Light-Matter Interaction

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Light-matter interactions occur when charged particles accelerate in time-dependent electric field. An accelerating charge particle generates light, and conversely electric fields cause forces on charges particle. In most scenarios, magnetic field does not directly interact with matter since it is easier to have charges, electric dipoles than magnetic dipoles.

Time-dependent charges can be described by a charge density $\rho(\mathbf{r},t)$. It is more often to use dipoles and currents to describe light-matter interaction. Polarization $\mathbf{P}$ (dipole) and currents density $\mathbf{J}$ have the relations

$$\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} = 0, \tag{1}$$

$$\mathbf{J} = \frac{\partial \mathbf{P}}{\partial t}. \tag{2}$$

1 Hamiltonian

1.1 Interaction Hamiltonian

According to classical mechanics, a charge particle has the Hamiltonian (SI units)

$$\mathcal{H} = \frac{(\mathbf{p} - q\mathbf{A})^2}{2m} + q\Phi(\mathbf{r},t), \tag{3}$$

where q is the charge of the particle not the position. $\Phi(\mathbf{r},t)$ is the electric potential. In the case of an electron, $q = -e$, we have

$$\mathcal{H} = \frac{(\mathbf{p} + e\mathbf{A})^2}{2m} - e\Phi(\mathbf{r},t). \tag{4}$$

We can decompose it into $\mathcal{H}_0$ and $\mathcal{H}_I$,

$$\mathcal{H}_0 = \frac{p^2}{2m}, \tag{5}$$

$$\mathcal{H}_I = \frac{e(\mathbf{p}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{p})}{2m} + \frac{e^2A^2}{2m} - e\Phi. \tag{6}$$

Typically, the term $\frac{e^2A^2}{2m}$ is dropped since the momentum of field $e\mathbf{A}$ is usually small than the electron's momentum $\mathbf{p}$ [1]. Since the momentum $\mathbf{p}$ is a differential operator, $\mathbf{p}\cdot\mathbf{A}$ is not equal

[1] Well, this is a sloppy argument. In electromagnetism, the higher-order terms of the vector potential$\mathbf{A}$ are relativistic. In this viewpoint, the term $\frac{e^2A^2}{2m}$ is proportional to $\frac{v^2}{c^2}$.

to $\mathbf{A}\cdot\mathbf{p}$. The vector potential $\mathbf{A}$ and Coulomb's potential Φ are not unique. The Maxwell's equations are invariant under the gauge transformations

$$\mathbf{A}' = \mathbf{A} + \nabla\lambda(\mathbf{r}, t), \tag{7}$$

$$\Phi' = \Phi - \frac{\partial\lambda(\mathbf{r}, t)}{\partial t}. \tag{8}$$

The fields are given by

$$\mathbf{B} = \nabla \times \mathbf{A}, \tag{9}$$

$$\mathbf{E} = -\nabla\Phi - \frac{\partial\mathbf{A}}{\partial t}. \tag{10}$$

The Gauge $\nabla\cdot\mathbf{A} = 0$ is frequently used in quantum optics. In this gauge, the interaction Hamiltonian (dropping $\frac{e^2A^2}{2m}$) becomes

$$\mathcal{H}_I = \frac{e(\mathbf{A}\cdot\mathbf{p})}{m} - e\Phi. \tag{11}$$

If $\Phi = 0$ is chosen [2], the interaction Hamiltonian becomes

$$\mathcal{H}_I = \frac{e(\mathbf{A}\cdot\mathbf{p})}{m} \tag{12}$$

$$= -\int dv\mathbf{A}\cdot\mathbf{J} \tag{13}$$

where we use $\int dv\mathbf{J} = \frac{-e\mathbf{p}}{m}$. Another choice is the Göppert-Mayer gauge,

$$\lambda = -(\mathbf{r} - \mathbf{r}_0)\cdot\mathbf{A}(\mathbf{r}_0). \tag{14}$$

In this gauge, we have

$$\mathbf{A}' = \mathbf{A}(\mathbf{r}) - \mathbf{A}(\mathbf{r}_0), \tag{15}$$

$$\Phi' = -e(\mathbf{r} - \mathbf{r}_0)\cdot\mathbf{E}(\mathbf{r}_0) \equiv -\mathbf{d}\cdot\mathbf{E}, \tag{16}$$

where the dipole operator is $-e(\mathbf{r} - \mathbf{r}_0)$. The so-called dipole approximation is when $\mathbf{A}(\mathbf{r})$ is almost a constant, i.e., $\mathbf{A}(\mathbf{r}) \simeq \mathbf{A}(\mathbf{r}_0)$. This approximation is valid when the charge distributions are within a small region. The interaction Hamiltonian becomes

$$\mathcal{H}_I = -\mathbf{E}\cdot\mathbf{d} \tag{17}$$

[2] In the region without charges $\nabla\cdot\mathbf{E} = 0$, we can define $\mathbf{E} = -\nabla\Phi$. Using the gauge transformation $\lambda = \int\Phi dt$, we can eliminate Φ and make $\nabla\cdot\mathbf{A} = 0$.

1.2 Total Hamiltonian

The total Hamiltonian of the light-matter is

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I + \mathcal{H}_F. \tag{18}$$

where

$$\mathcal{H}_F = \sum_m \int dv \left(\frac{\epsilon(\mathbf{r}) E_m^2(\mathbf{r})}{2} + \frac{B_m^2(\mathbf{r})}{2\mu(\mathbf{r})} \right). \tag{19}$$

The Hamiltonian of matter $\mathcal{H}_0$ is not necessary of the form of a free particle. In general, $\mathcal{H}_0$ describes a N-level system,

$$\mathcal{H}_0 = \sum_n E_n |E_n\rangle\langle E_n|. \tag{20}$$

The simplest case is a two level system (TLS)

$$\mathcal{H}_{TLS} = \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix}. \tag{21}$$

The interaction Hamiltonian for a a two level system is

$$\mathcal{H}_I = \begin{pmatrix} \langle E_c| - \mathbf{E}\cdot\mathbf{d}|E_c\rangle & \langle E_c| - \mathbf{E}\cdot\mathbf{d}|E_v\rangle \\ \langle E_v| - \mathbf{E}\cdot\mathbf{d}|E_c\rangle & \langle E_v| - \mathbf{E}\cdot\mathbf{d}|E_v\rangle \end{pmatrix} \tag{22}$$

$$= -\mathbf{E}\cdot\begin{pmatrix} \mathbf{d}_{cc} & \mathbf{d}_{cv} \\ \mathbf{d}_{vc} & \mathbf{d}_{vv} \end{pmatrix}, \tag{23}$$

where the dipole matrix element is $\mathbf{d}_{nn'} = \langle E_n|\mathbf{d}|E_{n'}\rangle$. In many cases, the diagonal elements of dipole matrices vanishes since the eigenfunctions are typical symmetric.

2 Classical Fields and Quantum Matter

We consider that the matter is described by a N-level system and treat the electric field $\mathbf{E}(\mathbf{r}, t)$ as a number. The Hamiltonian is

$$\mathcal{H} = \sum_n E_n |E_n\rangle\langle E_n| - \mathbf{E}\cdot\mathbf{d}. \tag{24}$$

In the case of a TLS system, the Hamiltonian is

$$\mathcal{H} = \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \mathbf{E}\cdot\begin{pmatrix} 0 & \mathbf{d}_{cv} \\ \mathbf{d}_{vc} & 0 \end{pmatrix}, \tag{25}$$

where we assume the diagonal elements of the dipole matrix are zeros. To solve the dynamics, we start with the interaction picture where state is

$$|\psi\rangle = C_c(t)e^{-i\omega_c t}|E_c\rangle + C_v(t)e^{-i\omega_v t}|E_v\rangle. \tag{26}$$

It is clear that without an external field $\mathbf{E}$, the coefficients $C_c(t)$ and $C_v(t)$ are constant in time. Plugging Eq. (26) in the Schrödinger equation, we obtain

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix} C_c \\ C_v \end{pmatrix} = -\mathbf{E}\cdot\begin{pmatrix} 0 & \mathbf{d}_{cv}e^{i(\omega_c-\omega_v)t} \\ \mathbf{d}_{vc}e^{i(\omega_v-\omega_c)t} & 0 \end{pmatrix}\begin{pmatrix} C_c \\ C_v \end{pmatrix}. \tag{27}$$

The dipole matrix elements in the interaction picture oscillate rapidly in time. The electric field $\mathbf{E} = \boldsymbol{\mathcal{E}}_\omega e^{-i\omega t} + \boldsymbol{\mathcal{E}}^*_\omega e^{i\omega t}$ needs to have a frequency $\omega \simeq (\omega_c - \omega_v)$ in order to create transition. We write

$$\omega = \omega_{cv} + \Delta, \tag{28}$$

where $\omega_{cv} = \omega_c - \omega_v$ and Δ is the detuning.

2.1 Rabi Model

Let the external field $\mathbf{E} = \mathbf{E}_0\cos\omega t = \mathbf{E}_0\left(\frac{e^{-i\omega t}+e^{i\omega t}}{2}\right)$. The equation of the coefficients is

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix} C_c \\ C_v \end{pmatrix} = \begin{pmatrix} 0 & \frac{V_0}{2}\left[e^{-i\Delta t} + e^{i(2\omega_{cv}+\Delta)t}\right] \\ \frac{V_0^*}{2}\left[e^{i\Delta t} + e^{-i(2\omega_{cv}+\Delta)t}\right] & 0 \end{pmatrix}\begin{pmatrix} C_c \\ C_v \end{pmatrix}. \tag{29}$$

where

$$V_0 = -\mathbf{E}_0\cdot\mathbf{d}_{cv}. \tag{30}$$

The equation needs to be solved numerically. The rotating-wave-approximation (RWA), where the high frequency terms are dropped is often used. Under the RWA, the equation reads

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix} C_c \\ C_v \end{pmatrix} = \begin{pmatrix} 0 & \frac{V_0}{2}e^{-i\Delta t} \\ \frac{V_0^*}{2}e^{i\Delta t} & 0 \end{pmatrix}\begin{pmatrix} C_c \\ C_v \end{pmatrix}. \tag{31}$$

Eliminating the variable C_v, we obtain the second-order differential equation

$$\ddot{C}_c + i\Delta\dot{C}_c + \frac{|V_0|^2}{4\hbar^2}C_c = 0. \tag{32}$$

The general solution is

$$C_c(t) = A_+e^{i\lambda_+ t} + A_-e^{i\lambda_- t} \tag{33}$$

with

$$\lambda_{\pm} = \Delta \pm \sqrt{\Delta^2 + \frac{|V_0|^2}{\hbar^2}} \equiv \Delta \pm \Omega_R. \tag{34}$$

The Rabi frequency $\Omega_R = \sqrt{\Delta^2 + \frac{|V_0|^2}{\hbar^2}}$. If initially $C_v(0) = 1$, the solution is

$$C_e = e^{i\frac{\Delta t}{2}} \frac{iV_0}{\hbar\Omega_R} \sin\frac{\Omega_R t}{2}, \tag{35}$$

$$C_g = e^{i\frac{\Delta t}{2}} \left[\cos\frac{\Omega_R t}{2} - i\frac{\Delta}{\Omega_R}\sin\frac{\Omega_R t}{2}\right]. \tag{36}$$

It can be checked that $|C_c|^2 + |C_v|^2 = 1$.

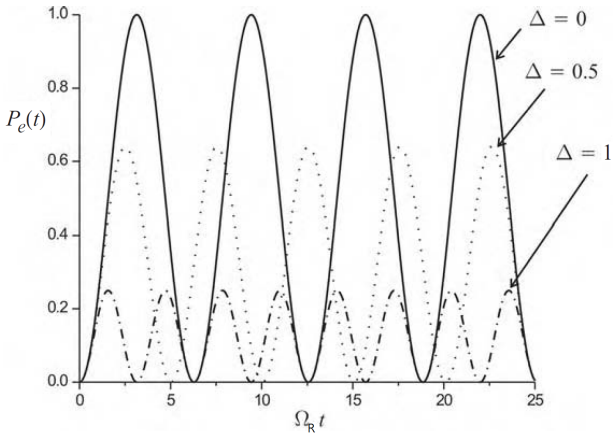

Figure 1: Population of the excited state as a function of time

2.2 Fermi's Golden Rule

If the external field is small, one can apply the perturbation method (for example, see Chapter 5 of Ref. [1]) to obtain (or from Eq. (35))

$$P_c(t) = |C_c|^2 = \frac{|V_0|^2 \sin^2\frac{\Delta t}{2}}{\hbar^2\Delta^2}. \tag{37}$$

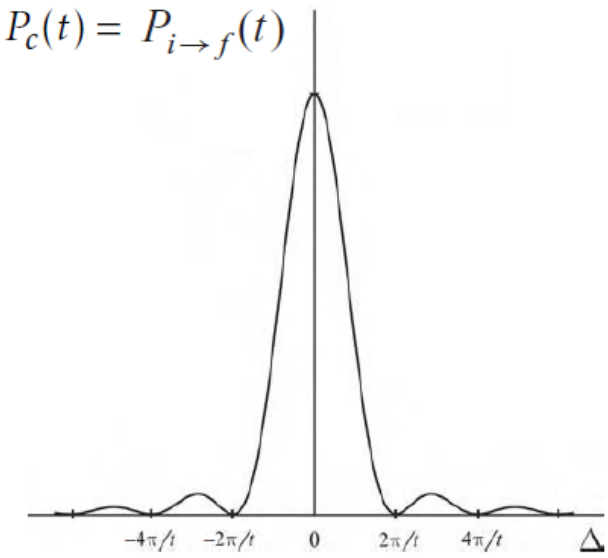

Figure 2: The transition probability $P_c(t)$ at a momentum t. When t is large, the function is approximately a delta function.

When t is large, the fraction is approximately a delta function

$$\frac{\sin^2 \frac{\Delta t}{2}}{\Delta^2} \simeq \frac{\pi t}{2}\delta(\Delta). \tag{38}$$

The transition rate $W_{v\to c}$ is

$$W_{v\to c} = \frac{P_c(t)}{t} = \frac{\pi}{2}\frac{|V_0|^2}{\hbar^2}\delta(\omega - \omega_{cv}) \tag{39}$$

$$= \frac{\pi}{2}\frac{|\mathbf{E}_0 \cdot \mathbf{d}_{cv}|^2}{\hbar^2}\delta(\omega - \omega_{cv}) \tag{40}$$

$$= \frac{\pi}{2}\frac{|\langle c|\mathbf{H}_I|v\rangle|^2}{\hbar^2}\delta(\omega - \omega_{cv}), \tag{41}$$

which is the famous Fermi's Golden rule.

3 Classical Matter and Quantum Fields

Currents and charges are treated as classical numbers. Time-dependent charges and currents are not independent variables. They are related by the continuity equation. This assumption is adequate when currents come form a lot of electrons and the quantum fluctuations are ignored. The typical problem is how a current source $\mathbf{j}(\mathbf{r}, t)$ interacts with photons. Thus, currents are

given functions, and the problem is to solve filed Hamiltonian.

$$\mathcal{H} = \mathcal{H}_F + \mathcal{H}_I \tag{42}$$

$$= \sum_m \hbar\omega_m a_m^\dagger a_m - \sum_m \mathbf{E}_m \cdot \mathbf{d} \tag{43}$$

$$= \sum_m \hbar\omega_m a_m^\dagger a_m - \sum_m \left(\frac{\boldsymbol{\mathcal{E}}_m a + \boldsymbol{\mathcal{E}}_m^* a^\dagger}{2} \right) \cdot \mathbf{d}, \tag{44}$$

where we should treat the dipole as a classical function with a frequency ω ,

$$\mathbf{d} = \mathbf{d}_0 e^{-i\omega t} = \int dv \mathbf{P} = \int dv \mathbf{P}_0 e^{-i\omega t} \tag{45}$$

$$= -i\omega \int dv \mathbf{J}. \tag{46}$$

We can also use vector potential operators via the relation

$$\mathbf{E} = -\frac{\partial}{\partial t}\mathbf{A}, \tag{47}$$

$$\mathbf{A} = \left(\frac{\boldsymbol{\mathcal{E}}_m a - \boldsymbol{\mathcal{E}}_m^* a^\dagger}{2i\omega} \right). \tag{48}$$

Considering a single mode and $\omega_m = \omega$, the Hamiltonian becomes

$$\mathcal{H} = \hbar\omega a^\dagger a - \int dv \mathbf{A} \cdot \mathbf{J}. \tag{49}$$

3.1 Generation of Coherent States

We are going to show a coherent state $|\alpha\rangle$ can be generate by a harmonic oscillating current density $\mathbf{J} = \mathbf{J}_0(\mathbf{r}) e^{i\omega t}$. [3] The Hamiltonian becomes

$$\mathcal{H} = \hbar\omega a^\dagger a + \left(V_0 a + V_0^* a^\dagger \right), \tag{50}$$

where

$$V_0 = i\omega e^{i\omega t} \int dv \boldsymbol{\mathcal{E}}_\omega(\mathbf{r}) \cdot \mathbf{J}_0(\mathbf{r}). \tag{51}$$

In the interaction picture, the interaction Hamiltonian becomes

$$\mathcal{H}_I = \left(V_I a + V_I^* a^\dagger \right), \tag{52}$$

where the interaction potenital is time-indepdenent and reads

[3] Physical currents should be real. We can think that $e^{i\omega t}$ comes from $\cos \omega t$

$$V_I = i\omega \int dv \boldsymbol{\mathcal{E}}_\omega(\mathbf{r}) \cdot \mathbf{J}_0(\mathbf{r}). \tag{53}$$

The evolution of a state is given by

$$|\psi(t)\rangle_I = \hat{\mathcal{T}}[e^{-i\int \frac{\mathcal{H}_I(t)}{\hbar} dt}]|\psi(0)\rangle_I \tag{54}$$

where $\hat{\mathcal{T}}[]$ denotes the time-ordering[4]. In this case, the interaction Hamiltonian in the interaction picture is time-independent,

$$|\psi(t)\rangle_I = e^{-i\frac{\mathcal{H}_I(t)}{\hbar}t}|\psi(0)\rangle_I \tag{55}$$

$$= e^{\alpha^* a - \alpha a^\dagger}|\psi(0)\rangle_I, \tag{56}$$

where

$$\alpha = i\frac{V_I^*}{\hbar}t \tag{57}$$

$$= \frac{i\omega \int dv \boldsymbol{\mathcal{E}}^*_\omega(\mathbf{r}) \cdot \mathbf{J}^*_0(\mathbf{r})}{\hbar} t. \tag{58}$$

Equation (56) is indeed the displacement operator. If the initial state is the ground state $|0\rangle$, the final state is a coherent state,

$$|\psi(t)\rangle_I = e^{\alpha^* a - \alpha a^\dagger}|0\rangle \tag{59}$$

$$= |\alpha\rangle. \tag{60}$$

One interesting observation is that $|\alpha| \sim t$ and the photon number $n \sim t^2$ grows quadratically.

4 Fully Quantum Approach

Both matter and field are quantized. The Hamiltonian is

$$\mathcal{H} = \sum_m \hbar\omega_m a_m^\dagger a_m + \sum_n E_n |E_n\rangle\langle E_n| - \mathbf{E}\cdot\mathbf{d}, \tag{61}$$

where the electric field operator is

$$\mathbf{E} = \sum_m \frac{\boldsymbol{\mathcal{E}}_m a_m + \boldsymbol{\mathcal{E}}^*_m a_m^\dagger}{2}, \tag{62}$$

and the dipole matrix operator is

$$\begin{pmatrix} \mathbf{d}_{11} & \mathbf{d}_{12} & \cdots \\ \mathbf{d}_{21} & \mathbf{d}_{22} & \\ \vdots & & \ddots \end{pmatrix}, \tag{63}$$

with $\mathbf{d}_{nn'} = \langle E_n|\mathbf{d}|E_{n'}\rangle$ and $\mathbf{d} = q\mathbf{r}$.

[4]Time-ordering is necessary if H_I is time-dependent and $[H_I(t_1), H_I(t_2)] \neq 0$

4.1 Two-Level System and Single-Mode Photons

The Hamiltonian is

$$\mathcal{H} = \hbar\omega a^\dagger a + \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \mathbf{E}\cdot\mathbf{d}. \tag{64}$$

where the electric field operator is

$$\mathbf{E} = \frac{\boldsymbol{\mathcal{E}}_\omega a + \boldsymbol{\mathcal{E}}_\omega^* a^\dagger}{2}, \tag{65}$$

and the dipole matrix operator is

$$\begin{pmatrix} 0 & \mathbf{d}_{cv} \\ \mathbf{d}_{vc} & 0 \end{pmatrix}, \tag{66}$$

where we assume that the diagonal terms vanish. The transition rate from $|n\rangle|E_c\rangle$ to $|n+1\rangle|E_v\rangle$ is obtained by

$$W_{\text{emission}} = \frac{\pi}{2}\frac{|\langle n+1|\langle E_v|\mathbf{H}_I|n\rangle|E_c\rangle|^2}{\hbar^2}\delta(\omega-\omega_{cv}) \tag{67}$$

$$= \frac{(n+1)\pi}{2}\frac{|\boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}|^2}{\hbar^2}\delta(\omega-\omega_{cv}). \tag{68}$$

An interesting result occurs when $n = 0$. The emission is not zero when $n = 0$. This is the phenomenon "spontaneous emission". When $n > 0$, it corresponds to the stimulated emission. The transition rate from $|n\rangle|E_v\rangle$ to $|n-1\rangle|E_c\rangle$ is obtained by

$$W_{\text{absorption}} = \frac{\pi}{2}\frac{|\langle n-1|\langle E_c|\mathbf{H}_I|n\rangle|E_v\rangle|^2}{\hbar^2}\delta(\omega-\omega_{cv}) \tag{69}$$

$$= \frac{n\pi}{2}\frac{|\boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{vc}|^2}{\hbar^2}\delta(\omega-\omega_{cv}). \tag{70}$$

4.2 Jaynes–Cummings Model

The TLS and single-mode photon Hamiltonian can be further simplified with the RWA,

The Hamiltonian is

$$\mathcal{H} = \hbar\omega a^\dagger a + \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \frac{1}{2}\begin{pmatrix} 0 & \boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}a + \boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{cv}a^\dagger \\ \boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{vc}a + \boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{vc}a^\dagger & 0 \end{pmatrix} \tag{71}$$

$$\simeq \hbar\omega a^\dagger a + \begin{pmatrix} E_c & 0 \\ 0 & E_v \end{pmatrix} - \frac{1}{2}\begin{pmatrix} 0 & \boldsymbol{\mathcal{E}}_\omega\cdot\mathbf{d}_{cv}a \\ \boldsymbol{\mathcal{E}}_\omega^*\cdot\mathbf{d}_{vc}a & 0 \end{pmatrix} \tag{72}$$

$$= \hbar\omega a^\dagger a + \frac{E_c+E_v}{2} + \frac{\hbar\omega_{cv}}{2}\sigma_z + \hbar\left(\lambda\sigma_+ a + \lambda^*\sigma_- a^\dagger\right) \tag{73}$$

where

$$\lambda = \frac{-1}{2}\boldsymbol{\mathcal{E}}_\omega \cdot \mathbf{d}_{cv}. \tag{74}$$

The average energy $\frac{E_c+E_v}{2}$ is only a constant so as irrelavent to dynamics. In most cases, it is possible to make λ real by choosing the phase of $\mathbf{d}_{cv}$. The Jaynes–Cummings Model is then obtained as

$$\mathcal{H}_{JC} = \hbar\omega a^\dagger a + \frac{\hbar\omega_{cv}}{2}\sigma_z + \hbar\lambda\left(\sigma_+ a + \sigma_- a^\dagger\right). \tag{75}$$

We have used the Pauli matrices

$$\sigma_z = |E_c\rangle\langle E_c| - |E_v\rangle\langle E_v| = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{76}$$

$$\sigma_+ = |E_c\rangle\langle E_v| = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \tag{77}$$

$$\sigma_- = |E_v\rangle\langle E_c| = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \tag{78}$$

The electron number operator is an identity,

$$N_e = |E_c\rangle\langle E_c| + |E_v\rangle\langle E_v|, \tag{79}$$

and the excitation number operator is

$$N_{ex} = |E_c\rangle\langle E_c| + a^\dagger a. \tag{80}$$

These numbers are conservative since the commutators vanish

$$[H, N_e] = 0, \tag{81}$$

$$[H, N_{ex}] = 0. \tag{82}$$

Exercise 1: Excitation Number

Show Eq. (82).

References

[1] J. J. Sakurai, *Modern Quantum Mechanics*, 1994 and 2010