The light matter Hamiltonian and how truncation breaks gauge invariance.

NTNU

Written by
Jan Haakon Melka Trabski

Supervised by
Tor Strømsem Haugland

2020

Abstract

The goal of the thesis is to give a fundamental understanding of how matter and light interact with each other, and a detailed derivation of the different matter light Hamiltonians. The main goal is to compare different models and see which model uses the least amount of computational time towards a good approximated solution and discuss the flaws and strengths of the different Hamiltonians and why some Hamiltonians work well compared to other models.

The result of the thesis is that one can transform Hamiltonians to different gauges, as it can be advantageous to solve different systems in certain gauges. One advantage is that a solution can be easier to obtain, as a smaller basis set is needed and as a consequence, less computational power is necessary. One key aspect is why some problems are preferable to solve in certain gauges, such as the matter-light Hamiltonian can be easily solved in the dipole gauge for high coupling terms compared to the Coulomb gauge. As the problem is not fully understood and there are three different main opinions on why invariance is not conserved in the Coulomb gauge. One opinion is that there is an asymmetric relation between the position and the momentum operator, different papers state that there is a loss in locality, or that the self energy term is not properly gauge transformed.

Contents

1 Introduction **1**

2 Quantization of light **3**

2.1 Multi Field Quantization 3

3 Hamiltonian interaction between light and matter **8**

3.1 The light matter Hamiltonian 8

3.2 Multipolar Hamiltonian 9

4 Derivation of different matter light Hamiltonian **12**

4.1 The Jaynes-Cummings Hamiltonian and Rabi Hamiltonian 12

4.2 Tavis-Cummings Hamiltonian and Dicke Hamiltonian 15

5 Discussion **16**

5.1 Minimal coupling Hamiltonian and PZW-Hamiltonian 16

5.2 The Different Hamiltonians 18

6 Conclusion **19**

7 Appendix **23**

7.1 Appendix A 23

7.2 Appendix B 24

7.3 Appendix B1 24

7.4 Appendix B2 26

7.5 Appendix C 27

1 Introduction

The interaction between matter and light is weak. However, with an ideal experimental setup, the interaction becomes significantly larger. The idea is to place an atom or a molecule inside a cavity in the presence of light being trapped between two mirrors placed with a distance, such that the distance divided by the wavelength gives either an integer or half an integer. This results in the light having a standing wave solution (See figure 1.1).[1; 2] Most theoretical studies for correlated matter light systems are performed within the long wavelength approximation. This is only valid when the molecule or atom is much smaller than the wavelength and the amplitude of the wave can be regarded as non-changing when interacting with the particle.[1; 3; 4; 5].

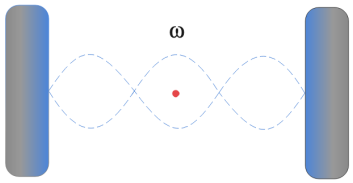

Figure 1.1: A standing wave of a single mode with frequency ω (the blue dashed line), a molecule or atom in the center (red dot) and mirrors which stand opposite of each other with a distance such that the distance gives a whole or half a integer when divided by the wavelength.

The approximation is well established within the derivation of the matter light Hamiltonian. it is assumed that the coupling term is subsumed in the kinetic energy term. A model such as Jaynes-Cummings neglects the self-interacting term, as the term contributes with constant energy.[6] However recent theoretical work highlights the importance of the self-interacting term for non-relativistic QED (quantum electrodynamics) in the long wavelength approximation. Some consequences of excluding the self-energy term are that depolarization shift is not properly described, and models lose their ground state.[6].

One important aspect of the matter light Hamiltonian is that it has one degree of freedom, chosen to be a gauge. As QED is an abelian unitary group and the gauges are invariant (the Lagrangian does not change) when transformed between local symmetry groups. it is possible to transform between gauges with a unitary transformation. [7; 8] The key idea of using such a transform, is that unitary transforms are linear and as a result, preserves the inner product of elements, which implies that the eigenvalues do not change under a unitary gauge transformation.[5; 9] A particularly gauge transformation that can transform a Coulomb gauge to a Dipole gauge is the PZW-transformation.[8]

To solve the Schrödinger equation, one can introduce projection operators. The importance of a projection operator is justified by that they can expand an arbitrary wave function in terms of a finite orthogonal basis. it is important to specify that the projection operators do not give an exact solution in terms of coefficients, as they are unknown and other approaches must be deduced to find the optimal coefficients for the arbitrary wave function, such as variational methods. A problem with truncated states is that gauge invariance breaks down, and commutation relations between operators collapse for an uncompleted basis. From a fundamental perspective, there are three hypothesis on why truncation breaks down gauge invariance and commutation relations. The first conjecture states that there is a fundamental asymmetry property between the momentum and position operator and as a result, the truncation works well when acting on the position operator, but not on the momentum operator.[10] The second conjecture states that the potential operator is not properly gauged transformed by the PZW. When the projection operator acts on the potential, the operator causes a shift in both position and momentum. A suggestion to correct the error is to let the projection operator act on the potential operator, before transforming it with the PZW-transformation.[11] The third conjecture suggests that not all operators are confined in the projection operators, this means that not all operators are truncated with respect to the projection operator. it is suggested that the self interacting term is not properly truncated with respect to the projection operator. This can be corrected by using the Thomas-Reiche-Kuhn sum rule on the self interacting term rule.[12]

One important concept that has not been mentioned, is that most models are approximated within a two-level system such as Jaynes-Cummings, Tavis-Cumming, Dicke, Rabi etc. The idea of the two-level model is that the basis only consists of two possible states, the ground state and the excited state. The reason for using such models, is because it is quite difficult to construct a proper model when regarding all the possible state simultaneously (see figure 1.2 as an illustration).[3; 13]

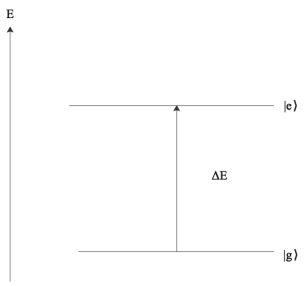

Figure 1.2: The figure represents the two-level system. The $|g\rangle$ the ground state, the $|e\rangle$ denotes the excited state and the ΔE denotes the energy difference between the two states

2 Quantization of light

To introduce the basic concepts of how matter and light interact with each other. Light must firstly be quantized with the help of Maxwell's equations. The foundation of classical electromagnetic theory states that light is an electromagnetic wave. The following chapter will contain a detailed derivation of how light can be quantized.

2.1 Multi Field Quantization

The quantum description of light requires the electromagnetic field to be quantized. First, a closer look at the Maxwell's equations is required. The equations should always be valid, but there are two different Maxwell equations depending on the system.[2; 14] For non-free space, the divergence of the electric field will be defined as the charge density (ρ) divided by the vacuum permittivity (ϵ_0). The derivation will be done in free space without any molecules. Thus, the Maxwell's equation can be written as the following equations.[2; 3; 14]

$$\nabla \cdot \mathbf{E} = 0 \tag{2.1}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2.2}$$

$$\nabla \times \mathbf{E} = -\frac{\partial B}{\partial t} \tag{2.3}$$

$$\nabla \times \mathbf{B} = \frac{1}{c^2}\frac{\partial E}{\partial t} \tag{2.4}$$

The first two equations above represent the divergence of an electric field ($\mathbf{E}$) and a magnetic field ($\mathbf{B}$) in an empty space. The operator informs if there is a singularity in space where the field origins from. The last two equations give a coherence between the magnetic and electric field. Or in other words, the curl of the field. Equations 2.1-2.4 will be the basis to construct an electromagnetic vector potential $\mathbf{A}$ in three dimensions and given those equations, it is possible to construct an electromagnetic vector potential that satisfies the equations above. [2; 3; 14; 15]

$$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t} - \nabla\phi \tag{2.5}$$

$$\mathbf{B} = \nabla \times \mathbf{A} \tag{2.6}$$

The divergence of the electric field and magnetic field gives the same result and leaves one degree of freedom left. A gauge transformation can be set for the last degree of freedom, and particularly the Coulomb gauge. which can be regarded as an conservation law, where electric field can't be created from nothing. [3] This is the whole essence of the derivation of a matter light Hamiltonian, and its because it is now achievable to transform the Hamiltonian between different families of gauges. This property is powerful, as the Hamiltonian could be difficult to solve in some gauges and easier in others. With the following established properties, it is now possible to derive the wave equation with the following divergence property from the Coulomb gauge.[16]

$$\nabla \cdot \mathbf{A} = 0 \tag{2.7}$$

combining equations 2.6, 2.5, 2.4 and 2.3 will give the the wave equation in three dimensions.

$$\nabla^2 \mathbf{A} = \frac{1}{c^2} \frac{\partial^2 \mathbf{A}}{\partial t^2} \tag{2.8}$$

There are multiple ways to find a valid vector-field that satisfies the condition from 2.8. One way is to use the frequency eigenmodes, another way is to choose proper boundary conditions.[17] In both cases the wave is inside a three-dimensional box with side length L. Because of linearity, the box can be written as a Fourier expansion of the k-modes, as the wave might be a combination of different fields. Thus, each k-solution can be considered part of a basis set, and a general solution can be represented as equation 2.9.[2; 3; 14]

$$\mathbf{A}(r,t) = \sum_{k} \sum_{\lambda=1}^{2} \mathbf{e}_{k,\lambda} \mathbf{A}_{k,\lambda}(t) e^{i\mathbf{k}\cdot\mathbf{r}} + \mathbf{e}_{k,\lambda} \mathbf{A}_{k,\lambda}(t)^* e^{-i\mathbf{k}\cdot\mathbf{r}} \tag{2.9}$$

Where k denotes the wave vector which points at the direction of propagation of the wave. It should be shown that the wave vector has the following generalized form.[2; 3; 14]

$$\mathbf{k} = \left(\frac{2\pi l}{L_x}, \frac{2\pi m}{L_y}, \frac{2\pi n}{L_z} \right) \tag{2.10}$$

with

$$l = 0, \pm 1, \pm 2, \ldots$$
$$m = 0, \pm 1, \pm 2, \ldots$$
$$n = 0, \pm 1, \pm 2, \ldots$$

Where ω_k denotes the frequency of the k^{th}-wave, $\mathbf{e}_{k,\lambda}$ is the polarization vector and the $\mathbf{A(t)}_{k,\lambda}$ is the amplitude for each Fourier mode. From the general solution it is possible to find an

expression for $\mathbf{A}(t)$ by using the assumption that $\mathbf{A}(r,t)$ is multiplicative separable and equation 2.8.[2; 3; 14]

$$\nabla^2\mathbf{A}(t)\cdot\mathbf{A}(r)-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}(r)\mathbf{A}(t)=0 \tag{2.11}$$

$$\mathbf{A}(t)\nabla^2\mathbf{A}(r)\ -\mathbf{A}(r)\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}(t)=0 \tag{2.12}$$

One fundamental postulate is it is impossible to distinguish which direction the wave propagates. Thus, the following equation must be satisfied.[2; 3; 14]

$$\mathbf{A}_k(t)=\mathbf{A}^*_{-k}(t) \tag{2.13}$$

Given a general solution for $\mathbf{A}(\mathbf{r})$. It is possible to derive a solution for $\mathbf{A}(t)$ using equations 2.12 and 2.13 .

$$\mathbf{A}(t)\nabla^2 e^{i\mathbf{k}\cdot\mathbf{r}}-e^{i\mathbf{k}\cdot\mathbf{r}}\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}(t)=0 \tag{2.14}$$

$$\left(-k^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{A}(t)=0 \tag{2.15}$$

The speed of light c can be rewritten using $c=\frac{\omega_k}{k}$, and the $\mathbf{A}(t)$ can be expressed as following.

$$\mathbf{A}(t)=A_k\cdot e^{-it\omega_k} \tag{2.16}$$

Thus, a complete solution of generality of the wave equation can be expressed as the equation below.

$$\mathbf{A}(r,t)=\sum_k\sum_{\lambda=1}^{2}\mathbf{e}_{k,\lambda}\mathbf{A}_{k,\lambda}e^{i\mathbf{k}\cdot\mathbf{r}-it\omega_k}+\mathbf{e}_{k,\lambda}\mathbf{A}^*_{k,\lambda}(t)e^{-i\mathbf{k}\cdot\mathbf{r}+it\omega_k} \tag{2.17}$$

It is now possible to express the magnetic field and the electric field as quantized waves, as there is coherence between the electromagnetic field and the two other fields.

$$\mathbf{E}(r,t)=i\sum_k\omega_k\sum_{\lambda=1}^{2}-\mathbf{e}_{k,\lambda}\mathbf{A}_{k,\lambda}e^{i\mathbf{k}\cdot\mathbf{r}-it\omega_k}-\mathbf{e}_{k,\lambda}\mathbf{A}^*_{k,\lambda}e^{-i\mathbf{k}\cdot\mathbf{r}+it\omega_k} \tag{2.18}$$

$$\mathbf{B}(r,t)=i\sum_k\sum_{\lambda=1}^{2}\frac{k\times\mathbf{e}_{k,\lambda}}{k}-\mathbf{e}_{k,\lambda}\mathbf{A}_{k,\lambda}e^{i\mathbf{k}\cdot\mathbf{r}-it\omega_k}-\mathbf{e}_{k,\lambda}\mathbf{A}^*_{k,\lambda}e^{-i\mathbf{k}\cdot\mathbf{r}+it\omega_k} \tag{2.19}$$

From classic mechanics, the total energy stored inside the cavity is given in 2.20.

$$E = \frac{1}{2}\int(\epsilon_0\mathbf{E}^2 + \mu_0\mathbf{B}^2)dV \tag{2.20}$$

By substituting the electric and magnetic field with equations 2.19 and 2.18 in equation 2.20, it is possible to solve the integral and find the total energy of the field (See Appendix A). The integrals reduces to the following expression.[3; 18; 19]

$$E = \sum_{\mathbf{k}}\sum_{\lambda}\epsilon_0 V\omega_k^2\left(\mathbf{A}_{\mathbf{k}\lambda}\mathbf{A}*_{\mathbf{k}\lambda} + \mathbf{A}*_{\mathbf{k}\lambda}\mathbf{A}_{\mathbf{k}\lambda}\right) \tag{2.21}$$

which is a general expression of the classical electromagnetic field with $\mathbf{A}_{\mathbf{k}\lambda}$ as the amplitudes of the waves. With the expression $k = 2n\pi/L$, the energy is a summation of quantified wave-modes. This is analogous to the harmonic oscillator. Thus, the amplitudes can be rewritten as a linear combination of the canonical variables $p_{\mathbf{k}\lambda}$ and $q_{\mathbf{k}\lambda}$ as in 2.22 and 2.23.[2; 14; 20]

$$\mathbf{A}_{k\lambda} = \frac{1}{2\omega_k\sqrt{(\epsilon_0 V)}}[\omega_k\hat{q}_{k\lambda} + i\hat{p}_{k\lambda}] \tag{2.22}$$

$$\mathbf{A}^*_{k\lambda} = \frac{1}{2\omega_k\sqrt{(\epsilon_0 V)}}[\omega_k\hat{q}_{k\lambda} - i\hat{p}_{k\lambda}] \tag{2.23}$$

When inserting these terms into 2.21, the following expression can be derived.

$$H = \frac{1}{2}\sum_{k}\sum_{\lambda}\left(\hat{p}^2_{\mathbf{k}\lambda} + \omega_k^2\hat{q}^2_{\mathbf{k}\lambda}\right) \tag{2.24}$$

The expression above is the harmonic oscillator, which means a proper combination of the canonical variables were given for the amplitudes, as for a single mode field, the annihilation and creation operators may be defined as

$$\hat{b}_{k\lambda} = \frac{1}{\sqrt{(2\hbar\omega_k)}}[\omega_k\hat{q}_{k\lambda} + i\hat{p}_{k\lambda}] \tag{2.25}$$

$$\hat{b}^\dagger_{k\lambda} = \frac{1}{\sqrt{(2\hbar\omega_k)}}[\omega_k\hat{q}_{k\lambda} - i\hat{p}_{k\lambda} \tag{2.26}$$

and one can observe that the amplitudes can be rewritten in the form of annihilation and creation operators.[3; 18]

$$\mathbf{A}_{k\lambda} = \frac{1}{\sqrt{(2\hbar\omega_k)}}\hat{b}_{k\lambda} \tag{2.27}$$

$$\mathbf{A}^*_{k\lambda} = \frac{1}{\sqrt{(2\hbar\omega_k)}}\hat{b}^\dagger_{k\lambda}] \tag{2.28}$$

The annihilation and a destruction operator for a specific wave-number k and polarization λ acting on a specific fock states $|n\rangle_{\mathbf{k}\lambda}$ which represents the amount of particle in a system is given below.

$$\begin{aligned}\hat{b}_{\mathbf{k}\lambda}|n\rangle_{\mathbf{k}\lambda} &= \sqrt{n}\,|n-1\rangle_{\mathbf{k}\lambda}\\ \hat{b}^\dagger_{\mathbf{k}\lambda}|n\rangle_{\mathbf{k}\lambda} &= \sqrt{n+1}\,|n+1\rangle_{\mathbf{k}\lambda}\end{aligned} \tag{2.29}$$

it is observed that the $\hat{b}^\dagger$ creates an additional photon, while $\hat{b}$ annihilates a photon. The $\hat{b}^\dagger$ is known as the creation operator while the $\hat{b}$ is known as the annihilation operator. In other literature they are typically written as $\hat{a}^\dagger$ and $\hat{a}$. For practical reasons, it is important to distinguish between boson operators and fermions operators. The difference between them is that bosons are of a integer spin such as photons, while ferions have half an integer spin such as electrons. As boson and fermion have the same operators and they commute. Thus, fermions creation and annihilation operators will be denoted with a. The $|n_{\mathbf{k}\lambda}\rangle$ is the state which represents the number of photons in a specific mode. The annihilation and creation operators obey the commutation relation given below.[3; 18]

$$[\hat{b}_{\mathbf{k}\lambda}, \hat{b}^\dagger_{\mathbf{k}\lambda}] = \delta_{\mathbf{k}\mathrm{k}'}\delta_{\lambda\lambda'} \tag{2.30}$$

with the number operator $\hat{n}$.

$$\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k'}\lambda} = \hat{n}_{\mathbf{k}\lambda} \tag{2.31}$$

Thus the Hamiltonian for the harmonic oscillator can be rewritten with respect to the annihilation and creation operators.

$$\begin{aligned}H &= \frac{1}{2}\sum_k\sum_\lambda\left(p^2_{\mathbf{k}\lambda} + \omega^2_k q^2_{\mathbf{k}\lambda}\right)\\ &= \frac{1}{2}\sum_k\sum_\lambda \hbar\omega_k(\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda} + \hat{b}_{\mathbf{k}\lambda}\hat{b}^\dagger_{\mathbf{k}\lambda})\\ &= \sum_k\sum_\lambda \hbar\omega_k(\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda} + \frac{1}{2})\end{aligned} \tag{2.32}$$

A commutator relation between the creation and annihilation operators was used to get the final Hamiltonian.[3; 18]

3 Hamiltonian interaction between light and matter

In the previous chapter, it was shown that light can be quantified from Maxwell's equations, and the annihilation and creation operators were derived. It is now appropriate to discuss the light matter Hamiltonian in the Coulomb gauge and the introduction of the minimal coupling term. As mentioned earlier, it is sometimes better to transform the Hamiltonians within the same local symmetry group, as the following chapter will include a derivation of the PZW-transformation, which transforms a Hamiltonian in the Coulomb gauge to the dipole gauge.

3.1 The light matter Hamiltonian

After quantizing the electromagnetic wave, it is now possible to describe the interaction between matter and light. The Hamiltonian is a sum of three terms, which are the atomic energy term ($\hat{H}_{\mathrm{a}}$), the electromagnetic field term ($\hat{H}_{\mathrm{c}}$) and a interaction term ($\hat{H}_{\mathrm{int}}$) as described in equation 3.5.[3; 18]

$$\hat{H} = \hat{H}_{\mathrm{c}} + \hat{H}_{\mathrm{a}} + \hat{H}_{\mathrm{int}} \tag{3.1}$$

From earlier derivations, the field Hamiltonian can be described as the equation under.

$$\hat{H}_c = \hbar \sum_k \omega_k (\hat{b}_k^\dagger \hat{b}_k + \frac{1}{2}) \tag{3.2}$$

it is possible to have multiple modes at the same time and the wave can be described as a linear combination of them and the energy will be a sum of all possibles energy states correlated to a wave mode present in the linear combination. It is typical to shift the reference point to $\hbar \sum_k \omega_k/2$ and consider only a single mode inside the cavity. Thus, equation 3.2 can be rewritten as equation 3.3.[3; 18]

$$\hat{H}_c = \hbar\omega_c (\hat{b}^\dagger \hat{b}) \tag{3.3}$$

As a brief summary, the expression above contains the number operator, which is the amount of bosons present in the system when acting on an arbitrary wave function. The ω_c is the angular frequency, which can be interpreted as the amount of periodic cycles per time. For the interaction term ($\hat{H}_{\mathrm{int}}$) and the atomic term ($\hat{H}_{\mathrm{a}}$) the Hamiltonian will be correlated. Because of the presence of an electromagnetic wave, which will affect the charged particle and shift the momentum operator as expressed in 3.4. [3; 18]

$$\hat{p}' = (\hat{p} - q\mathbf{A}(\mathbf{r}, t)) \tag{3.4}$$

it is possible to derive the expression given above by using the Lorentz force, Legendre transformations and Lagrange theory. Where $\mathbf{A}(\mathbf{r},t)$ is the electromagnetic field and q is the charge. Thus, the total Hamiltonian in the Coulomb gauge can be written as the equation below.[3; 18]

$$\hat{H} = \frac{(\hat{p} - q\mathbf{A}(\mathbf{r},t))^2}{2m} + \mathbf{V}(\mathbf{r}) + \hbar\omega(b^\dagger b) \tag{3.5}$$

The equation above is known as the minimal coupling Hamiltonian in the Coulomb gauge. The first term on the lefthand-side represents the coupling term between a charged particle and the electromagnetic wave. Thus, the total Hamiltonian is well defined and can be used to solve a system where charged particles and light are present. A problem with this Hamiltonian is it requires a large basis to get a good approximate solution up to a certain coupling value. One way to solve the problem is to use a unitary transformation to reduce the basis needed to describe the system numerically. This will not only solve the energy problem but will simultaneously save computational time for some models. Before such transformation can be considered, the dipole approximation (long wavelength approximation) must be established.[3; 18]

3.2 Multipolar Hamiltonian

It will be evident later why one wants to use the dipole approximation later, when the unitary transformation is introduced. It has been derived that the electromagnetic field has the expression given in 2.17. The expression given in 2.17, can be simplified further by inserting 2.27 and 2.28 into 2.17 and by only focusing on one of the polarizations and additionally only one single mode k. Thus, 2.17 can be further simplified to the following.[2]

$$\mathbf{A}(\mathbf{r},t) = \left(\frac{\hbar}{2\omega\epsilon_0 V}\right)^{\frac{1}{2}} \mathbf{e}(b^\dagger e^{i\mathbf{k}\cdot\mathbf{r} - it\omega_k} + be^{-i\mathbf{k}\cdot\mathbf{r} + it\omega_k}) \tag{3.6}$$

when applying the dipole approximation. The position part of the electromagnetic wave function can be written as a series of a Taylor expansion.[2; 20]

$$e^{i\mathbf{r}\cdot\mathbf{k}} = 1 + i\mathbf{r}\cdot\mathbf{k} + \frac{1}{2}(i\mathbf{r}\cdot\mathbf{k})^2 + \dots \tag{3.7}$$

It is appropriate to approximate equation 3.7 by only keeping the first term on the lefthand-side. This approximation is only valid when the wavelength is much larger than the molecule. As the amplitude doesn't change when it interferes with the molecule. Equation 3.6 can be rewritten as 3.8 .[2]

$$\mathbf{A}(t) = \left(\frac{\hbar}{2\omega\epsilon_0 V}\right)^{\frac{1}{2}} \mathbf{e}(b^\dagger e^{-it\omega_k} + be^{+it\omega_k}) \tag{3.8}$$

The equation above represents a rotating helix or in other words just a phase shift with respect to time. This means that the amplitude of the system is constant with time, and the equation 3.9 can be simplified to the following equation.[2; 20]

$$\mathbf{A} = \left(\frac{\hbar}{2\omega\epsilon_0 V}\right)^{\frac{1}{2}} \mathbf{e}(\hat{b} + \hat{b}^\dagger) \tag{3.9}$$

and let $\mathcal{A}$ denote the following

$$\mathcal{A} = \left(\frac{\hbar}{2\omega\epsilon_0 V}\right)^{\frac{1}{2}} \mathbf{e} \tag{3.10}$$

Equation 3.9 is widely used and has important applications for studying dipoles. it is should be mentioned that higher ordered terms in the dipole approximation are kept to study higher ordered poles such as quadrupoles, etc. Equation 3.9 is a key part of constructing a transformation from the Coulomb gauge to the dipole gauge. The transformation is known as the PZW-transformation (Power-Zienau-Woolley transformation). It is possible to shift the Hamiltonian, such that the energy is preserved by using the fact that Coulomb gauges are invariant when transformed between local symmetry groups. A unitary transformation is used to shift the Hamiltonian, and a special type of unitary transformation is when the length of the vector is preserved, then it is a linear unitary transformation. The argument for shifting is to reduce the basis needed to solve the Hamiltonian and in some instances, it is easier to solve. An operator $\hat{U}$ will be representing the linear unitary transformation with the following form.[2; 20; 21]

$$U = \exp(\frac{-i\hat{\mu}\mathcal{A}(\hat{b} + \hat{b}^\dagger)}{\hbar}). \tag{3.11}$$

Where $\hat{\mu}$ is the dipole moment operator can be used to check if certain transition state is allowed or not or if a molecule has a dipole moment present. By applying the unitary transformation on equation 3.5 will give the following equation to simplify.

$$\hat{U}\hat{H}\hat{U}^\dagger = \hat{U}(\hat{H}_{\text{int}} + \hat{H}_{\text{a}} + \hat{H}_{\text{c}})\hat{U}^\dagger \tag{3.12}$$

with

$$\hat{H}_{\text{a}} = \hat{p}^2 + \mathbf{V}(\mathbf{r}) \tag{3.13}$$

The Baker–Hausdorff lemma 3.14 can be used to find the unitary transformed Hamiltonian.[3; 20]

$$e^{\hat{S}^\dagger} p e^{\hat{S}} = p + i[p, \hat{S}] + \frac{(i)^2}{2}[\hat{S}, [p, \hat{S}]] + ... + \frac{(i)^n}{n!}[\hat{S}, [...[\hat{S}, [\hat{S}, [p, \hat{S}]]]] \tag{3.14}$$

Where $\hat{S}$ is a hermitian Hamiltonian. As the unitary transformation consists of boson and fermion operators, the commutator relation will not cancel out as easily. The transformation gave the following result (See appendix B for a detailed derivation) .[2; 14; 20]

$$\begin{aligned}\hat{U}\hat{H}_{\text{int}}\hat{U}^{\dagger} &= \hat{p}^2 \\ \hat{U}\hat{H}_{\text{c}}\hat{U}^{\dagger} &= \hat{b}^{\dagger}\hat{b} + i\mu\mathcal{A}(\hat{b}^{\dagger} - \hat{b}) + (\mu\mathcal{A})^2 \\ \hat{U}\hat{H}_{\text{a}}\hat{U}^{\dagger} &= \mathbf{V}(\mathbf{r})\end{aligned} \tag{3.15}$$

From equation 3.15, it is evident that the transformation acts as a momentum boost transformation. The total Hamiltonian can be rewritten with respect to the linear unitary transformation as the equation below.[2; 14; 20; 22]

$$\hat{H}_{\text{PZW}} = \hbar\omega_c(\hat{b}\hat{b^{\dagger}}) + i\omega_c\hat{\mu}\mathcal{A}(\hat{b^{\dagger}} - \hat{b}) + (\hat{\mu}\mathbf{A})^2 + \hat{p}^2 + \mathbf{V}(\mathbf{r}). \tag{3.16}$$

Form equation 3.16 the first term on the righthand-side represents the energy by the field. The second term in the interaction term between the particle and the field. The third term is the self interacting term. The two last terms are operators for the charged particle. The equation 3.16 can phase shift using the phase shift operator,

$$U(\theta) = e^{i\theta\hat{b}^{\dagger}\hat{b}} \tag{3.17}$$

and by applying the phase shift operator on 3.16, gives the following expression in 3.18. [23; 24]

$$\hat{H}_{\text{PZW}} = \hbar\omega_c(\hat{b}\hat{b^{\dagger}}) + \omega_c\hat{\mu}\mathcal{A}(\hat{b^{\dagger}} + \hat{b}) + (\hat{\mu}\mathbf{A})^2 + \hat{p}^2 + \mathbf{V}(\mathbf{r}). \tag{3.18}$$

This model serves as the basis of many models, and for some models the self interaction term is ignored, others use truncated states, such as projection operators. In the following pages, derivations of the other models will be studied, and in the end compared.[2; 14; 20]

4 Derivation of different matter light Hamiltonian

Equation 3.16 serves as the basis of many different matter light Hamiltonians. In this section, different matter light Hamiltonians will be derived. Most of the models are derived with respect to a truncated state, known as the two level model. Such models are Jaynes-Cummings model, Rabi model, Tavis-Cummings model and Dicke model.

4.1 The Jaynes-Cummings Hamiltonian and Rabi Hamiltonian

Equation 3.16 is the basis of the JC-Hamiltonian. The self interacting term is dropped.[3; 18] This is valid for this model, as the self-interacting term is constant for all possible states. For other models, there are a lot of disputes, if that is correct and the validity of the assumptions will be discussed later in the paper. The total Hamiltonian consists of the following parts.[3; 18]

$$H = \hat{H}_{\mathrm{a}} + \hat{H}_{\mathrm{c}} + \hat{H}_{\mathrm{int}} \tag{4.1}$$

The indices denotes the following: a for atom, c for cavity and int for interaction. with the following corresponding parts.[3; 18]

$$\hat{H}_{\mathrm{PZW}} = \hbar\omega_c(\hat{b}\hat{b^\dagger}) + \omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger} + \hat{b}) + (\hat{\mu}\mathbf{A})^2 + \hat{p}^2 + \mathbf{V}(\mathbf{r}). \tag{4.2}$$

The Hamiltonian above will serves as the basis to derive the Jaynes-Cummings Hamiltonian. The JC model assumes that there is only two possible states of for an atom, which is an excited state $|e\rangle$ and a ground state $|g\rangle$. This model is also known as the two level model for an atom or a molecule. This assumption is only valid if the photons have a frequency close to the transition energy. The next step is to rewrite the Hamiltonian to the known Jaynes-Cummings model. The first part can stand as it is, But the rest of the part in equation 4.2, must be rewritten with respect to the two level model. it is convenient to introduce the atomic transition operator.[3; 18]

$$\hat{\hat{\sigma}}_+ = |g\rangle\langle e|, \hat{\hat{\sigma}}_- = |e\rangle\langle g| \tag{4.3}$$

Since the basis consists of two states ($|g\rangle, \langle g|$), then the basis can be represented by a two times two matrix where each basis elements is in the column. where the diagonal element's are $\langle g|g\rangle$ and $\langle e|e\rangle$, and the off diagonal elements are $\langle g|e\rangle$ and $\langle e|g\rangle$. The atomic Hamiltonian can be represented as the equation below.[3; 18]

$$\hat{H}_{\mathrm{a}} = E_{\mathrm{e}}|g\rangle\langle g| + E_g|e\rangle\langle e| \tag{4.4}$$

E_{g} and E_e the different eigenstate of the different states. The reason for a negative sign is that the system is shifted such that the reference point is now the $1/2\hbar\omega_c$. The difference in the energy is ω_{a}.[3; 18]

$$\hat{H}_{\mathrm{a}} = \frac{1}{2}\omega_{\mathrm{a}}\hbar\hat{\sigma}_{\mathrm{z}} \tag{4.5}$$

The interaction terms must also be dealt with, as it is necessary to find the overlap matrix. Now it is necessary to calculate the expectation value of the dipole moment of the two level state. Let $\psi = e + g$.[3]

$$\langle\psi|\,\hat{d}\,|\psi\rangle = \langle e|\,\hat{d}\,|e\rangle + \langle e|\,\hat{d}\,|g\rangle + \langle g|\,\hat{d}\,|e\rangle + \langle e|\,\hat{d}\,|g\rangle \tag{4.6}$$

The off diagonal elements are nonzero for an atom, this can be explained by using symmetry arguments. When regarding the transition integrals given above, the product between two equal states results in an even function. An Addition observation is the atom can be regarded as having an inversion center, then the transition operator must be regarded as an odd function (ungerade). Thus, the product expressed by the diagonal elements, will be an odd function and the integral must be zero.[25] In contrast the off diagonal are by the same reasoning the only surviving terms. For a molecule, the same arguments cannot be used, as the molecules do not necessary need to have a inversion center and will have an active dipole moment. One way to set the diagonal elements to zero is to assume they are equal and shift the energy correspondent to the dipole moment the such that the diagonal elements becomes zero (change of reference).[3; 18]

$$\langle\psi|\,\hat{d}\,|\psi\rangle = \langle g|\,\hat{d}\,|e\rangle + \langle e|\,\hat{d}\,|g\rangle \tag{4.7}$$

$$\mathbf{g} = \langle g|\,\hat{d}\,|e\rangle = \langle e|\,\hat{d}\,|g\rangle \tag{4.8}$$

Let $\hat{d}$ represent the dipole operator and $\mathbf{g}$ as the transition operator. Combining equation 3.9 and the identity $\hat{H}_{\mathrm{int}} = \hat{\mathbf{d}}\mathbf{E}$ and rewriting $\mathbf{E}$ with respect to annihilation and creation operator, then the coupling constant g can be defined as. [3; 18]

$$g = \left(\frac{\omega}{2V\hbar\epsilon_0}\right)^{1/2}\mathbf{g}\cdot\mathbf{e} \tag{4.9}$$

Then the interaction Hamiltonian can be expressed as below.

$$\hat{H}_{\mathrm{int}} = \hbar g(\hat{\sigma}^{+} + \hat{\sigma}^{-})(b + b^{\dagger}) \tag{4.10}$$

The total Hamiltonian can be written as the following equation below.

$$\hat{H}_{\mathrm{Rabi}} = \hbar\omega_{\mathrm{c}}(\hat{b^\dagger}\hat{b}) + \hbar g(\hat{\sigma}^+ + \hat{\sigma}^-)(\hat{b} + \hat{b}^\dagger) + \frac{1}{2}\omega_{\mathrm{a}}\hbar\hat{\sigma}_{\mathrm{z}} \tag{4.11}$$

The equation 4.11 represents the Rabi Hamiltonian in the two level system. The Hamiltonian represents almost the same as the Jaynes-Cummings Hamiltonian, but the Jaynes-Cummings model is further approximated with a simplified interaction term. The interaction term can be transformed from the Schrödinger picture to the interacting picture with both a fast and a slow oscillating term. This can be done by using a unitary transform on the interacting term, as represented in the equation below. [18]

$$U' = e^{i\hat{H}_0 t} \tag{4.12}$$

with H_0 represented as the equation below

$$H_0 = b^\dagger b + \frac{\omega_a}{2}\hat{\sigma}^z \tag{4.13}$$

And as for the PZW-transformation the following commutator relations must be found by applying the Baker–Hausdorff lemma. This will give the following result.[18]

$$U'\hat{H}_{\mathrm{int}}U'^\dagger = \hbar\lambda(\hat{\sigma}^+\hat{b}e^{i(\omega_a-\omega_c)t} + \hat{\sigma}^-\hat{b}^\dagger e^{-i(\omega_a-\omega_c)t} + \hat{\sigma}^+\hat{b}^\dagger e^{i(\omega_a+\omega_c)t} + \hat{\sigma}^-\hat{b}^\dagger e^{-i(\omega_a+\omega_c)t}) \tag{4.14}$$

The four terms in this expression have a markedly different time behavior. While the first two terms vary slowly with time, $e^{\pm i(\omega_a-\omega_c)t}$, the last two terms vary rapidly with time, $e^{\pm i(\omega_a+\omega_c)t}$. The first two terms, corresponding to the absorption of a photon by the atom elevated to its excited state and the opposite process, an emission of a photon by the atom deexcited to the ground state, are therefore kept in the rotating wave approximation. This approximation is valid for $|\omega_a - \omega_c| << \omega_a$, i.e., close to resonance of the photon with the splitting of the two-level atom. With the rotating wave approximation the Jaynes-Cummings model can be derived, as represented below.[18]

$$\hat{H}_{\mathrm{JC}} = \hbar\omega_{\mathrm{c}}(\hat{b^\dagger}\hat{b}) + \hbar g(\hat{b}\hat{\sigma}_+ + \hat{b}^\dagger\hat{\sigma}_-) + \frac{1}{2}\omega_{\mathrm{a}}\hbar\hat{\sigma}_{\mathrm{z}} \tag{4.15}$$

The first term is the field energy term, the second term is the interaction term between light and matter and the last term is the atomic term.[18; 26]

4.2 Tavis-Cummings Hamiltonian and Dicke Hamiltonian

In addition to other Hamiltonians, one might want to study a system with more than one atom. Such models are the Tavis-Cummings Hamiltonian and the Dicke model. The difference in the Tavis-Cummings model is within the rotating wave approximation. The symbols are defined in section 4.1, but in addition, there are sums in the terms. The N term represents each individual atom and the interaction term between the wave and atom. Note that there is only one wave that is present in the cavity, so there is no point including combinations.[18]

$$H_{\mathrm{TC}} = \hbar\omega_{\mathrm{c}}(\hat{b^{\dagger}}\hat{b}) + \hbar\omega_{\mathrm{a}}\sum_{i}^{N}\hat{\sigma}_i^{+}\hat{\sigma}_i^{-} + \hbar\sum_{i}^{N} g_i(\hat{b}\hat{\sigma}_i^{+} + \hat{b^{\dagger}}\hat{\sigma}_i^{-}) \tag{4.16}$$

$$H_{\mathrm{Dicke}} = \hbar\omega_{\mathrm{c}}(\hat{b^{\dagger}}\hat{b}) + \hbar\omega_{\mathrm{a}}\sum_{i}^{N}\hat{\sigma}_i^{+}\hat{\sigma}_i^{-} + \hbar\sum_{i}^{N} g_i(\hat{b} + \hat{b^{\dagger}})(\hat{\sigma}_i^{+} + \hat{\sigma}_i^{-}) \tag{4.17}$$

5 Discussion

As a brief summary of the theory, four different Hamiltonians have been introduced with different applications. As QED is a unitary symmetry group, a transformation between gauges of local symmetry can be constructed. [7; 8] One such transformation is the PZW-transform 3.16, which can take a Hamiltonian in the Coulomb gauge and transform it into the dipole gauge within the long wavelength approximation.[10]. One small detail that should be mentioned is that the dipole gauge is also commonly known as the length gauge because the interacting term depends on the transition operator. In contrast, the Coulomb gauge is known as the velocity gauge, as the interacting term depends on the momentum. The reason for mentioning this is due some literature do not always use both names, even though they are synonyms.[16; 22]

Moreover, the different Hamiltonians that were derived in the dipole gauge have different applications that should be mentioned. One of the Hamiltonians derived was the Jaynes-Cummings Hamiltonian equation 4.15, which is used to study single atomic systems or single molecule and the frequencies between transition states. The Tavis-Cummings Hamiltonian equation 4.16 is used to study the same as Jayne-Cummings, but the model can be used for a many particle systems. Other models which are quite similar to the above mentioned Hamiltonians, but differ from them as they are beyond the rotating wave approximations are the Dicke-model equation 4.17 and Rabi model equation 4.11. The Rabi model is used for a single atomic systems, while the Dicke model is used for multi atomic systems.[27]

5.1 Minimal coupling Hamiltonian and PZW-Hamiltonian

As mentioned earlier the PZW-Hamiltonian is widely used regarding the light mattersystems. The main difference between the Hamiltonian is that the minimal coupling Hamiltonian given in the equation3.5 is in the Coulomb gauge while the PZW-Hamiltonian in the equation 3.16 is in the dipole gauge. [21; 28] Both models can be applied on the two-level model, where the total basis is truncated down to two possible states, the ground state and the excited state.[13] When applying a Hamiltonian on a truncated, some problems occur for the Hamiltonian in the Coulomb gauge. [10; 16; 22]

As written in the introduction, there are three hypotheses why the Hamiltonians applied on a truncated basis set in the Coulomb gauge performs poorer than the dipole gauge. One of the conjectures [10] performed a theoretical experiment, where they compared the energy in a well potential, given by the Rabi model in the dipole and Coulomb gauge. The goal was to compare the accuracy of the Rabi splittings given from the Rabi model on a truncated basis in the dipole and Coulomb gauge. The Rabi splitting calculated by the Rabi Hamiltonian in the Coulomb gauge gave less accurate Rabi splitting than the dipole gauge of the same sized truncated basis

set. This indicates that a large basis set is needed to study strong Rabi splitting values in the Coulumb gauge, compared to the same Hamiltonian in the dipole gauge. The observation was present due to an asymmetric behaviour between the momentum and position operator, which is the main reason why the Hamiltonian in the Coulomb gauge fails to produce equally good results as if the Hamiltonian was in the dipole gauge.[10] There is no suggestion from the paper on how to correct the problem, but newer research papers disagree and agree with the origin of the problem that was postulated and even suggested how to correct the problem.[4; 5; 10]

A different paper [11] did a similar experiment, but concluded more specifically why the gauge invariance broke down in the Coulomb gauge. The argument is that the potential operator in the light matter Hamiltonian is not properly gauge transformed by the PZW-transform. Meaning the locality in the potential is lost due to truncation, which is analogous to the commutation rule between the momentum and position operator is only preserved in a full basis. This means that the hamiltonian applied on truncated basis in Hilbert space, will cause a loss in locality: $\mathbf{V}(x) \rightarrow \mathbf{V'}(x, \hat{p})$ and can be fixed by applying the following substitution $\hat{p'} \rightarrow \hat{p} - q\hat{A}$ and will preserve the gauge invariance. Thus, to properly transform the Coulomb gauge with the PZW transformation, the transformation must be properly truncated by the projection operator. This means that the potential operator and the PZW operator must be acted on by the projection operator to get a gauge invariant Hamiltonian in the Coulomb gauge ($\hat{\mathbf{P}}\hat{\mathbf{U}}^{\dagger}\hat{\mathbf{P}}\cdot\hat{\mathbf{P}}\hat{\mathbf{V}}(x)\hat{\mathbf{P}}\cdot\hat{\mathbf{P}}\hat{\mathbf{U}}\hat{\mathbf{P}}$).[11]

A third paper [12] did a theoretical experiment where they transferred protons and electrons between two fixed ions to study the Rabi splitting in different gauges. They suggested that the problem comes from not all operators are properly confined when truncated, rather just loss of locality. This implies that the problem does not only apply to the potential operator, but for the dipole operator as the self energy term is not properly truncated. This was especially evident when the projection operator ($\hat{\mathbf{P}}$) acted on the PZW-transformation ($\hat{\mathbf{P}}\hat{U}\hat{\mathbf{P}}$) and then Taylor expanded $\hat{\mathbf{P}}\hat{U}\hat{\mathbf{P}} = \sum_n^{\infty} \frac{1}{n!}(\frac{-i}{\hbar})^n \hat{\mathbf{P}}\hat{\mu}^n\hat{\mathbf{P}}$) and analyzing the second order $\frac{1}{2}(\frac{-i}{\hbar})^2\hat{\mathbf{P}}\hat{\mu}(\hat{\mathbf{P}} + \hat{\mathbf{Q}})\hat{\mu}\hat{\mathbf{P}})$ where the identity is $(\hat{\mathbf{P}} + \hat{\mathbf{Q}})$, where $\hat{\mathbf{Q}}$ is higher order excited states. [11]. A suggestion to regain proper gauge invariance within the Coulomb gauge is to properly truncate the self energy term with the Thomas-Reich-Kuhn sum rule (Given in appendix C).[5]

In a brief summary, one of the main reason for not using the light matter Hamiltonian in the Coulomb gauge is that the gauge invariance breaks down and cannot accurately provide a correct Rabi splitting even for small couplings.[5] The second reason is that there is an asymmetry relation between the momentum and position operator in the Coulomb gauge. As a consequence, a larger basis set for a truncated basis applied on a Hamiltonian in the Coulomb gauge is required for the Hamiltonian to produce realistic results for even small couplings, in contrast to the dipole gauge.[10] There is some dispute about how to correct the problem, but both papers [11; 12] have proven that their way to correct the problem is functional, and result in a better description for a

higher coupling term when applying these corrections. One specification is that these corrections do not bypass the accuracy of the Hamiltonian in the dipole gauge. Resulting in a requirement of a larger basis in the Coulomb gauge to get an equally accurate Rabi splitting as in the dipole gauge.

5.2 The Different Hamiltonians

Now that the differences and problems of the Coulomb gauge and dipole gauge are established, it is time to discuss why there are so many models and the differences between them. For a single atomic system, there are two well known models, the Dicke model represented in equation 4.17 and the Jaynes Cumming model represented in equation 4.15. The main difference is that the Jaynes-Cummings model is within the rotating wave approximation. The Rabi model given in equation 4.11 can be transformed from the Schödinger picture to the interaction picture and will consist of four terms (See equation 4.14). The first two terms in equation 4.14 are slow oscillating terms with respect to time, while the last two terms are fast oscillating terms with respect to time.[18] When applying the wave approximation, the slow oscillating terms are neglected. This is a valid approximation when the applied electromagnetic radiation is near resonant with an atomic transition, and the intensity is low.[29] The reason for applying the approximation is that this simplifies the problem. For those instances the rotating wave approximation cannot be used, the function that needs to be solved is transcendental, which means that the equations can't be solved analytically and numerical approaches must be used. In some instances, the transcendental function can be expanded to a power series and an inversion of the function can be used to obtain an analytical solution.[30]

A specific case where the Rabi and the Jaynes-Cummings models are utilized is in the study of qubits.[27; 31] Within an area of circuit QED, where one wants to study single qubits and how the two level system interacts with the harmonic oscillator. In other words, study how nuclear spin interacts with a magnetic field. The model that is used is the Rabi Hamiltonian, but sometimes the wave function applied on that Rabi Hamiltonian can be a bit problematic to solve, but if the Rabi splittings between the qubits and the oscillator are much smaller than the qubit and oscillator frequencies, then a good approximation to drop the counter rotating terms (rotating wave approximation).[27] A different interest is to study larger systems with multiple atoms or a molecule, then the Tavis-Cummings or Dicke-model can be used to describe such systems. An Example where the Tavis-Cummings Hamiltonian is used is the study of quantum gates. [18; 32]

6 Conclusion

The Hamiltonian in the Coulomb gauge performs poorer than in the dipole gauge when studying coupling strength correlated to energy. The were mainly three opinions on why invariance breaks down in the Coulomb gauge [10; 11; 12], and only two of the papers provided a correction to the problem [11; 12]. The three postulated problems are given as follows: There is an asymmetric property between momentum and position operators. The second postulate is that there is a loss in locality when applying truncation on the potential in the Coulomb Gauge. which can be corrected by truncating the PZW transformation and applying the transformation on the minimal coupling Hamiltonian. The third postulate is that the operators are not properly truncated in the Coulomb gauge, especially the self interacting term. Which can be corrected if the self interacting term is properly gauge transformed.

References

[1] Raphael F Ribeiro, Luis A Martínez-Martínez, Matthew Du, Jorge Campos-Gonzalez-Angulo, and Joel Yuen-Zhou. Polariton chemistry: controlling molecular dynamics with optical cavities. *Chemical science*, 9(30):6325–6339, 2018.

[2] Yanhua Shih. *An introduction to quantum optics: photon and biphoton physics*. CRC press, 2020.

[3] Christopher Gerry, Peter Knight, and Peter L Knight. *Introductory quantum optics*. Cambridge university press, 2005.

[4] Anton Frisk Kockum, Adam Miranowicz, Simone De Liberato, Salvatore Savasta, and Franco Nori. Ultrastrong coupling between light and matter. *Nature Reviews Physics*, 1(1):19–40, 2019.

[5] Arkajit Mandal, Sebastian Montillo Vega, and Pengfei Huo. Polarized fock states and the dynamical casimir effect in molecular cavity quantum electrodynamics. *The Journal of Physical Chemistry Letters*, 11(21):9215–9223, 2020.

[6] Vasil Rokaj, Davis M Welakuh, Michael Ruggenthaler, and Angel Rubio. Light–matter interaction in the long-wavelength limit: no ground-state without dipole self-energy. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 51(3):034005, 2018.

[7] Makoto Morinaga. Deriving the static interaction between electric dipoles via the quantum gauge transformation. *arXiv preprint arXiv:1302.0920*, 2013.

[8] Guillermo Federico Quinteiro, DE Reiter, and T Kuhn. Formulation of the twisted-light–matter interaction at the phase singularity: The twisted-light gauge. *Physical Review A*, 91(3):033808, 2015.

[9] Werner H Greub. *Linear algebra*, volume 23. Springer Science & Business Media, 2012.

[10] Daniele De Bernardis, Philipp Pilar, Tuomas Jaako, Simone De Liberato, and Peter Rabl. *Breakdown of gauge invariance in ultrastrong-coupling cavity QED*, volume 98. APS, 2018.

[11] Omar Di Stefano, Alessio Settineri, Vincenzo Macrì, Luigi Garziano, Roberto Stassi, Salvatore Savasta, and Franco Nori. *Nature Physics*, 15(8):803–808, 2019.

[12] Michael AD Taylor, Arkajit Mandal, Wanghuai Zhou, and Pengfei Huo. Resolution of gauge ambiguities in molecular cavity quantum electrodynamics. *Physical Review Letters*, 125(12):123602, 2020.

[13] Leslie Allen and Joseph H Eberly. *Optical resonance and two-level atoms*, volume 28. Courier Corporation, 1987.

[14] Daniel F Walls and Gerard J Milburn. *Quantum optics*. Springer Science & Business Media, 2007.

[15] P.C.Matthhews. *Vector Calculus*, volume 1. Springer, 1998.

[16] José A Heras. How the potentials in different gauges yield the same retarded electric and magnetic fields. *American Journal of Physics*, 75(2):176–183, 2007.

[17] Rajan Singh, Mukesh Chandra, and BK Singh. Solutasdasion of 3-dimensional wave equation by method of separation of variables. *International Journal of Current Research and Review*, 7(14):54, 2015.

[18] Hans-Peter Eckle. *Models of Quantum Matter: A First Course on Integrability and the Bethe Ansatz*. Oxford University Press, USA, 2019.

[19] Tor Strømsem Haugland. Modeling chemistry in qed cavities using coupled cluster. 2019.

[20] David Parker Craig and Thiru Thirunamachandran. *Molecular quantum electrodynamics: an introduction to radiation-molecule interactions*. Courier Corporation, 1998.

[21] R Guy Woolley. Power-Zienau-Woolley representations of nonrelativistic qed for atoms and molecules. *Physical Review Research*, 2(1):013206, 2020.

[22] Morten Førre and Aleksander Skjerlie Simonsen. Generalized velocity-gauge form of the light-matter interaction hamiltonian beyond the dipole approximation. *Physical Review A*, 93(1):013423, 2016.

[23] Ulf Leonhardt. *Measuring the quantum state of light*, volume 22. Cambridge university press, 1997.

[24] Shen-Ping Bu, Guo-Feng Zhang, Jia Liu, and Zi-Yu Chen. Berry phase in a two-atom jaynes–cummings model with kerr medium. *Physica Scripta*, 78(6):065008, 2008.

[25] P.W Atkins. Atkins' physical chemistry. pages 303–326, 235–290, 2018.

[26] Fabio D Bonani. The Jaynes-Cummings model. $A\cdot$ *A*, 1(e2):2mec.

[27] S Agarwal, SM Hashemi Rafsanjani, and JH Eberly. Tavis-cummings model beyond the rotating wave approximation: Quasidegenerate qubits. *Physical Review A*, 85(4):043815, 2012.

[28] David L Andrews, Garth A Jones, A Salam, and R Guy Woolley. Perspective: Quantum hamiltonians for optical interactions. *The Journal of chemical physics*, 148(4):040901, 2018.

[29] Ying Wu and Xiaoxue Yang. Strong-coupling theory of periodically driven two-level systems. *Physical review letters*, 98(1):013601, 2007.

[30] Yimin Wang and Jing Yan Haw. Bridging the gap between the jaynes–cummings and Rabi models using an intermediate rotating wave approximation. *Physics Letters A*, 379(8):779–786, 2015.

[31] Lixian Yu, Shiqun Zhu, Qifeng Liang, Gang Chen, and Suotang Jia. Analytical solutions for the Rabi model. *Physical Review A*, 86(1):015803, 2012.

[32] Raffael Düll, Alexei Kulagin, Wanshun Lee, Yuri Ozhigov, Miao Huei-huei, and Zheng Keli. Quality of control in the Tavis-Cummings-Hubbard Model. *arXiv preprint arXiv:2103.04987*, 2021.

[33] Claude Cohen-Tannoudji, Jacques Dupont-Roc, and Gilbert Grynberg. *Atom-photon interactions: basic processes and applications*. 1998.

[34] Claude Cohen-Tannoudji, Jacques Dupont-Roc, and Gilbert Grynberg. *Photons and Atoms-Introduction to Quantum Electrodynamics*. 1997.

7 Appendix

7.1 Appendix A

The derivation for the total energy given in 2.21 will be derived in this part of the appendix. The derivation can be done by substituting the electric-field and magnetic-field with equation 2.18 and 2.19 gives the following expression

$$\begin{aligned} E = \frac{1}{2}\sum_{k}\sum_{\lambda}\int dV(\epsilon_0\omega_k\omega_{k'})(-\mathbf{A}_{k,\lambda}\mathbf{A}_{k',\lambda'}\mathbf{e}_{k,\lambda}\cdot\mathbf{e}_{k',\lambda'}e^{i(\mathbf{k}+\mathbf{k'})\mathbf{r}-(\omega_k+\omega'_k)} \\ -\mathbf{A}^*_{k,\lambda}\mathbf{A}^*_{k',\lambda'}\mathbf{e}_{k,\lambda}\cdot\mathbf{e}_{k',\lambda'}e^{i(\mathbf{k}+\mathbf{k'})\mathbf{r}-(\omega_k+\omega'_k)} + \mathbf{A}_{k,\lambda}\mathbf{A}^*_{k',\lambda'}\mathbf{e}_{k,\lambda}\cdot\mathbf{e}_{k',\lambda'}e^{i(\mathbf{k}+\mathbf{k'})\mathbf{r}} \\ \mathbf{A}^*_{k,\lambda}\mathbf{A}_{k',\lambda'}\mathbf{e}_{k,\lambda}\cdot\mathbf{e}_{k',\lambda'}e^{i(\mathbf{k}-\mathbf{k'})}) + \mu_0\mathbf{B}^2 \end{aligned} \tag{7.1}$$

The following relation must be used to further simplify the integral.

$$\int_V e^{\pm i(\mathbf{k}-\mathbf{k'})\mathbf{r}}dV = \delta_{k,\pm k}V \tag{7.2}$$

The magnetic part is analogues to the electric part. Using the equation 7.2. The integral can be solved, giving the following expression.[3; 27; 33]

$$\begin{aligned} E = \frac{1}{2}\sum_{\mathbf{k}}\sum_{\lambda,\lambda'=1,2} V\left((\mathbf{A}_{\mathbf{k}\lambda}\mathbf{A}^*_{\mathbf{k}\lambda'} + \mathbf{A}^*_{\mathbf{k}\lambda}\mathbf{A}_{\mathbf{k}\lambda'}\right) \\ \times\left(\epsilon_0\omega_k^2\mathbf{e}_{\mathbf{k}\lambda}\cdot\mathbf{e}_{\mathbf{k}\lambda'} + \frac{1}{\mu_0}\mathbf{k}\times\mathbf{e}_{\mathbf{k}\lambda}\cdot\mathbf{k}\times\mathbf{e}_{\mathbf{k}\lambda'}\right) \\ -\left(\mathbf{A}_{\mathbf{k}\lambda}\mathbf{A}_{-\mathbf{k}\lambda'}e^{-2i\omega_k t} + \mathbf{A}^*_{\mathbf{k}\lambda}\mathbf{A}^*_{-\mathbf{k}\lambda'}e^{2i\omega_k t}\right) \\ \times\left(\epsilon_0\omega_k^2 e_{\mathbf{k}\lambda}\cdot e_{-\mathbf{k}\lambda'} - \frac{1}{\mu_0}\mathbf{k}\times e_{\mathbf{k}\lambda}\cdot\mathbf{k}\times e_{-\mathbf{k}\lambda'}\right) \end{aligned} \tag{7.3}$$

The expression above can be further simplified by using the following identities.[3; 27; 33]

$$\begin{aligned} (\mathbf{k}\times e_{\mathbf{k}\lambda})\cdot(\mathbf{k}\times e_{\pm\mathbf{k}\lambda'}) = k^2 e_{\mathbf{k}\lambda}\cdot e_{\pm\mathbf{k}\lambda} \\ \mathbf{e}_{\mathbf{k}\lambda}\cdot\mathbf{e}_{\mathbf{k}\lambda'} = \delta_{\lambda,\lambda'} \end{aligned} \tag{7.4}$$

Rewriting this with some algebra gives the final expression for the energy

$$E = \sum_{\mathbf{k}}\sum_{\lambda}\epsilon_0 V\omega_k^2\left(\mathbf{A}_{\mathbf{k}\lambda}\mathbf{A}*_{\mathbf{k}\lambda} + \mathbf{A}*_{\mathbf{k}\lambda}\mathbf{A}_{\mathbf{k}\lambda}\right) \tag{7.5}$$

7.2 Appendix B

This part of the appendix will cover a detailed derivation of equation under

$$\hat{U}(\hat{H}_{\text{int}} + \hat{H}_{\text{a}} + \hat{H}_{\text{c}})\hat{U}^{\dagger} = \hbar\omega_c(\hat{b}\hat{b^{\dagger}}) + i\omega_c\hat{\mu}\mathcal{A}(\hat{b^{\dagger}} - \hat{b}) + (\hat{\mu}\mathcal{A})^2 + \hat{H}_{\text{a}}. \tag{7.6}$$

As stated in section 2.1, the Baker-Hausdorff lemma must be used to find the following commutator relations.

$$\begin{aligned}
&\hat{U}\hat{H}_{\text{int}}\hat{U}^{\dagger} = \hat{P}^2 \\
&\hat{U}\hat{H}_{\text{c}}\hat{U}^{\dagger} = \hat{b}^{\dagger}\hat{b} + \hat{\mu}\mathcal{A}(\hat{b}^{\dagger} - \hat{b}) + (\hat{\mu}\mathcal{A})^2 \\
&\hat{U}\hat{H}_{\text{a}}\hat{U}^{\dagger} = \mathbf{V}(\mathbf{r})
\end{aligned} \tag{7.7}$$

7.3 Appendix B1

The derivations will be done in a chronological order starting from above in 7.7. The relations given in 7.7 can be found by using Baker Hausdorff lemma. For the first expression, it is only necessary to include the first order commutator relation, as the rest will be zero. Before the first relation can be evaluated, a little trick must be applied, as the interaction Hamiltonian is squared. [3]

$$\hat{U}\hat{H}_{\text{int}}\hat{U}^{\dagger} = \hat{U}(\hat{p} - q\hat{A})^2\hat{U}^{\dagger}$$

The identity is inserted $1 = \hat{U}\hat{U}^{\dagger}$

$$\hat{U}(\hat{p} - q\hat{A})^2\hat{U}^{\dagger} = \hat{U}(\hat{p} - q\hat{A})\hat{U}^{\dagger}\hat{U}(\hat{p} - q\hat{A})\hat{U}^{\dagger}$$

Thus, the problem has been simplified and the product of each $\hat{U}(\hat{p} - q\hat{A})\hat{U}^{\dagger}$ will be equal and one have to only evaluate one of the terms.[3]

$$\hat{U}\hat{H}_{\text{int}}\hat{U}^{\dagger} = (\hat{p} - q\hat{A}) + [(\hat{p} - q\hat{A}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b} + \hat{b^{\dagger}})]$$

$$\frac{-i\mathcal{A}}{\hbar}[(\hat{p} - \hat{A}), \hat{\mu}(\hat{b} + \hat{b^{\dagger}})] = \frac{-i\mathcal{A}}{\hbar}[(\hat{p}, \hat{\mu}(\hat{b} + \hat{b^{\dagger}})] + \frac{-i\mathcal{A}}{\hbar}[-\hat{A}, \hat{\mu}(\hat{b} + \hat{b^{\dagger}})]$$

Now the equation above can be split up to two parts, one for the commutator relations between $\hat{U}\hat{p}\hat{U}^{\dagger}$ and the commutator relations between $\hat{U}\hat{q}A\hat{U}^{\dagger}$.[3]

$$\frac{-i\mathcal{A}}{\hbar}[(\hat{p},\hat{\mu}(\hat{b}+\hat{b^\dagger})] = \frac{-i\mathcal{A}}{\hbar}[\hat{p},\hat{\mu}\hat{b}] + [\hat{p},\hat{\mu}\hat{b^\dagger}]$$
$$\frac{-i\mathcal{A}}{\hbar}[-\hat{A},\hat{\mu}(\hat{b}+\hat{b^\dagger})] = \frac{-i\mathcal{A}}{\hbar}[-\hat{A},\hat{\mu}\hat{b^\dagger}] + [-\hat{A},\hat{\mu}\hat{b}]$$

$$\frac{-i\mathcal{A}}{\hbar}([\hat{p},\hat{\mu}\hat{b}] - [\hat{p},\hat{\mu}\hat{b^\dagger}]) = \frac{-i\mathcal{A}}{\hbar}([\hat{p},\hat{\mu}]\hat{b} + [\hat{p},\hat{b}]\hat{\mu} + [\hat{p},\hat{\mu}]\hat{b^\dagger} + [\hat{p},\hat{b^\dagger}]\hat{\mu})$$
$$\frac{-i\mathcal{A}}{\hbar}([-\hat{A},\hat{\mu}\hat{b^\dagger}] + [-\hat{A},\hat{\mu}\hat{b}]) = \frac{-i\mathcal{A}}{\hbar}([-\hat{A},\hat{\mu}]\hat{b^\dagger} + [-\hat{A},\hat{b^\dagger}]\hat{\mu} + [-\hat{A},\hat{\mu}]\hat{b} + [-\hat{A},\hat{b}]\hat{\mu})$$

$$\frac{-i\mathcal{A}}{\hbar}([\hat{p},\hat{\mu}]\hat{b} + [\hat{p},\hat{b}]\hat{\mu} + [\hat{p},\hat{\mu}]\hat{b^\dagger} + [\hat{p},\hat{b^\dagger}]\hat{\mu}) = \frac{-i\mathcal{A}}{\hbar}[\hat{p},\hat{\mu}]\hat{b} + [\hat{p},\hat{\mu}]\hat{b^\dagger})$$
$$\frac{-i\mathcal{A}}{\hbar}[-\hat{A},\hat{\mu}]\hat{b^\dagger} + [-\hat{A},\hat{b^\dagger}]\hat{\mu} + [-\hat{A},\hat{\mu}]\hat{b} + [-\hat{A},\frac{-i\mathcal{A}}{\hbar}(\hat{b}]\hat{\mu}) = [-\hat{A},\hat{b^\dagger}]\hat{\mu} + [-\hat{A},\hat{b}]\hat{\mu})$$

$$\frac{-i\mathcal{A}}{\hbar}([\hat{p},\hat{\mu}]\hat{b} + [\hat{p},\hat{b}]\hat{\mu} + [\hat{p},\hat{\mu}]\hat{b^\dagger} + [\hat{p},\hat{b^\dagger}]\hat{\mu}) = \frac{-i\mathcal{A}}{\hbar}([\hat{p},\hat{\mu}]\hat{b} + [\hat{p},\hat{\mu}]\hat{b^\dagger})$$
$$\frac{-i\mathcal{A}}{\hbar}([-\hat{A},\hat{\mu}]\hat{b^\dagger} + [-\hat{A},\hat{b^\dagger}]\hat{\mu} + [-\hat{A},\hat{\mu}]\hat{b} + [-\hat{A},(\hat{b}]\hat{\mu}) = \frac{-i\mathcal{A}}{\hbar}([-\hat{A},\hat{b^\dagger}]\hat{\mu} + [-\hat{A},\hat{b}]\hat{\mu})$$

$$\frac{-i\mathcal{A}}{\hbar}([\hat{p},\hat{\mu}]\hat{b} + [\hat{p},\hat{\mu}]\hat{b^\dagger}) = q\mathcal{A}(\hat{b}+\hat{b^\dagger})$$
$$\frac{-i\mathcal{A}}{\hbar}([-\hat{A},\hat{b^\dagger}]\hat{\mu} + [-\hat{A},\hat{b}]\hat{\mu}) = 0$$

Thus, the equation resolve to.[22]

$$\hat{U}\hat{H}_{\text{int}}\hat{U}^\dagger = \hat{P}^2$$

7.4 Appendix B2

Now the field energy Hamiltonian $\hat{H}_c$ will be evaluated. The same lemma will be used, but the commutator relation up to the second order must be evaluated, as the rest is zero.

$$\hat{U}\hat{H}_c\hat{U}^\dagger = i\omega_c\hat{b}^\dagger\hat{b} + [\hat{b}^\dagger\hat{b}, \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b}+\hat{b^\dagger})] + [[\hat{b}^\dagger\hat{b}, \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b}+\hat{b^\dagger})], \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b}+\hat{b^\dagger})]$$

$$[\hat{b^\dagger}\hat{b}, \hat{\mu}\mathcal{A}(\hat{b}+\hat{b^\dagger})] = [\hat{b^\dagger}\hat{b}, \hat{\mu}\mathcal{A}\hat{b}] + [\hat{b}\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b^\dagger})]$$

$$[\hat{b^\dagger}\hat{b}, \hat{\mu}\mathcal{A}\hat{b}] + [\hat{b^\dagger}\hat{b}, \hat{\mu}\mathcal{A}\hat{b^\dagger})] = [\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b}]\hat{b} + \hat{b^\dagger}[\hat{b}, \hat{\mu}\mathcal{A}\hat{b}] + [\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b^\dagger})]\hat{b} + \hat{b^\dagger}[\hat{b}, \hat{\mu}\mathcal{A}\hat{b^\dagger})]$$

$$[\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b}]\hat{b} + \hat{b^\dagger}[\hat{b}, \hat{\mu}\mathcal{A}\hat{b}] + [\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b^\dagger})]\hat{b} + \hat{b^\dagger}[\hat{b}, \hat{\mu}\mathcal{A}\hat{b^\dagger})] = [\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b}]\hat{b} + \hat{b^\dagger}[\hat{b}, \hat{\mu}\mathcal{A}\hat{b^\dagger})]$$

$$[\hat{b^\dagger}, \hat{\mu}\mathcal{A}\hat{b}]\hat{b} + \hat{b^\dagger}[\hat{b}, \hat{\mu}\mathcal{A}\hat{b^\dagger})] = \hat{\mu}\mathcal{A}((\hat{b^\dagger}\hat{b} - \hat{b}\hat{b^\dagger})\hat{b} + \hat{b^\dagger}(\hat{b}\hat{b^\dagger} - \hat{b^\dagger}\hat{b}))$$

$$\hat{\mu}\mathcal{A}((\hat{b^\dagger}\hat{b} - \hat{b}\hat{b^\dagger})\hat{b} + \hat{b^\dagger}(\hat{b}\hat{b^\dagger} - \hat{b^\dagger}\hat{b})) = i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger} - \hat{b})$$

Now for the second order term.

$$[[\hat{b^\dagger}\hat{b}, \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b}+\hat{b^\dagger})], \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b}+\hat{b^\dagger})] = [i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger} - \hat{b}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}\hat{b}] + [i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger} - \hat{b}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}\hat{b^\dagger}]$$

$$\begin{aligned}&[i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger} - \hat{b}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b}] + [i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger} - \hat{b}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}(\hat{b^\dagger}] = \\ &[i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}\hat{b}] + [i\omega_c\hat{\mu}\mathcal{A} - \hat{b}, \frac{-i}{\hbar}\hat{\mu}\mathcal{A}\hat{b^\dagger}]\end{aligned}$$

$$[i\omega_c\hat{\mu}\mathcal{A}(\hat{b^\dagger}), \frac{-i}{\hbar}\hat{\mu}\mathcal{A}\hat{b}] + [i\omega_c\hat{\mu}\mathcal{A} - \hat{b}, \frac{-i}{\hbar}\hat{\mu}\mathcal{A}\hat{b^\dagger}] = (\hat{\mu}\mathcal{A})^2$$

The last one is trivial.

7.5 Appendix C

The Thomas-Reich-Kuhn sum rule is a formula that sum over the energies for all possible transition states.[12; 34]

$$D = \sum_j [E_j - E_k](\langle k| \hat{x} |j\rangle)^2 \tag{7.8}$$

Where $\hat{x}$ is a operator, E_j and $\langle j|$ are the eigenstate of the Hamiltonian and correspondent eigenvalue. E_k and $|k\rangle$ are the particular state and the corresponding energy. where k usually denotes the ground state. In context to the matter light Hamiltonian, the Thomas-Reich-Kuhn can be rewritten as following.[12; 34]

$$D = \mathcal{A}^2 \sum_j [\omega_j - \omega_k](\langle k| \hat{\mu} |j\rangle)^2 \tag{7.9}$$