

Chapter 8

Density Matrix Formalism

8.1 Density Matrix Description

So far we have limited all discussions to so-called *pure states*, i.e. states that can be described by a wave function. $|\psi\rangle$. However, there exist states which cannot be described by a wave function, such as thermal states of motion, unpolarized light, or states prepared through a decoherence process such as spontaneous emission. Here a more general description is needed.

8.1.1 Basic Formalism

The general form of the density matrix of a state can be written as

$$\rho = \sum_i p_i |\psi_i\rangle \langle\psi_i|. \quad (8.1)$$

where the p_i coefficients are the relative weight of being found in state $|\psi_i\rangle$. So-called *pure* states have a single $p_j = 1$, while all other $p_{i \neq j} = 0$, resulting in the density matrix $\rho = |\psi_j\rangle \langle\psi_j|$. States which have more than one non-zero p_i are commonly referred to as *mixed* states.

Normalization of density matrices is enforced by the requirement that its trace is unity, i.e. $\text{Tr}(\rho) = 1$. This means

$$\begin{aligned} \text{Tr}(\rho) &= \sum_j \langle\psi_j| \rho |\psi_j\rangle \\ &= \sum_j \langle\psi_j| \sum_i p_i |\psi_i\rangle \langle\psi_i| \psi_j\rangle \\ &= \sum_i p_i \equiv 1 \end{aligned} \quad (8.2)$$

We should therefore interpret the p_i coefficients as the probability of being in state $|\psi_i\rangle$. In fact, for a general operator \hat{O} we extend it's expectation value to mixed states:

$$\begin{aligned}
\langle \hat{O} \rangle &= \sum_i p_i \langle \psi_i | \hat{O} | \psi_i \rangle \\
&= \sum_j \sum_i p_i \langle \psi_i | \psi_j \rangle \langle \psi_j | \hat{O} | \psi_i \rangle \\
&= \sum_j \sum_i p_j \langle \psi_i | \psi_j \rangle \langle \psi_j | \hat{O} | \psi_i \rangle \\
&= \sum_i \langle \psi_i | \left(\sum_j p_j |\psi_j\rangle \langle \psi_j| \hat{O} \right) | \psi_i \rangle \\
&= \sum_i \langle \psi_i | \rho \hat{O} | \psi_i \rangle = \text{Tr}(\rho \hat{O})
\end{aligned} \tag{8.3}$$

where in the second line we have inserted the identity operator $\mathbb{1} = \sum_j |\psi_j\rangle \langle \psi_j|$. We could have inserted the identity on the other side of \hat{O} , and we would have obtained $\langle \hat{O} \rangle = \text{Tr}(\hat{O} \rho)$. Therefore we have

$$\langle \hat{O} \rangle = \text{Tr}(\rho \hat{O}) = \text{Tr}(\hat{O} \rho) \tag{8.4}$$

Note also that, if given a density matrix, it is easy to determine if it is a mixed or pure state by looking at $\text{Tr}(\rho^2)$:

$$\begin{aligned}
\text{Tr}(\rho^2) &= \sum_k \langle \psi_k | \sum_j p_j |\psi_j\rangle \langle \psi_j| \sum_i p_i |\psi_i\rangle \langle \psi_i| \psi_k \rangle \\
&= \sum_k \sum_j \sum_i \delta_{kj} \delta_{ji} \delta_{ik} p_j p_i \\
&= \sum_i p_i^2
\end{aligned} \tag{8.5}$$

$$= \sum_i p_i^2 \tag{8.6}$$

For a pure state, and *only* for a pure state, this expression equals unity, i.e. $\text{Tr}(\rho^2) = \text{Tr}(\rho) = 1$.

8.1.2 Matrix Representation

In any N -dimensional Hilbert space, we are free to choose a basis $\{|i\rangle\}_{i=1}^N$ which spans space. In this basis the density matrix can be written as:

$$\rho = \begin{pmatrix} \rho_{00} & \rho_{01} & \dots & \rho_{0N} \\ \rho_{10} & \rho_{11} & \dots & \rho_{1N} \\ \vdots & \vdots & \ddots & \vdots \\ \rho_{N0} & \rho_{N1} & \dots & \rho_{NN} \end{pmatrix} \tag{8.7}$$

The diagonal terms $\rho_{ii} \in (\mathbb{R} \geq 0)$ are called the *populations*, and must obey $\sum_{i=1}^N \rho_{ii} = 1$. The off-diagonal terms $\rho_{ij} \in \mathbb{C}$ are called the *coherences*, and must obey $\rho_{ij} = \rho_{ji}^*$, since the density matrix is Hermitian.

8.1.3 Density Matrix Time Evolution

We can see how the density matrix evolves in time by using the Schrödinger equation:

$$\begin{aligned}
\frac{d\rho}{dt} &= \sum_i p_i \left[\left(\frac{d}{dt} |\psi_i\rangle \right) \langle\psi_i| + |\psi_i\rangle \left(\frac{d}{dt} \langle\psi_i| \right) \right] \\
&= \sum_i p_i \left[\frac{H}{i\hbar} |\psi_i\rangle \langle\psi_i| + |\psi_i\rangle \langle\psi_i| \frac{H}{-i\hbar} \right] \\
&= -\frac{i}{\hbar} (H\rho - \rho H) = -\frac{i}{\hbar} [H, \rho]
\end{aligned} \tag{8.8}$$

This is known as the von Neumann equation, and it is a direct analogue of the Schrödinger equation for density matrices. If the Hamiltonian does not depend on time, this can be easily solved to give

$$\rho(t) = e^{-iHt/\hbar} \rho(0) e^{iHt/\hbar} \tag{8.9}$$

which makes sense, since consistency demands that:

$$\begin{aligned}
\rho(t) &= \sum_i p_i |\psi_i(t)\rangle \langle\psi_i(t)| \\
&= \sum_i p_i e^{-iHt/\hbar} |\psi_i(0)\rangle \langle\psi_i(0)| e^{iHt/\hbar} \\
&= e^{-iHt/\hbar} \rho(0) e^{iHt/\hbar}
\end{aligned} \tag{8.10}$$

8.1.4 Partial Trace

Consider a quantum mechanical system comprised of two coupled sub-systems A and B . The coupled system's state is given by the density matrix ρ_{AB} in the total Hilbert space \mathcal{H}_{AB} is made up of states in the tensor product $\mathcal{H}_A \otimes \mathcal{H}_B$ of the Hilbert spaces of the subsystems. The partial trace of the combined state with respect to subsystem B leaves us with the reduced density matrix of subsystem A , denoted by ρ_A . Mathematically, this means:

$$\text{Tr}_B(\rho_{AB}) = \rho_A \tag{8.11}$$

Suppose we have an operator X , which is defined in subspace A . In order for the partial trace to be a sensible operation, we must have the following:

$$\text{Tr}(X\rho_A) = \text{Tr}[(X \otimes \mathbb{1}_B)\rho_{AB}] \tag{8.12}$$

We can verify this is true by:

$$\begin{aligned}
\text{Tr}[(X \otimes \mathbb{1}_B)\rho_{AB}] &= \sum_a \sum_b (\langle a| \otimes \langle b|) (X \otimes \mathbb{1}_B) \rho_{AB} (|a\rangle \otimes |b\rangle) \\
&= \sum_a \sum_b \sum_{b'} (\langle a| \otimes \langle b|) (X \otimes |b'\rangle \langle b'|) \rho_{AB} (|a\rangle \otimes |b\rangle) \\
&= \sum_a \sum_b \langle a| X \langle b| \rho_{AB} |b\rangle (|a\rangle) \\
&= \sum_a \langle a| X \rho_A |a\rangle = \text{Tr}(X\rho_A)
\end{aligned} \tag{8.13}$$

In practice the partial trace is often used to "trace out" a system we have no knowledge of and/or we want to average over. In fact, it provides a way to uniquely describe subsystem A in the event we have no access to measurements in subsystem B .

8.2 Open Quantum Systems and Lindblad Master Equation

An *open quantum system* allows for coupling between the system of interest and the external environment, or *bath*. In general, the bath has an infinite number of degrees of freedom and is too complex to simulate or describe analytically, but with some simplifying assumptions we can obtain a powerful result known as the master equation in Lindblad form, also known as the Gorini–Kossakowski–Sudarshan–Lindblad equation (GKSL equation), or simply the Lindbladian. We present here a somewhat simplified derivation of the Lindblad master equation for a weakly interacting system and bath.

A general interaction between a system and bath can be described by the Hamiltonian

$$H_T = H + H_B + H_{SB} \quad (8.14)$$

where H describes the system dynamics, H_B the bath dynamics, and H_{SB} describes the system-bath interaction. We have implied the tensor multiplication of the system and bath's Hilbert space, and their respective Hamiltonians would be more accurately written as $H \otimes \mathbb{1}_B$ and $\mathbb{1} \otimes H_B$.

The total (system and bath) state is given by the total density matrix ρ_T , which evolves according to the von Neumann equation:

$$\dot{\rho}_T = -\frac{i}{\hbar} [H + H_B + H_{SB}, \rho_T] \quad (8.15)$$

Generally, we are only interested in the system dynamics, and therefore we want eliminate the bath's degrees of freedom by taking the partial trace over the bath: $\rho_S = \text{Tr}_B(\rho_T)$. We will first, however, need to determine how ρ_T evolves.

It is useful to move to the interaction picture, where we define $H_0 = H + H_B$ and $\tilde{\rho}_T = e^{-iH_0t/\hbar} \rho_T e^{iH_0t/\hbar}$ to see that

$$\begin{aligned} \dot{\tilde{\rho}}_T &= e^{iH_0t/\hbar} \left(i \frac{H_0}{\hbar} \rho_T + \dot{\rho}_T - i \rho_T \frac{H_0}{\hbar} \right) e^{-iH_0t/\hbar} \\ &= -\frac{i}{\hbar} e^{iH_0t/\hbar} (-H_0 \rho_T + [H_0 + H_{SB}, \rho_T] + \rho_T H_0) e^{-iH_0t/\hbar} \\ &= -\frac{i}{\hbar} e^{iH_0t/\hbar} (H_{SB} \rho_T - \rho_T H_{SB}) e^{-iH_0t/\hbar} \\ &= -\frac{i}{\hbar} e^{iH_0t/\hbar} (H_{SB} e^{-iH_0t/\hbar} e^{iH_0t/\hbar} \rho_T - \rho_T e^{-iH_0t/\hbar} e^{iH_0t/\hbar} H_{SB}) e^{-iH_0t/\hbar} \\ &= -\frac{i}{\hbar} \left[\tilde{H}_{SB}, \tilde{\rho}_T \right] \end{aligned} \quad (8.16)$$

where we have made the substitutions $\tilde{H}_{SB} = \exp(iH_0t/\hbar)H_{SB}\exp(-iH_0t/\hbar)$ to yield the effective interaction Hamiltonian, and $\tilde{\rho}_T = \exp(iH_0t/\hbar)\rho_T\exp(-iH_0t/\hbar)$.

Eq. 8.16 can be integrated to yield

$$\tilde{\rho}_T(t) = \tilde{\rho}_T(0) - \frac{i}{\hbar} \int_0^t dt' \left[\tilde{H}_{SB}(t'), \tilde{\rho}_T(t') \right] \quad (8.17)$$

which we can plug back into the von Neuman equation:

$$\dot{\tilde{\rho}}_T = -\frac{i}{\hbar} \left[\tilde{H}_{SB}, \tilde{\rho}_T(0) \right] - \frac{1}{\hbar^2} \int_0^t dt' \left[\tilde{H}_{SB}(t), \left[\tilde{H}_{SB}(t'), \tilde{\rho}_T(t') \right] \right] \quad (8.18)$$

We can safely assume that the system and bath were uncorrelated before the interaction was turned on, and therefore the first term on the right side of Eq. 8.18 vanishes. We now take the partial trace over the bath's degrees of freedom to arrive at an equation for $\tilde{\rho}_S = \text{Tr}_B(\tilde{\rho}_T) \equiv \tilde{\rho}$

$$\dot{\tilde{\rho}} = -\frac{1}{\hbar^2} \int_0^t dt' \text{Tr}_B \left(\left[\tilde{H}_{SB}(t), \left[\tilde{H}_{SB}(t'), \tilde{\rho}_T(t') \right] \right] \right) \quad (8.19)$$

This equation for the dynamics of the system is exact, but it still requires knowledge of the bath's dynamics. To move past this point we make what is known as the *Born approximation*: The system and bath interact only weakly, and the bath is "large", and so the bath's eigenstates are not changed by the interaction. Since the system and bath were initially decoupled, this means that $\tilde{\rho}_T(t) = \tilde{\rho}(t) \otimes \tilde{\rho}_B(0)$. Eq. 8.19 then becomes

$$\dot{\tilde{\rho}} = -\frac{1}{\hbar^2} \int_0^t dt' \text{Tr}_B \left(\left[\tilde{H}_{SB}(t), \left[\tilde{H}_{SB}(t'), \tilde{\rho}(t') \otimes \tilde{\rho}_B(0) \right] \right] \right) \quad (8.20)$$

We now make the assumption that the time derivative of the system's density matrix only depends on the density matrix at the current time, but not on the density matrix at any prior time, which also means that any correlations within the bath are quickly lost. This is the *Born-Markov approximation* and allows us to make the replacement $\tilde{\rho}(t') = \tilde{\rho}(t)$ in Eq. 8.20:

$$\dot{\tilde{\rho}} = -\frac{1}{\hbar^2} \int_0^t dt' \text{Tr}_B \left(\left[\tilde{H}_{SB}(t), \left[\tilde{H}_{SB}(t'), \tilde{\rho}(t) \otimes \tilde{\rho}_B(0) \right] \right] \right) \quad (8.21)$$

A general form of the interaction Hamiltonian is given by:

$$\tilde{H}_{SB}(t) = \hbar \sum_i S_i(t) \otimes B_i(t) = \hbar \sum_i S_i^\dagger(t) \otimes B_i^\dagger(t) \quad (8.22)$$

Due to the Hermiticity of the Hamiltonian, these two are equivalent and we can choose which version to insert. In the following we choose the non-daggered (daggered) form to replace $H_{SB}(t')$ ($H_{SB}(t)$) in the first and third terms, and opposite for the second and fourth term.

which can be plugged into Eq. 8.21 and expanded to yield:

$$\begin{aligned} \dot{\tilde{\rho}} = & - \sum_i \sum_j \int_0^t dt' \{ S_i^\dagger(t) S_j(t') \tilde{\rho}(t) \text{Tr} [B_i^\dagger(t) B_j(t') \tilde{\rho}_B(0)] \\ & - S_i(t) \tilde{\rho}(t) S_j^\dagger(t') \text{Tr} [B_i(t) \tilde{\rho}_B(0) B_j^\dagger(t')] \\ & - S_j(t') \tilde{\rho}(t) S_i^\dagger(t) \text{Tr} [B_j(t') \tilde{\rho}_B(0) B_i^\dagger(t)] \\ & + \tilde{\rho}(t) S_j^\dagger(t') S_i(t) \text{Tr} [\tilde{\rho}_B(0) B_j^\dagger(t') B_i(t)] \} \end{aligned} \quad (8.23)$$

We now use the cyclic property of traces to see that:

$$\text{Tr} [B_j(t') \tilde{\rho}_B B_i^\dagger(t)] = \text{Tr} [\tilde{\rho}_B B_i^\dagger(t) B_j(t')] = \text{Tr} [B_i^\dagger(t) B_j(t') \tilde{\rho}_B] \equiv \frac{h_{ij}}{2} \delta(t' - t) \quad (8.24)$$

$$\text{Tr} [B_i(t) \tilde{\rho}_B B_j^\dagger(t')] = \text{Tr} [\tilde{\rho}_B B_j^\dagger(t') B_i(t)] = \text{Tr} [B_j^\dagger(t') B_i(t) \tilde{\rho}_B] \equiv \frac{h_{ji}}{2} \delta(t' - t) \quad (8.25)$$

where the final equivalence is made assuming that correlations within the bath die out very quickly compared to the system's evolution. The h_{ij} factors are general numerical factors which result from performing the trace over the bath. This is another consequence of the weak system-bath interaction. Now Eq. 8.23 simplifies to

$$\dot{\tilde{\rho}} = - \sum_i \sum_j \left\{ \frac{h_{ij}}{2} [S_i^\dagger(t) S_j(t) \tilde{\rho}(t) - S_j(t) \tilde{\rho}(t) S_i^\dagger(t)] + \frac{h_{ji}}{2} [\tilde{\rho}(t) S_j^\dagger(t) S_i(t) - S_i(t) \tilde{\rho}(t) S_j^\dagger(t)] \right\} \quad (8.26)$$

Since i and j sum over the same operators we are free to swap the indices on the third and fourth terms to obtain (dropping the explicit time-dependent notation):

$$\begin{aligned} \dot{\tilde{\rho}} = & - \sum_i \sum_j \left\{ \frac{h_{ij}}{2} [S_i^\dagger S_j \tilde{\rho} - S_j \tilde{\rho} S_i^\dagger] + \frac{h_{ij}}{2} [\tilde{\rho} S_i^\dagger S_j - S_j \tilde{\rho} S_i^\dagger] \right\} \\ = & \sum_i \sum_j h_{ij} \left\{ S_j \tilde{\rho} S_i^\dagger - \frac{1}{2} (S_i^\dagger S_j \tilde{\rho} + \tilde{\rho} S_i^\dagger S_j) \right\} \end{aligned} \quad (8.27)$$

Finally, it is possible to diagonalize the matrix h using an appropriate unitary transformation $\Gamma = dh d^\dagger$, such that

$$\Gamma_i = \sum_k \sum_l d_{ik} h_{kl} d_{li}^* \quad (8.28)$$

$$L_i = \sum_k d_{ik} S_k \quad (8.29)$$

Using this, and moving back to the Schrödinger picture, Eq. 8.27 to the standard form of the quantum master equation in Lindblad form:

$$\begin{aligned} \dot{\rho} = & - \frac{i}{\hbar} [H, \rho] + \sum_i \Gamma_i \left[L_i \rho L_i^\dagger - \frac{1}{2} (L_i^\dagger L_i \rho + \rho L_i^\dagger L_i) \right] \\ = & - \frac{i}{\hbar} [H, \rho] + \sum_i \mathcal{D}[\sqrt{\Gamma_i} L_i](\rho) \end{aligned} \quad (8.30)$$

where $\mathcal{D}[L](\rho)$ is known as the *Lindblad superoperator* acting on ρ . It is "super" because it acts on the density matrix from both the left and right side, unlike a conventional operator which only acts on one side. Note that often the jump operators are defined as $\tilde{L}_i = \sqrt{\Gamma_i} L_i$ to yield the Lindbladian:

$$\begin{aligned}\dot{\rho} &= -\frac{i}{\hbar} [H, \rho] + \sum_i \left[\tilde{L}_i \rho \tilde{L}_i^\dagger - \frac{1}{2} \left(\tilde{L}_i^\dagger \tilde{L}_i \rho + \rho \tilde{L}_i^\dagger \tilde{L}_i \right) \right] \\ &= -\frac{i}{\hbar} [H, \rho] + \sum_i \mathcal{D}[\tilde{L}_i](\rho)\end{aligned}\quad (8.31)$$

The Lindblad superoperator has some nice properties. It is trace-preserving on ρ , and it is Hermitian, so ρ will stay Hermitian as it evolves in time.

8.3 Three-level Systems

We will now apply our knowledge of density matrices to three-level systems in a general way. A combination of two- and three-level systems allow for an understanding of the majority of the atomic physics experimental toolbox. Two-level systems allow for the description of such phenomena as Doppler cooling, optical molasses, polarization gradient cooling, etc. Three-level systems, on the other hand, allow for richer behavior including two-photon (Raman) transitions and Raman sideband cooling, electromagnetically induced transparency (EIT), EIT cooling, as well as entangling (two qubit) gates. In this section we will go through a framework description which is generally useful for the description of three-level systems.

8.3.1 Operators in rotating frame

We start from the master equation for a system which evolves coherently according to a Hamiltonian $H = H_0 + H_I$. Typically we work in a basis where H_0 is diagonal and H_I are the off-diagonal interaction terms. We also define an arbitrary Hamiltonian H_D which commutes with the H_0 part of the original Hamiltonian, i.e. $[H_0, H_D] = 0$, and is typically therefore also diagonal. We can move the von Neumann equation from the Schrödinger picture, as shown in Eq. 8.8, to a rotating frame defined by:

$$\tilde{\rho} = e^{iH_D t/\hbar} \rho e^{-iH_D t/\hbar} \quad (8.32)$$

Note that the case where $H_D = H_0$ we are simply moving to the interaction picture. The time derivative of $\tilde{\rho}$ is then (similarly to the derivation in Eq. 8.16):

$$\begin{aligned}\dot{\tilde{\rho}} &= e^{iH_D t/\hbar} \left(i \frac{H_D}{\hbar} \rho + \dot{\rho} - i \rho \frac{H_D}{\hbar} \right) e^{-iH_D t/\hbar} \\ &= -\frac{i}{\hbar} [H_0 - H_D, \tilde{\rho}] - \frac{i}{\hbar} e^{iH_D t/\hbar} \left(H_I \rho - \rho H_I + \sum_i \mathcal{D}[L_i](\rho) \right) e^{-iH_D t/\hbar} \\ &= -\frac{i}{\hbar} [H_0 - H_D + \tilde{H}_I, \tilde{\rho}] + \sum_i \mathcal{D}[\tilde{L}_i](\tilde{\rho}) \equiv -\frac{i}{\hbar} [\tilde{H}, \tilde{\rho}] + \sum_i \mathcal{D}[\tilde{L}_i](\tilde{\rho})\end{aligned}\quad (8.33)$$

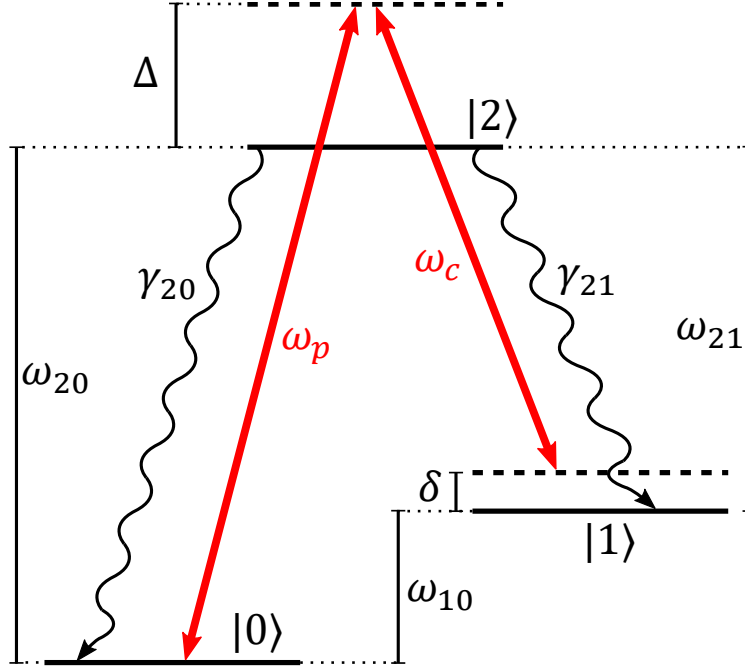


Figure 8.1: A general three-level system. All frequencies are positive as picture. Applied probe and coupling fields are shown in red.

Here, we have defined $\tilde{H}_I = \exp(iH_D t/\hbar)H_I \exp(-iH_D t/\hbar)$, and similar for the \tilde{L}_i jump operators. In general, the expectation value of an operator A is:

$$\begin{aligned}
 \langle A \rangle &= \text{Tr}(A\rho) \\
 &= \sum_i \sum_j p_j \langle \psi_i | A | \psi_j \rangle \langle \psi_j | \psi_i \rangle \\
 &= \sum_i \sum_j p_j \langle \psi_i | e^{-iH_D t/\hbar} e^{iH_D t/\hbar} A e^{-iH_D t/\hbar} e^{iH_D t/\hbar} | \psi_j \rangle \langle \psi_j | e^{-iH_D t/\hbar} e^{iH_D t/\hbar} | \psi_i \rangle \quad (8.34) \\
 &= \sum_i \sum_j p_j \langle \tilde{\psi}_i | \tilde{A} | \tilde{\psi}_j \rangle \langle \tilde{\psi}_j | \tilde{\psi}_i \rangle = \text{Tr}(\tilde{A}\tilde{\rho})
 \end{aligned}$$

Here we define $\tilde{A} = \exp(iH_D t/\hbar)A \exp(-iH_D t/\hbar)$ as the operator A in the rotating frame.

8.3.2 Semi-classical three-level atom Hamiltonian

Let us now consider the specific case of a three-level system in the Λ configuration (see) in a harmonic potential interacting with two classical electric fields, as shown in Fig. 8.1. We can describe the energy levels of the atom using:

$$H_0^{(A)} = \hbar\omega_{10} |1\rangle \langle 1| + \hbar\omega_{20} |2\rangle \langle 2| \quad (8.35)$$

where the (A) superscript denotes the atomic part of the Hamiltonian, and we have chosen the energy of the $|0\rangle$ state to be zero. Since the atom is moving in a harmonic potential,

and therefore the motional energy is given by

$$H_0^{(M)} = \hbar\nu \left(a^\dagger a + \frac{1}{2} \right) \quad (8.36)$$

where the (M) superscript denotes the motional component of the Hamiltonian. Our total non-interacting part of the Hamiltonian is then $H_0 = H_0^{(A)} + H_0^{(M)}$.

Now let's consider the interaction Hamiltonian H_I . From Sec. 7.1.3, we know that

$$H_I = -q\vec{E} \cdot \hat{\mathbf{r}} \quad (8.37)$$

In this case we are applying two electric fields, one of which drives the $|0\rangle \rightarrow |2\rangle$ transition which we label as the "probe" (p subscript) field and which drives the $|1\rangle \rightarrow |2\rangle$ transition which we name the "coupling" (c subscript) field. H_I therefore is made up of two terms, which we label as $H_I^{(p)}$ and $H_I^{(c)}$, with $H_I = H_I^{(p)} + H_I^{(c)}$. For $H_I^{(p)}$, an applied electric field $E_p(x, t) = E_0^{(p)} \hat{\epsilon}_p \cos(\omega_p t - \vec{k}_p \cdot \vec{x} + \phi_p)$, and an atomic dipole moment $\hat{\mathbf{r}}_{02} = \hat{r}(d_{02}|0\rangle\langle 2| + d_{02}^*|2\rangle\langle 0|)$, we get

$$\begin{aligned} H_I^{(p)} &= -qE_0^{(p)} \hat{\epsilon}_p \cos(\omega_p t - k_p x + \phi_p) \hat{r} (d_{02}|0\rangle\langle 2| + d_{02}^*|2\rangle\langle 0|) \\ &= (\hbar\Omega_p|0\rangle\langle 2| + \hbar\Omega_p^*|2\rangle\langle 0|) \cos(\omega_p t - \vec{k}_p \cdot \vec{x} + \phi_p) \end{aligned} \quad (8.38)$$

where we have defined $\hbar\Omega_p = -qE_0^{(p)} d_{02}(\hat{\epsilon}_p \cdot \hat{\mathbf{r}})$. Similarly, for the coupling beam we get

$$H_I^{(c)} = (\hbar\Omega_c|1\rangle\langle 2| + \hbar\Omega_c^*|2\rangle\langle 1|) \cos(\omega_c t - \vec{k}_c \cdot \vec{x} + \phi_c) \quad (8.39)$$

With all the Hamiltonians suitably defined, we now define a frame which rotates according to

$$H_D = \hbar(\omega_p - \omega_c) |1\rangle\langle 1| + \hbar\omega_p |2\rangle\langle 2| \quad (8.40)$$

and therefore

$$\begin{aligned} H_0 - H_D &= \hbar[\omega_{10} - (\omega_p - \omega_c)] |1\rangle\langle 1| + \hbar(\omega_{20} - \omega_p) |2\rangle\langle 2| + \hbar\nu \left(a^\dagger a + \frac{1}{2} \right) \\ &= -\hbar\delta |1\rangle\langle 1| - \hbar\Delta |2\rangle\langle 2| + \hbar\nu \left(a^\dagger a + \frac{1}{2} \right) \end{aligned} \quad (8.41)$$

where Δ is commonly referred to as the *single-photon detuning* (or sometimes just the *detuning*), and δ is the *two-photon detuning*.

For the interaction Hamiltonian we will need the exponential of H_D :

$$e^{\pm iH_D t/\hbar} = |0\rangle\langle 0| + e^{\pm i(\omega_p - \omega_c)t} |1\rangle\langle 1| + e^{\pm i\omega_p t} |2\rangle\langle 2| \quad (8.42)$$

and therefore we get for the probe and coupling beam interactions

$$\tilde{H}_I^{(p)} = (\hbar\Omega_p e^{-i\omega_p t} |0\rangle\langle 2| + \hbar\Omega_p^* e^{i\omega_p t} |2\rangle\langle 0|) \cos(\omega_p t - \vec{k}_p \cdot \vec{x} + \phi_p) \quad (8.43)$$

$$\tilde{H}_I^{(c)} = (\hbar\Omega_c e^{-i\omega_c t} |1\rangle\langle 2| + \hbar\Omega_c^* e^{i\omega_c t} |2\rangle\langle 1|) \cos(\omega_c t - \vec{k}_c \cdot \vec{x} + \phi_c) \quad (8.44)$$

At this point it is instructive to consider the frequency scales of typical atomic systems. The atomic energy levels are usually in the visible part of the electromagnetic spectrum. For light with a wavelength around 600 nm the frequency of the light is $\omega \approx 2\pi \cdot 500$ THz. However, the linewidth of dipole-allowed excited states is typically around $\Gamma \approx 2\pi \cdot 10$ MHz, and therefore we typically (but not always, for example while driving Raman transitions) are trying to drive near-resonant processes, and therefore the single photon detunings $\Delta \approx \Gamma$. The two-photon detuning δ is often used to resonantly drive a two-photon transition, and is typically very small $\delta \leq 2\pi \cdot 10$ kHz. The motional frequency depends on the trap depth, but for a typical ion trap $\nu \approx 2\pi \cdot 1$ MHz. In summary, we typically have

$$\omega_{02}, \omega_{12}, \omega_{01} \gg \Delta, \Gamma > \nu > \delta \quad (8.45)$$

however this is not always true. For example, in the case where $|0\rangle$ and $|1\rangle$ are states in the same hyperfine ground level of an atom, and we want to drive two-photon Raman transitions between them, we will likely have:

$$\omega_{02}, \omega_{12} \gg \Delta \gg \omega_{01} \gg \Gamma > \nu > \delta \quad (8.46)$$

The point of this consideration is that optical frequencies are very large. If we expand the cosine terms in Eq. 8.43 and Eq. 8.44 we will get terms similar to:

$$e^{\pm i\omega t} \cos(\omega t) = \frac{1}{2} (1 + e^{\pm i2\omega t}) \quad (8.47)$$

The complex exponential term here oscillates very quickly compared to any interesting dynamics arising from the interaction. These terms will therefore average to 0 over any appreciable timescale, and we will be left only with the DC term. Another way to look at this is to consider that these terms appear in the Hamiltonian, which we have to integrate over to solve the $i\hbar\dot{\rho} = H\rho - \rho H$ von Neumann equation. Since $\int e^{i2\omega t} \propto 1/\omega$, their contribution to the overall integral will be negligible, since ω is very large. Ignoring these rapidly oscillating terms is known as the *rotating wave approximation* (RWA), and is very commonly used in atomic physics.

Eq. 8.43 and Eq. 8.44, using the RWA, then become

$$\tilde{H}_I^{(p)} = \frac{\hbar}{2} \left(\Omega_p e^{-i(\vec{k}_p \cdot \vec{x} - \phi_p)} |0\rangle \langle 2| + \Omega_p^* e^{i(\vec{k}_p \cdot \vec{x} - \phi_p)} |2\rangle \langle 0| \right) \quad (8.48)$$

$$\tilde{H}_I^{(c)} = \frac{\hbar}{2} \left(\Omega_c e^{-i(\vec{k}_c \cdot \vec{x} - \phi_c)} |1\rangle \langle 2| + \Omega_c^* e^{i(\vec{k}_c \cdot \vec{x} - \phi_c)} |2\rangle \langle 1| \right) \quad (8.49)$$

The next thing to consider is the atom's motion in the harmonic potential. Recall that for a harmonic oscillator, we know that $\vec{x} = \sqrt{\hbar/2m\nu} \hat{\epsilon}_x (a^\dagger + a)$, and therefore

$$\vec{k}_p \cdot \vec{x} = \sqrt{\frac{\hbar}{2m\nu}} k_p (\hat{k}_p \cdot \hat{\epsilon}_x) (a^\dagger + a) \equiv \eta_p (a^\dagger + a) \quad (8.50)$$

$$\vec{k}_c \cdot \vec{x} = \sqrt{\frac{\hbar}{2m\nu}} k_c (\hat{k}_c \cdot \hat{\epsilon}_x) (a^\dagger + a) \equiv \eta_c (a^\dagger + a) \quad (8.51)$$

where η , as defined in the above equation, is known as the Lamb-Dicke parameter. It is a measure of how well localized the atom is in the harmonic potential. Note that it relies on the overlap (dot product) between the electric field propagation direction and the direction of motion in the trap. When a system with a mean number of motional quanta n satisfies the condition $\sqrt{n}\eta < 1$, it is said to be in the *Lamb-Dicke regime*, and it is thus possible to Taylor expand the exponential such that

$$e^{i\eta(a^\dagger+a)} = 1 + i\eta(a^\dagger + a) + \mathcal{O}(\eta^2) \quad (8.52)$$

which often greatly simplifies analytic treatments. If this expansion is made, the zero-order ($\propto \eta^0$) term is often referred to as the *carrier transition*, and the first-order ($\propto \eta^1$) terms are called the *motional sidebands*.

The final form of our interaction Hamiltonian is therefore

$$\begin{aligned} \tilde{H}_I &= \tilde{H}_I^{(p)} + \tilde{H}_I^{(c)} \\ &= \frac{\hbar}{2} \left(\Omega_p e^{-i\eta_p(a^\dagger+a)} |0\rangle \langle 2| + \Omega_p^* e^{i\eta_p(a^\dagger+a)} |2\rangle \langle 0| \right) \\ &\quad + \frac{\hbar}{2} \left(\Omega_c e^{-i\eta_c(a^\dagger+a)} |1\rangle \langle 2| + \Omega_c^* e^{i\eta_c(a^\dagger+a)} |2\rangle \langle 1| \right) \end{aligned} \quad (8.53)$$

where we have absorbed the electric field phase into the Rabi frequency $\Omega_p \rightarrow \Omega_p e^{i\phi_p}$ and $\Omega_c \rightarrow \Omega_c e^{i\phi_c}$ for simplicity.

The last piece we need to describe this system is the Lindblad, or jump, term as shown in Eq. 8.31. The excited state $|2\rangle$ can decay to either of the two ground states $|0\rangle$ or $|1\rangle$, with rates given by γ_{20} and γ_{21} , resp. The jump operators in the Schrödinger picture are given by $L_{20} = \sqrt{\gamma_{20}} |0\rangle \langle 2|$ and $L_{21} = \sqrt{\gamma_{21}} |1\rangle \langle 2|$. In the rotating frame we see that

$$\tilde{L}_{20} = \sqrt{\gamma_{20}} e^{-i\omega_p t} L_{20} \quad (8.54)$$

$$\tilde{L}_{21} = \sqrt{\gamma_{21}} e^{-i\omega_c t} L_{21} \quad (8.55)$$

Clearly $\tilde{L}^\dagger \tilde{L} = L^\dagger L$, and $\tilde{L} \rho \tilde{L}^\dagger = L \rho L^\dagger$ for both jump operators. Therefore the spontaneous emission part of the master equation does not change, and we have:

$$\mathcal{D}[\tilde{L}_i](\tilde{\rho}) = \mathcal{D}[L_i](\tilde{\rho}). \quad (8.56)$$

Note also that $L_{20}^\dagger L_{20} = \gamma_{20} |2\rangle \langle 2|$ and $L_{21}^\dagger L_{21} = \gamma_{21} |2\rangle \langle 2|$

$$\begin{aligned} \sum_{i=0}^1 \mathcal{D}[L_{2i}](\tilde{\rho}) &= (\gamma_{20} |0\rangle \langle 0| + \gamma_{21} |1\rangle \langle 1|) \langle 2|\tilde{\rho}|2\rangle - \frac{\gamma_{20} + \gamma_{21}}{2} (|2\rangle \langle 2| \tilde{\rho} + \tilde{\rho} |2\rangle \langle 2|) \\ &= (\gamma_{20} |0\rangle \langle 0| + \gamma_{21} |1\rangle \langle 1|) \tilde{\rho}_{22} - \frac{\gamma_{20} + \gamma_{21}}{2} (|2\rangle \langle 2| \tilde{\rho} + \tilde{\rho} |2\rangle \langle 2|) \end{aligned} \quad (8.57)$$

The first term can be identified as the ground state populations increasing when a spontaneous decay takes the atom's state from $|2\rangle$ to one of the ground states. The other two terms govern the decay of the excited state and the coherences which couple the excited state to the ground states.

Between Eq. 8.33, Eq. 8.41, Eq. 8.53, and Eq. 8.57 we now have all the necessary components to write down the full master equation governing the time evolution of $\tilde{\rho}$.

8.3.3 Raman Transitions

We consider now a simple case where we ignore the motional component. We can write down the coupled differential equations generated by the master equation in matrix form to get the set of coupled differential equations for this problem. We define $\tilde{\rho}_{ij} = \langle i|\tilde{\rho}|j\rangle$ and obtain the following equations of motion for the populations:

$$\begin{aligned}\dot{\tilde{\rho}}_{00} &= -\frac{i}{2} (\Omega_p \tilde{\rho}_{20} - \Omega_p^* \tilde{\rho}_{02}) + \gamma_{20} \tilde{\rho}_{22} \\ \dot{\tilde{\rho}}_{11} &= -\frac{i}{2} (\Omega_c \tilde{\rho}_{21} - \Omega_c^* \tilde{\rho}_{12}) + \gamma_{21} \tilde{\rho}_{22} \\ \dot{\tilde{\rho}}_{22} &= -\dot{\tilde{\rho}}_{00} - \dot{\tilde{\rho}}_{11}\end{aligned}\tag{8.58}$$

and for the coherences:

$$\begin{aligned}\dot{\tilde{\rho}}_{01} = \dot{\tilde{\rho}}_{10}^* &= -i\delta \tilde{\rho}_{01} - \frac{i}{2} (\Omega_p \tilde{\rho}_{21} - \Omega_c^* \tilde{\rho}_{02}) \\ \dot{\tilde{\rho}}_{02} = \dot{\tilde{\rho}}_{20}^* &= -i\Delta \tilde{\rho}_{02} - \frac{i}{2} [\Omega_p (\tilde{\rho}_{22} - \tilde{\rho}_{00}) - \Omega_c \tilde{\rho}_{01}] - \frac{\gamma_{20} + \gamma_{21}}{2} \tilde{\rho}_{02} \\ \dot{\tilde{\rho}}_{12} = \dot{\tilde{\rho}}_{21}^* &= -i(\Delta - \delta) \tilde{\rho}_{12} - \frac{i}{2} [\Omega_c (\tilde{\rho}_{22} - \tilde{\rho}_{11}) - \Omega_p \tilde{\rho}_{10}] - \frac{\gamma_{20} + \gamma_{21}}{2} \tilde{\rho}_{12}\end{aligned}\tag{8.59}$$

Now we make the assumption that both beams are very far detuned from resonance, and therefore $\Delta \gg \Omega_c, \Omega_p, \gamma_{20}, \gamma_{21}, \delta$. The population in the $|2\rangle$ state will stay very small, due to spontaneous emission depumping the state quickly. In addition, the coherences involving the $|2\rangle$ state will change very slowly. The equations for $\dot{\tilde{\rho}}_{02}$ and $\dot{\tilde{\rho}}_{12}$ are solved for their steady state solutions:

$$\begin{aligned}\dot{\tilde{\rho}}_{02} = \dot{\tilde{\rho}}_{20}^* = 0 &\implies -i\Delta \tilde{\rho}_{02} + \frac{i}{2} [\Omega_p \tilde{\rho}_{00} + \Omega_c \tilde{\rho}_{01}] = 0 \\ &\implies \tilde{\rho}_{02} = \tilde{\rho}_{20}^* = \frac{\Omega_p}{2\Delta} \tilde{\rho}_{00} + \frac{\Omega_c}{2\Delta} \tilde{\rho}_{01}\end{aligned}\tag{8.60}$$

$$\begin{aligned}\dot{\tilde{\rho}}_{12} = \dot{\tilde{\rho}}_{21}^* = 0 &\implies -i(\Delta - \delta) \tilde{\rho}_{12} + \frac{i}{2} [\Omega_c \tilde{\rho}_{11} + \Omega_p \tilde{\rho}_{10}] = 0 \\ &\implies \tilde{\rho}_{12} = \tilde{\rho}_{21}^* = \frac{\Omega_c}{2\Delta} \tilde{\rho}_{11} + \frac{\Omega_p}{2\Delta} \tilde{\rho}_{10}\end{aligned}\tag{8.61}$$

Using these equations we can write a set of coupled differential equations which depend only on the $|0\rangle$ and $|1\rangle$ states:

$$\begin{aligned}\dot{\tilde{\rho}}_{00} &= i \frac{\Omega_p^* \Omega_c}{4\Delta} \tilde{\rho}_{01} - i \frac{\Omega_p \Omega_c^*}{4\Delta} \tilde{\rho}_{10} \\ \dot{\tilde{\rho}}_{11} &= -i \frac{\Omega_p^* \Omega_c}{4\Delta} \tilde{\rho}_{01} + i \frac{\Omega_p \Omega_c^*}{4\Delta} \tilde{\rho}_{10} = -\dot{\tilde{\rho}}_{00} \\ \dot{\tilde{\rho}}_{01} &= -i \left(\delta + \frac{|\Omega_p|^2 - |\Omega_c|^2}{4\Delta} \right) \tilde{\rho}_{01} - i \frac{\Omega_p \Omega_c^*}{4\Delta} (\tilde{\rho}_{11} - \tilde{\rho}_{00})\end{aligned}\tag{8.62}$$

By assuming slowly varying coherences and a vanishing excited state population, we have effectively eliminated the excited state from this three-level system, and turned it into a

two-level system. This method is (not surprisingly) called *adiabatic elimination*. The result in Eq. 8.62 can be written in von Neumann form as follows:

$$\begin{aligned}\dot{\tilde{\rho}}_{\text{eff}} &= -\frac{i}{\hbar}[H_{\text{eff}}, \tilde{\rho}_{\text{eff}}] \\ H_{\text{eff}} &= \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega \\ \Omega^* & -2\delta' \end{pmatrix} \\ \rho_{\text{eff}} &= \begin{pmatrix} \tilde{\rho}_{00} & \tilde{\rho}_{01} \\ \tilde{\rho}_{10} & \tilde{\rho}_{11} \end{pmatrix}\end{aligned}\tag{8.63}$$

Here we have defined the effective Rabi frequency Ω and effective detuning δ' :

$$\Omega = \frac{\Omega_p \Omega_c^*}{2\Delta} \quad \delta' = \delta + \frac{|\Omega_p|^2 - |\Omega_c|^2}{4\Delta}\tag{8.64}$$

The modification to the detuning is due to the differential AC Stark shift induced on the two states by the applied electric fields. The form of Eq. 8.63 is exactly analogous to a driven two-level system without spontaneous emission, even though there is nothing which directly drives a transition between the $|0\rangle$ and $|1\rangle$ states! Not only that, but the process is coherent, meaning that phase relationships between the states, and therefore the coherence terms in the density matrix, are preserved.

It's worth looking at the effective Rabi frequency more closely. We recall that

$$\Omega_p \Omega_c^* \propto E_0^{(p)} E_0^{(c)*} e^{i(\phi_p - \phi_c)}\tag{8.65}$$

This dependence should make it clear that, in order to drive this transition effectively, you need intense power in your laser beams, as well as good intensity stability. Most critically, however, is that the relative phase between the laser beams needs to be stable. Due to the short wavelength, this means that laser beam path length differences must be controlled to $\ll \lambda \approx 500$ nm.

Also, note that while the coherent two-photon transition rate scales as $1/\Delta$, the incoherent scattering rate of a dipole transition scales as $1/\Delta^2$. This means that to drive clean two-photon transitions we should use large intensity laser fields and large detunings in order to ensure the two-photon rate dominates.