Chapter 10

Two-Photon Transitions

10.1 Raman Transitions

Raman transition is where two optical fields are used to drive a transition, where one photon is "virtually" absorbed and another is virtually emitted through an excited state. The absorption or emission is virtual since the actual laser frequencies are detuned substantially from the transition frequency (well beyond the natural linewidth of the transition), and the probability of an actual photon being absorbed and spontaneously emitted is very low. The virtual absorption and emission is driven by strong fields through a stimulated process, so it is sometimes called *stimulated Raman transition*. Also, there are two photons involved, one from each beam, and therefore it is an example of a "two-photon" transition process.

The Raman transition can also be understood using the model described in Fig. 9.1, where the detuning Δ to the excited state is maintained to be significant (larger than the excited state linewidth γ), and the differential detuning δ is small. In this analysis, we would also add the phase of the two fields φ_i for the laser field $\mathcal{E}_i$ ($i = 0,\, 1$). The interaction Hamiltonian (Eq. 9.2) for the Jaynes-Cummings model is slightly modified to include the phase of the optical fields

$$\hat{H}'(t) = -\frac{\hbar}{2}\left[\Omega_0 e^{-i(\omega_0 t+\varphi_0)} \left|2\right\rangle \left\langle 0\right| + \Omega_1 e^{-i(\omega_1 t+\varphi_1)} \left|2\right\rangle \left\langle 1\right|\right] + h.c. \tag{10.1}$$

We can follow the same procedure in Section 9.1.2 to reach the differential equation for the $c_i(t)$ coeffcients

$$\begin{aligned}
\dot{c}_0(t) + \frac{iE_0}{\hbar}c_0(t) &= \frac{i}{2}\Omega_0 e^{i[(\omega_{20}+\Delta)t+\varphi_0]}c_2(t), && (10.2)\\
\dot{c}_1(t) + \frac{iE_1}{\hbar}c_1(t) &= \frac{i}{2}\Omega_1 e^{i[(\omega_{21}+\Delta-\delta)t+\varphi_1]}c_2(t), && (10.3)\\
\dot{c}_2(t) + \frac{iE_2}{\hbar}c_2(t) &= \frac{i}{2}\left[\Omega_0 e^{-i[(\omega_{20}+\Delta)t+\varphi_0]}c_0(t) + \Omega_1 e^{-i[(\omega_{21}+\Delta-\delta)t+\varphi_1]}c_1(t)\right]. && (10.4)
\end{aligned}$$

where we used the relationship between frequencies $\omega_0 = \omega_{20} + \Delta$ and $\omega_1 = \omega_{21} + \Delta - \delta$. We make the transformation $c_i'(t) = c_i(t)e^{iE_i t/\hbar}$ for the two ground states ($i = 0,\, 1$), just as in Section 9.1.2. For the excited state, we make the transformation $c''_2(t) = c_2(t)e^{i(E_2/\hbar+\Delta)t}$. The differential equations reduce to

$$\dot{c}'_0(t) = \frac{i}{2}\Omega_0 e^{i\varphi_0} c''_2(t), \tag{10.5}$$

$$\dot{c}'_1(t) = \frac{i}{2}\Omega_1 e^{-i(\delta t-\varphi_1)} c''_2(t), \tag{10.6}$$

$$\dot{c}''_2(t) - i\Delta c''_2(t) = \frac{i}{2}\left[\Omega_0 e^{-i\varphi_0} c'_0(t) + \Omega_1 e^{i(\delta t-\varphi_1)} c'_1(t)\right]. \tag{10.7}$$

Since the detuning Δ is large compared to the linewidth of the level $|2\rangle$ defined by its spontaneous emission rate, the population of this state is expected to be very small if the Rabi frequencies induced by the fields Ω_0 and Ω_1 are also small compared to the detuning. Under these conditions, the time derivative of the coefficient $c''_2(t)$ – the first term on the LHS of Eq. 10.7 – is small compared to the second term, $|\dot{c}''_2(t)| \ll |\Delta c''_2(t)|$. So, Eq. 10.7 can be approximated as

$$c''_2(t) \simeq -\frac{1}{2\Delta}\left[\Omega_0 e^{-i\varphi_0} c'_0(t) + \Omega_1 e^{i(\delta t-\varphi_1)} c'_1(t)\right]. \tag{10.8}$$

Using this equation, Eqs. 10.5 and 10.6 can be simplified to

$$\dot{c}'_0(t) = -\frac{i}{4\Delta}\left[\Omega_0^2 c'_0(t) + \Omega_0\Omega_1 e^{i(\delta t+\Delta\varphi)} c'_1(t)\right], \tag{10.9}$$

$$\dot{c}'_1(t) = -\frac{i}{4\Delta}\left[\Omega_0\Omega_1 e^{-i(\delta t+\Delta\varphi)} c'_0(t) + \Omega_1^2 c'_1(t)\right], \tag{10.10}$$

where we defined $\Delta\varphi \equiv \varphi_0 - \varphi_1$. This pair of equations now only involve the time-dependent coefficient of the two ground states, and the population of the excited state is ignored. This process of eliminating the excited state population in the limit of large detuning is called *adiabatic elimination* process.

From the RHS of Eqs. 10.9 and 10.10, we note that there are terms that correspond to simple shift of frequency. We define $\delta_i = \Omega_i^2/4\Delta$ $(i = 0, 1)$, called the *light shift* (also known as the AC Stark shift), and shift the resonance frequency by defining $c''_i(t) \equiv c'_i(t)e^{i\delta_i t}$, to obtain the final set of equations

$$\dot{c}''_0(t) = -\frac{i\Omega}{2} e^{i[(\delta+\Delta\delta)t+\Delta\varphi]} c''_1(t), \tag{10.11}$$

$$\dot{c}''_1(t) = -\frac{i\Omega}{2} e^{-i[(\delta+\Delta\delta)t+\Delta\varphi]} c''_0(t), \tag{10.12}$$

where $\Delta\delta \equiv \delta_0 - \delta_1$ is the *differential light shift*, and $\Omega \equiv \Omega_0\Omega_1/(2\Delta)$ is the *effective Rabi frequency* for the Raman transition. In the limit of zero Raman detuning ($\delta = 0$), this equation reduces to the simple Rabi flopping of two-level system described in Section 9.2.2 (Eqs. 9.28 and 9.29). In the presence of detuning, the time-evolution of the system is identical to the magnetic resonance considered in Section 3.2.3 (direct comparison to Eqs. 3.40 and 3.41).

A quick summary of the stimulated Raman transition is as follows:

- With large detuning $\Delta \gg \gamma$, Ω_0, Ω_1, we can adiabatically eliminate the population of the excited state, and reach an effective two-level system dynamics for the two ground states driven by an effective single field.
- The ground state energy levels are shifted by an amount $\delta_i = \Omega_i^2/(4\Delta)$ (for $i = 0, 1$), known as the light shift (or the AC Stark shift).
- The Rabi frequency for the two states is given by $\Omega = \Omega_0\Omega_1/(2\Delta)$, with a detuning of δ plus the differential light shift of $\Delta\delta = \delta_0 - \delta_1$.
- The "phase" of effective field that is driving the Rabi oscillations is defined by the difference of the local phase of the two optical fields, $\Delta\varphi = \varphi_0 - \varphi_1$.
- The Raman transition involves a "virtual" absorption of a photon from the laser beam $\mathcal{E}_0$ and a "virtual" emission of a photon into the laser beam $\mathcal{E}_1$. The recoil of these photon absorption and emission events will lead to corresponding momentum kicks on the center-of-mass motion of the atom.

There are a couple of interesting consequences depending on the geometry of the beams that drive the Raman transitions.

- If the two laser beams that are driving the Raman transitions are propagating along an identical beam path (co-propagating), the optical phases φ_0 and φ_1 tend to experience exactly the same beam path. In this case, $\Delta\delta = 0$, and the Raman transition tends to be stable over the optical beam path.
- Under the co-propagating condition, the absorption of a photon from the first beam and the emission of a photon into the second beam exerts momentum kicks to the atom that cancels each other out. So, in co-propagating beam geometry, the atom undergoing a Raman transition does not see any significant change in its momentum.
- If the two laser beams that are driving the Raman transitions are not propagating along the same path (non-co-propagating), the phase difference between the two beams at the location of the atom is sensitive to the optical beam path of both beams.
- When the two beams are not co-propagating, the net momentum kick on the atom will be the difference between the momentum exerted on the atom due to the absorption of the first photon and that due to the emission of the second photon, namely $\Delta\vec{p} = \hbar(\vec{k}_0 - \vec{k}_1) = \hbar\Delta\vec{k}$, where $\vec{k}_0$ and $\vec{k}_1$ are the wavevectors of the laser beam $\mathcal{E}_0$ and $\mathcal{E}_1$, respectively.
- The differential light shift is dependent on the intensity of each beam hitting the atom, and can be compensated with the detuning of the optical frequency difference δ between the two beams.

10.2 Two-Photon Excitation

Another example of a two-photon transition process is where an excitation to a higher energy level is achieved using two laser beams, where the energy arising from absorption (or

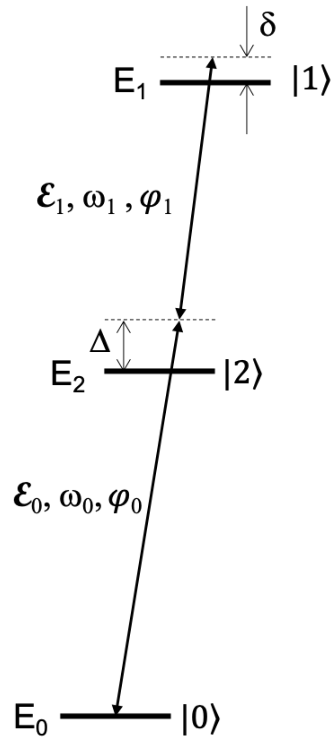

Figure 10.1: Schematic of a 3-level atomic system driven by two fields, for a two-photon excitation/de-excitation. The transition is between two energy levels $|0\rangle$ and $|1\rangle$ using an intermediate state with energy level $|2\rangle$, and two laser fields $\mathcal{E}_0$ and $\mathcal{E}_1$, with frequencies ω_1, ω_2 and phases φ_0, φ_1, driving the transition between the state $|0\rangle$ and $|1\rangle$ and the excited state $|2\rangle$, respectively. The frequency ω_0 is detuned from the $|0\rangle \leftrightarrow |2\rangle$ transition by an amount Δ, and the difference between the sum of the two laser frequencies can be detuned from the energy difference $E_1 - E_0$ of the two states $|0\rangle$ and $|1\rangle$ by another amount δ.

emission) of two photons, one from each beam, makes up for the total energy difference of the twp energy levels involved, as shown in Fig. 10.1.

The energy relationship in this case, compared to the Raman transition case, is given by

$$\omega_0 = \omega_{20} + \Delta, \;\; \omega_1 = \omega_{12} + \delta - \Delta. \tag{10.13}$$

The interaction Hamiltonian describing the Jaynes-Cummings model in Eq. 10.1 is now replaced with

$$\hat{H}'(t) = -\frac{\hbar}{2}\left[\Omega_0 e^{-i(\omega_0 t + \varphi_0)} |2\rangle \langle 0| + \Omega_1 e^{-i(\omega_1 t + \varphi_1)} |1\rangle \langle 2|\right] + h.c. \tag{10.14}$$

The resulting differential equations for the coefficients for this case is given by

$$\dot{c}_0(t) + \frac{iE_0}{\hbar}c_0(t) = \frac{i}{2}\Omega_0 e^{i[(\omega_{20}+\Delta)t+\varphi_0]}c_2(t), \quad (10.15)$$

$$\dot{c}_1(t) + \frac{iE_1}{\hbar}c_1(t) = \frac{i}{2}\Omega_1 e^{-i[(\omega_{12}-\Delta+\delta)t+\varphi_1]}c_2(t), \quad (10.16)$$

$$\dot{c}_2(t) + \frac{iE_2}{\hbar}c_2(t) = \frac{i}{2}\left[\Omega_0 e^{-i[(\omega_{20}+\Delta)t+\varphi_0]}c_0(t) + \Omega_1 e^{i[(\omega_{12}-\Delta+\delta)t+\varphi_1]}c_1(t)\right]. \quad (10.17)$$

Upon adiabatic elimination of the population in $|2\rangle$ state and accounting for the light shifts for the $|0\rangle$ and $|1\rangle$ levels of $\delta_i = \Omega_i^2/(4\Delta)$ (for $i = 0, 1$) similar to the Raman case, we end up with the differential equations for $c_i''(t) = c_i(t)e^{i(E_i/\hbar+\delta_i)t}$,

$$\dot{c}_0''(t) = -\frac{i\Omega}{2}e^{i[(\delta+\Delta\delta)t+\varphi_0+\varphi_1]}c_1''(t), \quad (10.18)$$

$$\dot{c}_1''(t) = -\frac{i\Omega}{2}e^{-i[(\delta+\Delta\delta)t+\varphi_0+\varphi_1]}c_0''(t), \quad (10.19)$$

where $\Delta\delta = \delta_0 - \delta_1$ is the differential light shift, and $\Omega = \Omega_0\Omega_1/(2\Delta)$ is the effective Rabi frequency for the transition between the two states $|0\rangle$ and $|1\rangle$.

A few things to note in the two-photon excitation/de-excitation case

- With large detuning $\Delta \gg \gamma_2, \Omega_0, \Omega_1$ (where γ_2 is the linewidth of the intermediate state $|2\rangle$ determined by its spontaneous emission), we can adiabatically eliminate the population of the state $|2\rangle$, and reach an effective two-level system dynamics for the ground state $|0\rangle$ and the excited state $|1\rangle$ driven by an effective single field.
- The energy levels of the ground state $|0\rangle$ and the excited state $|1\rangle$ are shifted by amounts $\delta_i = \Omega_i^2/(4\Delta)$ (for $i = 0, 1$), known as the light shift (or the AC Stark shift).
- The Rabi frequency for the two states is given by $\Omega = \Omega_0\Omega_1/(2\Delta)$, with a detuning of δ plus the differential light shift of $\Delta\delta = \delta_0 - \delta_1$.
- The "phase" of effective field that is driving the Rabi oscillations is defined by the sum of the local phases of the two optical fields, $\varphi_0 + \varphi_1$.
- The two-photon transition involves "virtual" absorption of a photon from each of the laser beams $\mathcal{E}_0$ and $\mathcal{E}_1$. The recoil of these photon absorption events will lead to corresponding momentum kicks on the center-of-mass motion of the atom.

The consequences depending on the geometry of the beams that drive the two-photon transitions include

- If the two laser beams that are driving the two-photon transition are propagating along an identical beam path (co-propagating), the optical phases φ_0 and φ_1 will add up, and the phase of the two-photon transition is sensitive to the optical beam path.
- Under the co-propagating condition, the absorption of the two photons exerts momentum kicks to the atom that is in the same direction. So, in co-propagating beam geometry, the atom undergoing a two-photon transition will see full recoil on its momentum from both photons.

- If the two laser beams that are driving the Raman transitions are not propagating along the same path (non-co-propagating), the sum of the phase of the two beams at the location of the atom is sensitive to the optical beam path of both beams.
- When the two beams are not co-propagating, the net momentum kick on the atom will be the sum of the the momentum exerted on the atom due to the absorption of the two photons, namely $\vec{p} = \hbar(\vec{k}_0 + \vec{k}_1) = \hbar\vec{k}$, where $\vec{k}_0$ and $\vec{k}_1$ are the wavevectors of the laser beam $\mathcal{E}_0$ and $\mathcal{E}_1$, respectively, and $\vec{k} = \vec{k}_0 + \vec{k}_1$. When the two beams are counter-propagating, the momenta of the two photons tend to cancel. However, unlike the Raman case where the difference in the photon energy for the two photons are very small, the frequency (and therefore the wavevector) for the photons involved in a two-photon transition can be quite different, so the momentum cancellation is usually not guaranteed even in the counter-propagating case.
- The differential light shift is dependent on the intensity of each beam hitting the atom, and can be compensated with the detuning of the optical frequency difference δ between the two beams.

Two-photon excitation mechanisms are often used to excite a ground-state atom to a highly excited state with large orbitals, as the energy scale for those transitions tend to be in the ultraviolet region. These highly excited atoms are called Rydberg atoms, and can feature very large dipole moment due to the orbital angular momentum of the electron.