Solution Set 10 – Two-Photon Processes

Problem 1. Spontaneous Two-Photon Emission (10 pts)

(a) The Göppert-Mayer formula

▷ Find the excitation rate $\Gamma_{ab}(\omega_1)$ when one beam has frequency ω_1. Your expression should include a resonance condition which constrains ω_2.

Beginning with the first equation of the assignment, we find the probability of being in excited state $|b\rangle$.

$$\left|a_b^{[2]}\right|^2 = \left(\frac{1}{4\hbar^2}\right)^2 \left|\sum_f \left\{\frac{H_{bf,2}H_{fa,1}}{\omega_1 - \omega_{fa}} + \frac{H_{bf,1}H_{fa,2}}{\omega_2 - \omega_{fa}}\right\}\right|^2 \frac{\sin^2\left(\left(\omega_{ba} - \omega_1 - \omega_2\right)t/2\right)}{\left(\left(\omega_{ba} - \omega_1 - \omega_2\right)t/2\right)^2} \quad (1)$$

Using arguments presented in class, we replace

$$\frac{\sin^2\left(\left(\omega_{ba} - \omega_1 - \omega_2\right)t/2\right)}{\left(\left(\omega_{ba} - \omega_1 - \omega_2\right)t/2\right)^2} \rightarrow 2\pi t\delta\left(w_{ba} - w_1 - w_2\right). \quad (2)$$

Then, taking time derivative we get,

$$\Gamma_{ab}(\omega_1) = \frac{\pi}{8\hbar^4}\left|\sum_f \left\{\frac{H_{bf,2}H_{fa,1}}{\omega_1 - \omega_{fa}} + \frac{H_{bf,1}H_{fa,2}}{\omega_2 - \omega_{fa}}\right\}\right|^2 \delta(\omega_{ba} - (\omega_1 + \omega_2)) \quad (3)$$

▷ Show that your expression is mathematically equivalent to $\cdots$

Making the substitution,

$$H_{ij,k} = \langle i\,|eE_k\hat{e_k}\cdot\vec{r}|\,j\rangle \quad (4)$$

We get,

$$\Gamma_{ab}(\omega_1) = \frac{\pi E_1^2E_2^2e^4}{8\hbar^4}\left|\sum_f \left\{\frac{\langle b|\mathbf{r}\cdot\hat{\mathbf{e}}_\mathbf{2}|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_\mathbf{1}|a\rangle}{\omega_1 - \omega_{fa}} + \frac{\langle b|\mathbf{r}\cdot\hat{\mathbf{e}}_\mathbf{1}|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_\mathbf{2}|a\rangle}{\omega_2 - \omega_{fa}}\right\}\right|^2 \delta(\omega_{ba} - (\omega_1 + \omega_2)) \quad (5)$$

Finally, using the fact that the delta function forces $\omega_{ba} = \omega_1 + \omega_2$, we can write

$$\omega_1 - \omega_{fa} = \omega_1 - (\omega_{ba} + \omega_{fb}) = -\left(\omega_{fb} + \omega_2\right) \quad (6)$$

$$\omega_2 - \omega_{fa} = -(\omega_{fb} + \omega_1) \tag{7}$$

Using these relations, and using the fact that $\vec{r}$ is hermitian, we arrive at,

$$\Gamma_{ab}(\omega_1) = \frac{\pi E_1^2 E_2^2 e^4}{8\hbar^4} \left| \sum_f \left\{ \frac{\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_1|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_2|b\rangle}{\omega_2 + \omega_{fb}} + \frac{\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_2|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_1|b\rangle}{\omega_1 + \omega_{fb}} \right\} \right|^2 \delta(\omega_{ba} - (\omega_1 + \omega_2)). \tag{8}$$

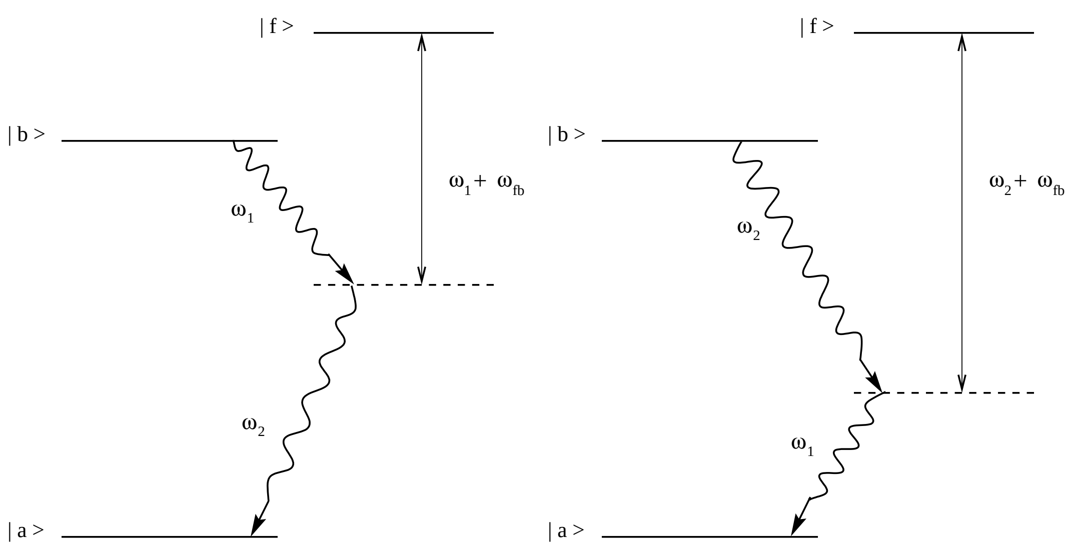
From the principle of detailed balance, we know that

$$\Gamma_{ba}(\omega_1) = \Gamma_{ab}(\omega_1) \tag{9}$$

where $\Gamma_{ba}(\omega_1)$ is the rate of two-photon stimulated emission with one of the photons having frequency ω_1. As noted in the assignment, it can be explicitly shown that $\Gamma_{ba}(\omega_1)$ is equal to the expression in Eq. 8 by applying the appropriate perturbation theory expressions for two-photon emission. One simply swaps a and b and changes the signs of ω_1 and ω_2 everywhere.

▷ Give a physical interpretation for the terms in the sum.

For a two-photon emission process, it is easiest to use Eq. 8 to make an intuitive physical interpretation of the sum. Reading the first term inside the curly brackets from right to left, we essentially have the product of the amplitude for a dipole transition at frequency ω_2 from state $|b\rangle$ to state $|f\rangle$ with the amplitude for a transition at frequency ω_1 from state $|f\rangle$ to state $|a\rangle$. As usual for a second-order process, the term is weighted by the inverse of the detuning from the intermediate state (see diagram below). The second term in the curly brackets represents the other pathway involving intermediate state $|f\rangle$: transition from $|b\rangle$ to $|f\rangle$ by a photon of frequency ω_1 and transition from $|f\rangle$ to $|a\rangle$ via a photon of frequency ω_2. The sum over all possible intermediate states thus gives the total amplitude for transition from $|b\rangle$ to $|a\rangle$ by means of two photons with frequencies ω_1 and ω_2. Note that interference can occur between various terms of the sum. A similar interpretation can also be made for a two-photon absorption process.



▷ Derive the expression for $A(\omega_1)\mathrm{d}\omega_1$.

We now find an expression for the two-photon absorption rate from modes of an ambient quantized electromagnetic field, and this will immediately give us the two-photon spontaneous emission rate. We suppose the field is contained in a box of volume V with the usual periodic boundary conditions. Consider a coherent mode $|\alpha\rangle$. The expectation values of photon number and electric field amplitude are,

$$\langle n\rangle = \langle \alpha \left| a^\dagger a \right| \alpha\rangle = |\alpha|^2 \tag{10}$$

$$\langle E\rangle = \sqrt{\frac{2\pi\hbar\omega}{V}} \langle \alpha \left| a - a^\dagger \right| \alpha\rangle \tag{11}$$

$$\langle E\rangle = \sqrt{\frac{2\pi\hbar\omega}{V}} 2 * Im\left(\alpha\right) \tag{12}$$

Note that $\alpha = re^{-i\omega t}$, so the amplitude of the field is,

$$\langle E_0\rangle = \sqrt{\frac{2\pi\hbar\omega}{V}} 2 * r = \sqrt{\frac{8\pi\hbar\omega\langle n\rangle}{V}} \tag{13}$$

Accordingly, we can rewrite Eq. 8,

$$\Gamma_{ab}(\omega_1) = \frac{\pi e^4}{8\hbar^4}\left(\frac{8\pi n_1\hbar\omega_1}{V}\right)\left(\frac{8\pi n_2\hbar\omega_2}{V}\right)|S|^2\,\delta(\omega_{ba} - (\omega_1 + \omega_2)) \tag{14}$$

$$= \frac{(2\pi)^3 e^4 n_1 n_2 \omega_1\omega_2}{\hbar^2 V^2}|S|^2\,\delta(\omega_{ba} - (\omega_1 + \omega_2)). \tag{15}$$

where S represents the sum over f.

To take into consideration that there are actually a continuum of field modes with frequencies in the vicinity of ω_1 and ω_2, we will now replace $n_1 n_2 \delta(\omega_{ba} - (\omega_1 + \omega_2))$ with $n_1(\omega_1)\, n_2(\omega_2)$, the numbers of photons per unit frequency at these frequencies. It is understood that $\omega_2 = \omega_{ba} - \omega_1$; what we are doing amounts to integrating the rate of Eq. 15 with the density of *occupied* states of the ambient field. The number of photons per unit frequency at frequency ω and with wavevectors pointing into solid angle element $d\Omega$ is

$$n(\omega) = \frac{\overline{n(\omega)} V \omega^2\, d\Omega}{(2\pi)^3 c^3}, \tag{16}$$

where $\overline{n(\omega)}$ is the average number of photons per mode at frequency ω; it could be the Planck distribution, for example. The appropriate density of occupied modes for two-photon absorption is then given by

$$n_1(\omega_1)\, n_2(\omega_2) = \frac{\overline{n(\omega_1)}\,\overline{n(\omega_2)} V^2 \omega_1^2 \omega_2^2\, d\Omega_1\, d\Omega_2}{(2\pi)^6 c^6}. \tag{17}$$

Note that we have not yet accounted for the fact that there are two possible polarizations in each mode.

Putting everything together, we find that that Eq. 15 leads to the following rate for absorption of photons propagating into $d\Omega_1$ and $d\Omega_2$, with one photon having frequency between ω_1 and $\omega_1 + d\omega_1$:

$$\Gamma_{ab}(\omega_1)\, d\omega_1\, d\Omega_1\, d\Omega_2 = \sum_{\text{pol.}} \frac{e^4 \overline{n(\omega_1)}\,\overline{n(\omega_2)}}{(2\pi)^3 \hbar^2 c^6} \omega_1^3 \omega_2^3 |S|^2\, d\omega_1\, d\Omega_1\, d\Omega_2, \tag{18}$$

where the sum is over all possible polarizations eigenstates of the two photons. To go from this stimulated absorption rate to the corresponding spontaneous emission rate from $|b\rangle$ to $|a\rangle$, all we have to do is remove the factor of $\overline{n(\omega_1)}\,\overline{n(\omega_2)}$. The spontaneous emission rate for particular directions of the photons is therefore

$$\Gamma^o_{ba}(\omega_1)\,d\omega_1\,d\Omega_1\,d\Omega_2 = \sum_{\text{pol.}} \frac{e^4}{(2\pi)^3\hbar^2c^6}\omega_1^3\omega_2^3|S|^2\,d\omega_1\,d\Omega_1\,d\Omega_2. \tag{19}$$

To get the total spontaneous emission rate per unit frequency, $A(\omega_1)$, all that remains is to integrate over all 4π steradians of solid angle for both photons.

$$\begin{aligned} A(\omega_1) &= \int d\Omega_1\,d\Omega_2\,\Gamma^o_{ba}(\omega_1) && (20)\\ &= 2\times 2\times 4\pi\times 4\pi\times \left\langle \frac{e^4}{(2\pi)^3\hbar^2c^6}\omega_1^3\omega_2^3|S|^2 \right\rangle_{\text{avg}} && (21)\\ &= \frac{8e^4}{\pi\hbar^2c^6}\omega_1^3\omega_2^3\langle|S|^2\rangle_{\text{avg}} && (22)\end{aligned}$$

Here, the averaging brackets indicate an average over all possible polarizations and directions of propagation for the emitted photons. The factor of 2×2 accounts for the sum over polarizations. This is the formula of Göppert-Mayer.

As explained in the paper of Breit and Teller, it's a simple matter to evaluate the average in the formula for the situation where both $|a\rangle$ and $|b\rangle$ are S-states. In this case, all the possible intermediate states are P-states. Neglecting fine and hyperfine splittings, each P-level is triply degenerate, and it's convenient to use the linearly independent orbitals p_x, p_y, and p_z which have the symmetry properties of the cartesian coordinates x,y, and z, respectively. Then for each P-level, one can group the numerators of the sum S according to

$$\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_1|p_x\rangle\langle p_x|\mathbf{r}\cdot\hat{\mathbf{e}}_2|b\rangle + \langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_1|p_y\rangle\langle p_y|\mathbf{r}\cdot\hat{\mathbf{e}}_2|b\rangle + \langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_1|p_z\rangle\langle p_z|\mathbf{r}\cdot\hat{\mathbf{e}}_2|b\rangle = (\hat{\mathbf{e}}_1\cdot\hat{\mathbf{e}}_2)z_{ap_z}z_{p_za} \tag{23}$$

where we have used the symmetry properties of the orbitals,

$$\langle s|\mathbf{r}\cdot\hat{\mathbf{u}}|p_i\rangle = u_i\langle s|r_i|p_i\rangle = u_i\langle s|z|p_z\rangle. \tag{24}$$

Here, $\hat{\mathbf{u}}$ is any unit vector, i is any cartesian component, and $|s\rangle$ is an S-orbital. With these simplifications, it is evident that the sum can now be expressed

$$S = (\hat{\mathbf{e}}_1\cdot\hat{\mathbf{e}}_2)\sum_f z_{af}z_{fb}\left(\frac{1}{\omega_2+\omega_{fb}}+\frac{1}{\omega_1+\omega_{fb}}\right) \tag{25}$$

where the sum over f is understood to be restricted to those states which transform like z. The problem of taking the average of $|S|^2$ is now reduced to taking the average of $(\hat{\mathbf{e}}_1\cdot\hat{\mathbf{e}}_2)^2$ over all possible directions of $\hat{e}_1$ and $\hat{e}_2$, which turns out to be 1/3. Thus, if fine and hyperfine corrections to the energy levels are neglected, the Göppert-Mayer formula becomes

$$A(\omega_1)d\omega_1 = \frac{8e^4}{3\pi\hbar^2c^6}\omega_1^3\omega_2^3\left|\sum_f z_{af}z_{fb}\left(\frac{1}{\omega_1+\omega_{fb}}+\frac{1}{\omega_2+\omega_{fb}}\right)\right|^2 d\omega_1. \tag{26}$$

(b) $2S$ natural lifetime

▷ Use the total spontaneous decay rate to obtain the lifetime τ of the $2S$ state. Express the $2S$ lifetime in seconds.

To make an estimate of the $2S$ state natural lifetime, we now substitute the Bohr radius a_o for the matrix elements, use the definition of the fine structure constant, $\alpha = e^2/(\hbar c)$, and plug into the equation given in the assignment,

$$\frac{1}{\tau} = A_\tau = \frac{1}{2} \times \frac{8\alpha^2}{3\pi} \frac{a_o^4}{c^4} \int_0^{\omega_{ba}} d\omega_1 \, \omega_1^3 (\omega_{ba} - \omega_1)^3 \left(\frac{1}{\omega_1} - \frac{1}{\omega_{ba} - \omega_1} \right)^2, \tag{27}$$

where we have expressed ω_2 in terms of ω_1. As suggested by the assignment, we are considering only the $2P$ term in the sum over f. Also, we have set $\omega_{fb} = 0$. In other words, we take the $2S$ and $2P$ states to be degenerate. This is an excellent approximation since the $1S$-$2S$ separation is about 2.5×10^{15} Hz, and the fine structure splitting and Lamb shift are only about 10^{10} Hz and 10^9 Hz, respectively.

If we define a dimensionless frequency parameter $x \equiv \omega_1/\omega_{ba}$, then the total spontaneous decay rate becomes

$$A_\tau = \frac{4\alpha^2}{3\pi} \frac{a_o^4 \omega_{ba}^5}{c^4} \int_0^1 dx \, G(x) \tag{28}$$

where

$$G(x) = x^3 (1-x)^3 \left(\frac{1}{x} + \frac{1}{1-x} \right)^2 \tag{29}$$

$$= x(1-x). \tag{30}$$

The simple function $G(x)$ describes the two-photon decay spectrum in this approximation. Since

$$\int_0^1 dx \, G(x) = \int_0^1 dx \, (x - x^2) = \frac{1}{6}, \tag{31}$$

the inverse lifetime is then

$$A_\tau = \frac{2\alpha^2}{9\pi} \frac{a_o^4 \omega_{ba}^5}{c^4} \tag{32}$$

$$\approx \frac{2}{9\pi} \left(\frac{1}{137} \right)^2 \frac{(2\pi)^5 a_o^4 c}{\lambda_{ba}^5} \tag{33}$$

$$\approx 3.2 \text{ s}^{-1}. \tag{34}$$

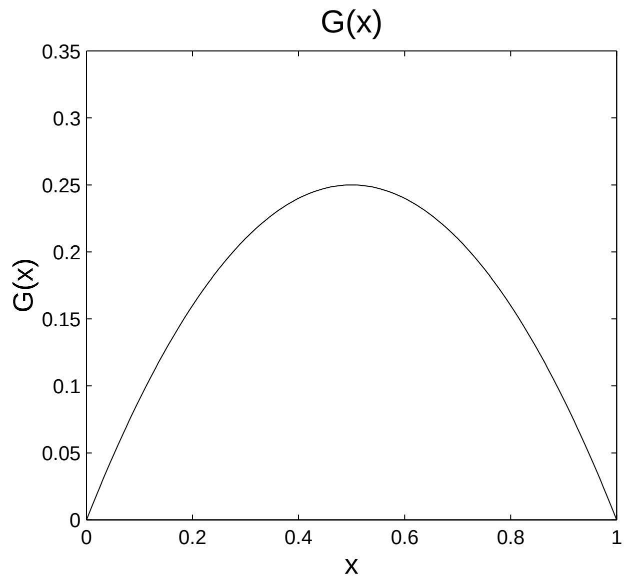
Here, we have used the Lyman-α wavelength, $\lambda_{ba} = 122\,\text{nm}$. One could also use atomic units for this calculation and make the arithmetic a little simpler. Since $a_o = 1$, $\omega_{ba} = 3/8$, and $c = \alpha^{-1}$ in atomic units,

$$A_\tau \cong \frac{2}{9\pi} \left(\frac{3}{8} \right)^5 \left(\frac{1}{137} \right)^6 \cong 7.9 \times 10^{-17}. \tag{35}$$

This last quantity has units of inverse time. Using the fact that 1 atomic unit of time equals 2.42×10^{-17} s, we again find $A_\tau \sim 3 \text{ s}^{-1}$. Thus, we have estimated that the $2S$ natural lifetime is on the order of 1/3 s, correct to order of magnitude.

▷ Plot the spectrum $A(\omega_1)$ over its entire range.

The emission spectrum, $G(x)$, is a simple parabola, symmetric with respect to $x = 1/2$. If a more exact numerical calculation is performed including other P-states and the continuum, one finds that the spectrum is parabolic in the center and steeper than a parabola near the edges (see below).



A more accurate calculation from J. H. Tung, *et al.* Phys. Rev. A. **30**, 1175 (1984).

Problem 2. Bragg Scattering

(a) Calculate the two-photon Rabi frequency for the Raman process.

The electric fields of the two beams can be expressed

$$E_1 = \frac{\mathcal{E}_1}{2}\left(e^{i(kz-\omega_1 t)} + e^{-i(kz-\omega_1 t)}\right) \quad (36)$$
$$E_2 = \frac{\mathcal{E}_2}{2}\left(e^{i(-kz-\omega_2 t)} + e^{-i(-kz-\omega_2 t)}\right) \quad (37)$$

where $\mathcal{E}_1 = \mathcal{E}_2 = E_o$ and $\omega_2 = \omega_1 + \Delta\omega$. The interaction Hamiltonian then has the form

$$H = -\frac{1}{2}\left(e^{i(kz-\omega_1 t)} + e^{-i(kz-\omega_1 t)}\right)\mathcal{E}_1\hat{\mathbf{e}}_1 \cdot \mathbf{D} - \frac{1}{2}\left(e^{-i(kz+\omega_2 t)} + e^{i(kz+\omega_2 t)}\right)\mathcal{E}_2\hat{\mathbf{e}}_2 \cdot \mathbf{D}. \quad (38)$$

The only difference from the case of two-photon excitation treated in Lecture 20 (and sections 9.2-9.3 of the old course notes) is that there is a spatial dependence in the factors $e^{\pm ikz}$—these factors will disappear when matrix elements are evaluated. (Since the two beams have the same polarization, we can simply use a scalar dipole operator $D = \hat{\mathbf{e}}_\mathbf{1} \cdot \mathbf{D} = \hat{\mathbf{e}}_\mathbf{2} \cdot \mathbf{D}$.)

We can now employ the second-order perturbation theory method of Lecture 20 (or sections 9.2 and 9.3). The two-photon Rabi frequency is defined in terms of the one-photon Rabi frequencies for each of the beams, which are in turn defined in terms of interaction matrix elements. The relevant matrix elements for this problem are

$$H_{ki,1} = -E_o\langle k|De^{+ikz}|i\rangle \quad (39)$$
$$H_{fk,2} = -E_o\langle f|De^{-ikz}|k\rangle. \quad (40)$$

Let's evaluate $H_{ki,1}$ explictly.

$$H_{ki,1} = -E_o\langle e, \hbar k|De^{+ikz}|g, 0\rangle \quad (41)$$
$$= -E_o\langle e|D|g\rangle \int_0^L dz\, \frac{e^{-ikz}}{\sqrt{L}} e^{+ikz} \frac{e^{-i(0)z}}{\sqrt{L}} \quad (42)$$
$$= -E_o D_{eg} \quad (43)$$

where plane wave eigenstates for a 1-dimensional box of length L (periodic boundary conditions) have been used for the momentum states. Likewise, one also finds $H_{fk,2} = -E_o D_{eg}$. Note that we do not need to make the usual assumption that both beams are nearly resonant ($\Delta \ll \omega_{eg}$) in order to drop the counter-rotating terms. Rather, the matrix elements of the counter-rotating terms vanish entirely because they do not conserve momentum. (Consider $\langle k|De^{-ikz}|i\rangle$, for example.)

Following the definition from Lecture 20 (section 9.3), the two-photon Rabi frequency ω_{R2} is

$$\omega_{R2} = \frac{\omega_R^{(1)}\omega_R^{(2)}}{2\Delta} \quad (44)$$
$$= \frac{|H_{ki,1}|^2|H_{fk,2}|^2}{2\hbar^2\Delta} \quad (45)$$
$$= \frac{E_o^2 D_{eg}^2}{2\hbar^2\Delta}. \quad (46)$$

▷ What should $\Delta\omega$ be in order to realize the Raman resonance condition?

To realize resonance, we must conserve energy:

$$\Delta\omega = \frac{-(E_f - E_i)}{\hbar} \tag{47}$$
$$= -\frac{1}{\hbar}\frac{(2\hbar k)^2}{2m} \tag{48}$$
$$= -\frac{2\hbar k^2}{m}. \tag{49}$$

In other words, the frequency difference between the beams must supply the kinetic energy associated with momentum $2\hbar k$, which is equal to 4 times the single-photon recoil energy. As an example, for a sodium atom excited on its D-line, this frequency difference is on the order of $2\pi \times 100$ kHz. Thus, $\Delta\omega/\omega = \Delta k/k \sim 10^{-9}$. This is why it's a very good approximation to use the same wavevector magnitude k for both beams.

▷ If H' is the perturbation due to E_1 and E_2, what is $\langle i|H'|f\rangle$?

We want to find the effective matrix element H'_{if} for this Raman process which couples $|i\rangle$ to $|f\rangle$. One way to do this, which avoids all mention of Rabi frequencies, is to use the expression for the transition rate, analogous to that derived in Lecture 20 (see pg. 4 of Lecture 20 notes or eq. 9.14):

$$\Gamma_{if} = \frac{\pi}{8\hbar^4}\frac{E_o^4 D_{eg}^4}{\Delta^2} f(\Delta\omega). \tag{50}$$

Here, $f(\Delta\omega)$ is a spectral distribution function. The same rate can be expressed in terms of H'_{if} using Fermi's Golden Rule,

$$\Gamma_{if} = \frac{2\pi}{\hbar^2}|H'_{if}|^2 f(\Delta\omega). \tag{51}$$

Solving for the magnitude of H'_{if}, we obtain

$$|\langle i|H'|f\rangle| = \frac{E_o^2 D_{eg}^2}{4\hbar\Delta}. \tag{52}$$

We can come up with the same answer if we remember that, for a real sinusoidal perturbation, the magnitude of the interaction matrix element is just $\hbar/2$ times the Rabi frequency. In this case,

$$|\langle i|H'|f\rangle| = \frac{\hbar\omega_{R2}}{2}. \tag{53}$$

One must be careful, though—conventions for defining the Rabi frequency are unfortunately not universal!

(b) Calculate the AC Stark shift $U(z,t)$ due to the total electric field $E_1 + E_2$.

The AC Stark shift for this two-level atom in the oscillating electric field is given by

$$U(z,t) = -\frac{\overline{[E(z,t)]^2} D_{eg}^2}{2\hbar\Delta}, \tag{54}$$

where the overline bar indicates a time average over optical frequencies. This can be derived using either the dressed atom picture or perturbation theory in the near-resonant limit. (Recall

Problem 1 of Homework #5; see also notes from Lecture 10). In the near-resonant limit (rotating wave approximation), the polarizability becomes

$$\alpha(\omega) = \frac{D_{eg}^2}{\hbar}\left(\frac{1}{\omega_{eg}-\omega}+\frac{1}{\omega_{eg}+\omega}\right) \tag{55}$$

$$\cong \frac{D_{eg}^2}{\hbar\Delta}, \tag{56}$$

where ω is the frequency of the electric field, and $\Delta = \omega_{eg} - \omega$ is defined as in the problem. The AC Stark shift is then given by

$$U = -\frac{1}{2}\alpha(\omega)\overline{[E(t)]^2}\,, \tag{57}$$

which leads to Eq. 54. Note that in the DC limit ($\omega \ll \omega_{eg}$), this expression would be larger by a factor of two because the contribution from the rotating and counter-rotating terms is then approximately equal.

What we need to do now is calculate the time average of the total electric field experienced by the atom. As mentioned already, the frequency $\Delta\omega$ is many orders of magnitude less than optical frequencies. Since ω is responsible for the interesting physics of the problem, we want to average over a time slow compared to optical frequencies but fast compared to $\Delta\omega$. Making use of complex notation,

$$\overline{(E_1+E_2)^2} = \frac{E_o^2}{4}\overline{\left(e^{i(kz-\omega_1 t)}+e^{-i(kz-\omega_1 t)}+e^{i[-kz-(\omega_1+\Delta\omega)t]}+e^{-i[-kz-(\omega_1+\Delta\omega)t]}\right)^2}\,. \tag{58}$$

Squaring the quantity under the time average leads to 16 terms. There are four terms equal to unity, two terms oscillating in time with frequency $-\Delta\omega$, two terms oscillating with frequency $\Delta\omega$, and the remaining terms oscillate at optical frequencies. Taking the time average then leaves us with

$$\overline{(E_1+E_2)^2} = \frac{E_o^2}{4}\left(4+2e^{i(2kz+\Delta\omega t)}+2e^{-i(2kz+\Delta\omega t)}\right) \tag{59}$$

$$= E_o^2[1+\cos(2kz+\Delta\omega t)]. \tag{60}$$

It follows that the external potential seen by an atom due to the AC Stark shift is

$$U(z,t) = -\frac{E_o^2 D_{eg}^2}{2\hbar\Delta}[1+\cos(2kz+\Delta\omega t)]. \tag{61}$$

This form is reasonable since the field due to superposition of E_1 and E_2 is just a "standing wave" moving with phase velocity $\Delta\omega/(2k)$.

(c) What is the coupling $\langle i|U(z,t)|f\rangle$ due to the mechanical potential presented by the AC Stark shift? Compare this with the perturbation matrix element obtained in part (a).

The potential due to the AC Stark shift has Fourier components at spatial frequencies $\pm 2k$:

$$U(z,t) = \frac{E_o^2 D_{eg}^2}{2\hbar\Delta}\left[1+\frac{1}{2}e^{i(2kz+\Delta\omega t)}+\frac{1}{2}e^{-i(2kz+\Delta\omega t)}\right]. \tag{62}$$

Using the same plane wave momentum states as in part (a), we find that only one of the components contributes to the coupling between $|i\rangle$ and $|f\rangle$:

$$\begin{aligned}
\langle i|U(z,t)|f\rangle &= \langle g,0|U(z,t)|g,2\hbar k\rangle && (63)\\
&= \langle g|g\rangle\langle 0|U(z,t)|2\hbar k\rangle && (64)\\
&= 1\times\frac{1}{L}\int_0^L dz\; U(z,t)e^{i2kz} && (65)\\
&= \frac{1}{L}\int_0^L dz\,\frac{E_o^2 D_{eg}^2}{4\hbar\Delta}e^{-i2(kz+\Delta\omega t)}e^{i2kz} && (66)\\
&= \frac{E_o^2 D_{eg}^2}{4\hbar\Delta}e^{-i\Delta\omega t}. && (67)
\end{aligned}$$

Thus, up to a phase factor, the matrix element is identical to that derived in part (a). In other words, the light shift potential picture and the stimulated Raman picture predict the same transition rates.

Solutions compiled by Stephen Moss, 27 April 2000. Revision by Jaroslaw Labaziewicz, 7 May 2006; and minor edits by Monika Schleier-Smith, 5 May 2008.