be modelled.

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