

Homework 3

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1 Two-photon absorption in a three-level system

In this problem, we use perturbation theory to investigate two-photon absorption within a three-level atom, with states $|a\rangle, |b\rangle$ and $|c\rangle$ with energy eigenvalues $E_a = \hbar\omega_a, E_b = \hbar\omega_b$, and $E_c = \hbar\omega_c$ such that $E_c > E_b > E_a$; here $|a\rangle$ and $|c\rangle$ are assumed to have even parity and $|b\rangle$ has odd parity. In the problem that follows, we drive this three-level atom with a monochromatic field $E(t) = E_0 \cos(\omega t)$ producing an interaction of the form $H_{\text{int}} = -\hat{\mu} \cdot E(t)$, and we use time-dependent perturbation theory to find the evolution of our quantum state of the form $|\psi\rangle = \sum_n \gamma_n e^{-i\omega_n t} |n\rangle$.

(a) Assuming that our atom starts in the ground state (i.e. $\gamma_a^{(0)} = 1$), use second-order perturbation theory to find $\gamma_c^{(2)}(t)$. Through these calculations, we will assume that states $|b\rangle$ and $|c\rangle$ have finite upper state lifetimes. To emulate population decay, be sure to include a phenomenological damping into your susceptibility by making the replacement $\omega_b \rightarrow \omega_b - i\Gamma_b$ and $\omega_c \rightarrow \omega_c - i\Gamma_c$ where Γ_a and Γ_b are small compared to the transition frequencies.

The time dependent perturbation theory is summarized as

$$\langle k | \psi(t) \rangle = \sum_{i=0}^{\infty} \gamma_k^{(i)} e^{-i\omega_k t}, \quad (1)$$

$$\gamma_k^{(0)} = \text{const.}, \quad \frac{d\gamma_k^{(i+1)}}{dt} = \frac{1}{i\hbar} \sum_n H_{kn}^{(1)} \gamma_n^{(i-1)} e^{i(\omega_k - \omega_n)t}. \quad (2)$$

In the current case, due to parity conservation in dipole transition, only transitions $a \rightarrow b$ and $b \rightarrow c$ are possible. Therefore in the first order perturbation theory, only $\gamma_b^{(1)}$ is non-zero. We have

$$\frac{d\gamma_b^{(1)}}{dt} = \frac{1}{i\hbar} H_{ba}^{\text{dipole}} \gamma_a^{(0)} e^{i(\omega_b - \omega_a)t}, \quad (3)$$

$$\begin{aligned} \gamma_b^{(1)} &= -\frac{\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0}{i\hbar} \cdot \frac{1}{2} \int_{-\infty}^t dt' \left(e^{i(\omega + \omega_b - \omega_a)t'} + e^{i(-\omega + \omega_b - \omega_a)t'} \right) \\ &= \frac{\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0}{\hbar} \cdot \frac{1}{2} \left(\frac{e^{i(\omega + \omega_b - \omega_a)t}}{\omega + \omega_b - \omega_a} + \frac{e^{i(-\omega + \omega_b - \omega_a)t}}{-\omega + \omega_b - \omega_a} \right). \end{aligned} \quad (4)$$

Here since we are interested in the response properties of the three-level system under plane waves, not, say, how fast the atom goes away from its initial state, we have pushed the lower bound to $-\infty$, and since each excited state has a finite lifetime, the $-1/(\pm\omega + \omega_b - \omega_a)$ term is simply thrown away, following the practice in the last homework and in the lectures.

Similarly, from

$$\frac{d\gamma_c^{(2)}}{dt} = \frac{1}{i\hbar} H_{cb}^{\text{dipole}} \gamma_b^{(1)} e^{i(\omega_c - \omega_b)t} = -\frac{1}{i\hbar} \cdot \frac{1}{2} \boldsymbol{\mu}_{cb} \cdot \mathbf{E}_0 (e^{i(\omega + \omega_c - \omega_b)t} + e^{i(-\omega + \omega_c - \omega_b)t}) \cdot \gamma_b^{(1)}, \quad (5)$$

we get

$$\begin{aligned} \gamma_c^{(2)} &= \frac{\boldsymbol{\mu}_{cb} \cdot \mathbf{E}_0}{\hbar} \frac{\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0}{\hbar} \cdot \frac{1}{4} \left(\frac{e^{i(2\omega + \omega_c - \omega_a)t}}{(2\omega + \omega_c - \omega_a)(\omega + \omega_b - \omega_a)} + \right. \\ &\quad \frac{e^{i(-2\omega + \omega_c - \omega_a)t}}{(-2\omega + \omega_c - \omega_a)(-\omega + \omega_b - \omega_a)} + \frac{e^{i(\omega_c - \omega_a)t}}{(\omega_c - \omega_a)(\omega + \omega_b - \omega_a)} + \\ &\quad \left. \frac{e^{i(\omega_c - \omega_a)t}}{(\omega_c - \omega_a)(-\omega + \omega_b - \omega_a)} \right). \end{aligned} \quad (6)$$

Since each excited state has finite lifetime, we do the substitution $\omega_{b,c} \rightarrow \omega_{b,c} - i\Gamma_{b,c}$ and get

$$\gamma_c^{(2)} = \frac{\boldsymbol{\mu}_{cb} \cdot \mathbf{E}_0}{\hbar} \frac{\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0}{\hbar} \cdot \frac{1}{4} \left(\frac{e^{i(2\omega + \omega_c - \omega_a)t}}{(2\omega + \omega_c - \omega_a - i\Gamma_c)(\omega + \omega_b - \omega_a - i\Gamma_b)} + \frac{e^{i(-2\omega + \omega_c - \omega_a)t}}{(-2\omega + \omega_c - \omega_a - i\Gamma_c)(-\omega + \omega_b - \omega_a - i\Gamma_b)} + \frac{e^{i(\omega_c - \omega_a)t}}{(\omega_c - \omega_a - i\Gamma_c)(\omega + \omega_b - \omega_a - i\Gamma_b)} + \frac{e^{i(\omega_c - \omega_a)t}}{(\omega_c - \omega_a - i\Gamma_c)(-\omega + \omega_b - \omega_a - i\Gamma_b)} \right). \quad (7)$$

Here in principle we should also include the imaginary parts of $\omega_{b,c}$ in the oscillating factors $e^{i(\dots)t}$; if we do so, eventually we should multiply $e^{\Gamma_c t}$ to the expression above. But similar to the case in the last homework, when evaluating $\langle \psi |$, we should do the substitution $\omega_{b,c} \rightarrow \omega_{b,c} + i\Gamma_{b,c}$, and in the c component of $\langle \psi |$ we get a $e^{i(\Gamma_c)t}$ factor, which cancels the $e^{\Gamma_c t}$ factor in $|\psi\rangle$; on the other hand, the imaginary parts in the denominator do not cancel and have real physical consequences when we calculate the expectation values. So we can ignore the $e^{\Gamma_c t}$ factor in $\gamma_c^{(2)}$ and still get everything right.

(b) Assuming that our energy spacing meets the condition for resonant twophoton absorption (i.e., $\omega_c - \omega_a \cong 2\omega$), perform the rotating wave approximation to simplify your result from part (a). [Hint: all of the terms with fast-oscillating phases should vanish] Use this result find the probability, P_c , that the atom occupies state $|c\rangle$. Since this probability, P_c , is independent of time it can also be viewed as the steady-state population of state $|c\rangle$.

Since $\omega_c - \omega_a \simeq 2\omega$, only the $-2\omega + \omega_c - \omega_a$ term is the slowly-oscillating term, and by rotating wave approximation

$$\gamma_c^{(2)} \approx \frac{\boldsymbol{\mu}_{cb} \cdot \mathbf{E}_0}{\hbar} \frac{\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0}{\hbar} \cdot \frac{1}{4} \frac{e^{i(-2\omega + \omega_c - \omega_a)t}}{(-2\omega + \omega_c - \omega_a - i\Gamma_c)(-\omega + \omega_b - \omega_a - i\Gamma_b)}. \quad (8)$$

Thus the steady-state population on state c is

$$P_c = |\psi_c^{(2)}|^2 = \frac{|\boldsymbol{\mu}_{cb} \cdot \mathbf{E}_0|^2 |\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0|^2}{16\hbar^4} \frac{1}{((\omega_c - \omega_a - 2\omega)^2 + \Gamma_c^2)((\omega_b - \omega_a - \omega)^2 + \Gamma_b^2)}. \quad (9)$$

(c) Notice that the excited state population satisfies the following rate equation $dP_c/dt = -2\Gamma_c P_c + R_c$, where R_c is the two-photon absorption rate for the atom. Use this expression to find R_c and the two-photon absorption crosssection, $\sigma_{TPA}(I)$, at steady state; here, I is the intensity of light. For the purposes of this calculation, assume the atom lives in a material with refractive index n . [Hint: $R_c = I\sigma_{TPA}(I)/\hbar\omega$]

When equilibrium hasn't be achieved, since $\psi_c^{(2)} \sim e^{-\Gamma_c t} \Rightarrow P_c \sim e^{-2\Gamma_c t}$, the rate equation for P_c is

$$\frac{dP_c}{dt} = -2\Gamma_c P_c + R_c, \quad (10)$$

where R_c is the two-photon absorption rate for the atom. Now since the system is already in equilibrium, we have

$$R_c = 2\Gamma_c P_c = \frac{|\boldsymbol{\mu}_{cb} \cdot \mathbf{E}_0|^2 |\boldsymbol{\mu}_{ba} \cdot \mathbf{E}_0|^2}{8\hbar^4} \frac{\Gamma_c}{((\omega_c - \omega_a - 2\omega)^2 + \Gamma_c^2)((\omega_b - \omega_a - \omega)^2 + \Gamma_b^2)}. \quad (11)$$

The two-photon absorption (TPA) cross section can be determined by

$$R_c \cdot \hbar\omega = I\sigma_{TPA} = \text{energy absorbed per second}. \quad (12)$$

The intensity is the time average of Poynting vector,

$$I = \frac{1}{2} \epsilon_0 c n |\mathbf{E}_0|^2, \quad (13)$$

and therefore by replacing E_0^2 by I , we get

$$\sigma_{TPA} = \frac{\omega I}{2n^2 \epsilon_0^2 c^2 \hbar^3} |\boldsymbol{\mu}_{cb} \cdot \hat{\mathbf{e}}|^2 |\boldsymbol{\mu}_{ba} \cdot \hat{\mathbf{e}}|^2 \frac{\Gamma_c}{((\omega_c - \omega_a - 2\omega)^2 + \Gamma_c^2)((\omega_b - \omega_a - \omega)^2 + \Gamma_b^2)}. \quad (14)$$

Here $\hat{\mathbf{e}}$ is the polarization direction of the incident beam.

(d) It takes $dt = dz/(c/n)$ for a plane wave beam to propagate dz , where n is the refractive index. During this period of time, the energy loss due to second order effect is (note that there are two photons absorbed a time)

$$dI = \underbrace{N}_{\text{number density}} \cdot \underbrace{R_c \cdot 2\hbar\omega}_{\Delta E \text{ per second per atom}} \cdot dt = N \frac{n}{c} R_c \hbar\omega dz. \quad (15)$$

The definition of β_2 is

$$\frac{dI}{dz} = -\alpha I - \beta_2 I^2, \quad (16)$$

and we find

$$\begin{aligned} \beta_2 &= \frac{N}{I^2} \cdot \frac{n}{c} R_c \cdot 2\hbar\omega \\ &= \frac{\omega N}{n\epsilon_0^2 c^3 \hbar^3} |\boldsymbol{\mu}_{cb} \cdot \hat{\mathbf{e}}|^2 |\boldsymbol{\mu}_{ba} \cdot \hat{\mathbf{e}}|^2 \frac{\Gamma_c}{((\omega_c - \omega_a - 2\omega)^2 + \Gamma_c^2)((\omega_b - \omega_a - \omega)^2 + \Gamma_b^2)}. \end{aligned} \quad (17)$$

2 Quantum treatment of nonlinear susceptibility

(a) In this section we generalize the procedure in the first problem. The external electric field now is

$$\mathbf{E}(t) = \sum_p \mathbf{E}_p e^{-i\omega_p t} + \text{c.c.} \quad (18)$$

Here to simplify the derivation, we follow the convention in Boyd and slightly misuse the notation in the following way:

- \sum_p means to sum over all optical modes p , and the positive- and negative-frequencies.
- $\mathbf{E}(\omega_p)$, likewise, means \mathbf{E}_p when $\omega_p > 0$, and \mathbf{E}_p^* when $\omega_p < 0$.

In this way $\sum_p F(\mathbf{E}(\omega_p)^*, -\omega_p) e^{i\omega_p t}$ can be replaced by $\sum_p F(\mathbf{E}(\omega_p), \omega_p) \times e^{-i\omega_p t}$.

The zeroth order state is

$$\gamma_n^{(0)} = \delta_{n,g}. \quad (19)$$

Therefore the first order perturbation is determined by

$$\frac{d\gamma_m^{(1)}}{dt} = \frac{1}{i\hbar} \underbrace{(-\boldsymbol{\mu}_{mg} \cdot \sum_p \mathbf{E}(\omega_p) e^{-i\omega_p t})}_{H_{\text{int}}} e^{i(\omega_m - \omega_g)t}, \quad (20)$$

and therefore after integration we get

$$\gamma_m^{(1)}(t) = \frac{1}{\hbar} \sum_p \frac{\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)}{(\omega_m - \omega_g - \omega_p)} e^{i(\omega_m - \omega_g - \omega_p)t}. \quad (21)$$

The second order perturbation is determined by

$$\frac{d\gamma_m^{(2)}}{dt} = \frac{1}{i\hbar} \sum_n \underbrace{(-\boldsymbol{\mu}_{mn} \cdot \sum_q \mathbf{E}(\omega_q) e^{-i\omega_q t})}_{H_{\text{int}}} e^{i(\omega_m - \omega_n)t} \cdot \frac{1}{\hbar} \underbrace{\sum_p \frac{\boldsymbol{\mu}_{ng} \cdot \mathbf{E}(\omega_p)}{(\omega_n - \omega_g - \omega_p)} e^{i(\omega_n - \omega_g - \omega_p)t}}_{\gamma_n^{(1)}}, \quad (22)$$

and after integration we get

$$\gamma_m^{(2)}(t) = \frac{1}{\hbar^2} \sum_{p,q} \sum_n \frac{(\boldsymbol{\mu}_{mn} \cdot \mathbf{E}(\omega_q))(\boldsymbol{\mu}_{ng} \cdot \mathbf{E}(\omega_p))}{(\omega_m - \omega_g - \omega_p - \omega_q)(\omega_n - \omega_g - \omega_p)} e^{i(\omega_m - \omega_g - \omega_p - \omega_q)t}. \quad (23)$$

(b) The second order polarization is given by

$$\langle \boldsymbol{\mu} \rangle^{(2)} = \langle \psi^{(2)} | \boldsymbol{\mu} | \psi^{(0)} \rangle + \langle \psi^{(1)} | \boldsymbol{\mu} | \psi^{(1)} \rangle + \langle \psi^{(0)} | \boldsymbol{\mu} | \psi^{(2)} \rangle, \quad (24)$$

and therefore (note that we still need a $e^{-i\omega_i t}$ factor besides γ_i)

$$\begin{aligned} \langle \boldsymbol{\mu} \rangle^{(2)} = & \frac{1}{\hbar^2} \sum_{p,q} \sum_{m,n} e^{i\omega_m t} \frac{(\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(-\omega_q))(\boldsymbol{\mu}_{gn} \cdot \mathbf{E}(-\omega_p))\boldsymbol{\mu}_{mg}}{(\omega_m^* - \omega_g - \omega_p - \omega_q)(\omega_n^* - \omega_g - \omega_p)} e^{-i(\omega_m - \omega_g - \omega_p - \omega_q)t} e^{-i\omega_g t} \\ & + \frac{1}{\hbar^2} \sum_{m,n} \sum_{p,q} e^{i\omega_m t} \frac{\boldsymbol{\mu}_{gm} \cdot \mathbf{E}(-\omega_p)}{(\omega_m^* - \omega_g - \omega_p)} e^{-i(\omega_m - \omega_g - \omega_p)t} \boldsymbol{\mu}_{mn} \frac{\boldsymbol{\mu}_{ng} \cdot \mathbf{E}(\omega_q)}{(\omega_n - \omega_g - \omega_q)} e^{i(\omega_m - \omega_g - \omega_p)t} e^{-i\omega_n t} \\ & + \frac{1}{\hbar^2} \sum_{p,q} \sum_{m,n} e^{i\omega_g t} \frac{(\boldsymbol{\mu}_{mn} \cdot \mathbf{E}(\omega_q))(\boldsymbol{\mu}_{ng} \cdot \mathbf{E}(\omega_p))\boldsymbol{\mu}_{gm}}{(\omega_m - \omega_g - \omega_p - \omega_q)(\omega_n - \omega_g - \omega_p)} e^{i(\omega_m - \omega_g - \omega_p - \omega_q)t} e^{-i\omega_m t}. \end{aligned} \quad (25)$$

Here to simply the form of the denominator, we have attract $i\Gamma_{m,n}$ into $\omega_{m,n}$. After simplification and changing the signs of $\omega_{p,q}$ (see the discussion at the beginning of problem (a)) we have

$$\begin{aligned} \langle \boldsymbol{\mu} \rangle^{(2)} = & \frac{1}{\hbar^2} \sum_{p,q} \sum_{m,n} e^{-i(\omega_p + \omega_q)t} \left(\frac{(\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_q))(\boldsymbol{\mu}_{gn} \cdot \mathbf{E}(\omega_p))\boldsymbol{\mu}_{mg}}{(\omega_m^* - \omega_g + \omega_p + \omega_q)(\omega_n^* - \omega_g + \omega_p)} \right. \\ & + \frac{(\boldsymbol{\mu}_{gm} \cdot \mathbf{E}(\omega_p))(\boldsymbol{\mu}_{ng} \cdot \mathbf{E}(\omega_q))\boldsymbol{\mu}_{mn}}{(\omega_m^* - \omega_g + \omega_p)(\omega_n - \omega_g - \omega_q)} \\ & \left. + \frac{(\boldsymbol{\mu}_{mn} \cdot \mathbf{E}(\omega_q))(\boldsymbol{\mu}_{ng} \cdot \mathbf{E}(\omega_p))\boldsymbol{\mu}_{gm}}{(\omega_m - \omega_g - \omega_p - \omega_q)(\omega_n - \omega_g - \omega_p)} \right). \end{aligned} \quad (26)$$

After swapping p and q and m and n in some of the terms, we get Boyd (3.2.26). The expression can also be obtained in a many-body perturbation theoretic way: we set the chemical potential to somewhere between ω_g and the first excited state, and then (26) can be derived from the two photon in, one photon out electron loop diagram in the same way Lindhard function is derived from the electron-hole loop.

(c) $\chi^{(2)}$ is (26) divided by $E(\omega_p)$ and $E(\omega_q)$; so it's proportional to $\mu_{mn}\mu_{ng}\mu_{gm}$. When the system has inversion symmetry and therefore eigenstates in the system are all parts of representations of the inversion operation, and therefore have definite parities, in order for this product to be non-zero, we need $|m\rangle$, $|n\rangle$, $|g\rangle$ to all have different parities. But this is not possible: if $|g\rangle$ and $|m\rangle$ have different parities and $|m\rangle$ and $|n\rangle$ have different parities, and the parities of $|g\rangle$ and $|n\rangle$ have to be the same, because the parity of a state is either even or odd: and then μ_{ng} is zero. So whenever the eigenstates have definite parities (in other words, the system has inversion symmetry), $\chi^{(2)}$ vanishes.

3 Three-wave interaction

3.1 Classical stimulated Raman effect

When the polarizability depends on a phonon mode, we have

$$L = \int d^3\mathbf{r} \left(\frac{1}{2} \epsilon(q) \dot{\mathbf{A}}^2 - \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 \right) + L_{\text{non-interactive}}[q] - H_{\text{dipole}}, \quad (27)$$

and from this Lagrangian we can verify that the displacement vector is

$$\mathbf{D} = \frac{\delta L}{\delta(-\dot{\mathbf{A}})} = \epsilon(q) \mathbf{E} + \boldsymbol{\mu}_{\text{linear}}. \quad (28)$$

If on the other hand, the electric field is seen as a given external field, the effective Hamiltonian about the phonon mode then is

$$H = \frac{p^2}{2m} + \frac{1}{2} m \Omega_0^2 q^2 + \frac{1}{2} \epsilon(q) \dot{\mathbf{A}}^2 + H_{\text{dipole}}, \quad (29)$$

and after Taylor expansion of $\epsilon(q)$ we get

$$H = \frac{p^2}{2m} + \frac{1}{2}m\Omega_0^2 q^2 + \frac{1}{2} \left. \frac{\partial \alpha}{\partial q} \right|_0 qE^2 - eqE, \quad (30)$$

where e is the charge of the dipole induced by that phonon mode.

(a) The optical force on coordinate q is therefore

$$F_q = -\frac{\partial H}{\partial q} = -m\Omega_0^2 q - \underbrace{\frac{1}{2} \left. \frac{\partial \alpha}{\partial q} \right|_0 E^2 + eE}_{\text{produced by light}}. \quad (31)$$

(b) Next, we consider the presence of two driving fields such that $E(t) = E_1(t) + E_2(t)$, where $E_1(t) = E_1^0 \cos(\omega_1 t)$ and $E_2(t) = E_2^0 \cos(\omega_2 t)$. The driving frequencies are related by $\omega_1 = \omega_2 + \Omega$, permitting us to view ω_1 as the (blue) pump-field and ω_2 as the (red) Stokes-field. (Here, we assume that $\Omega \approx \Omega_o$ and $\Omega_o \ll \omega_1, \omega_2$.)

The force on q then is

$$F_q = -m\Omega_0^2 q + e(E_1^0 \cos \omega_1 t + E_2^0 \cos \omega_2 t) - \frac{1}{2} \left. \frac{\partial \alpha}{\partial q} \right|_0 (E_1^0 \cos \omega_1 t + E_2^0 \cos \omega_2 t)^2. \quad (32)$$

(c) Find the equation of motion for $q(t)$, keeping only frequency terms close to the Raman resonance. (Hint, only the cross-terms between E_1 and E_2 should survive.)

Using the product-to-sum formula, we find the only term in F_q whose frequency is close to the Raman resonance is the $\omega_1 - \omega_2$ term in the qE^2 term. So the equation of motion for the Ω component of q is (below we use q to refer to the Ω component of q)

$$m\ddot{q} = F_q(\Omega) = -m\Omega_0^2 q - \frac{1}{2} \left. \frac{\partial \alpha}{\partial q} \right|_0 E_1^0 E_2^0 \cos \Omega t. \quad (33)$$

(d) Next, include damping with an amplitude decay, $1/\tau$, and find an expression for the steady-state phonon amplitude. [Hint: it makes more sense to work with the envelope equations (i.e. $q = \text{Re}[\tilde{q} \cdot e^{-i\Omega t}]$). Remember, at steadystate, $\dot{\tilde{q}} = 0$.]

Adding the damping term to (33), we have

$$\ddot{q} + \frac{1}{\tau} \dot{q} + \Omega_0^2 q = -\frac{1}{2m} \left. \frac{\partial \alpha}{\partial q} \right|_0 E_1^0 E_2^0 \cos \Omega t. \quad (34)$$

Using the envelope ansatz

$$q = \frac{1}{2}(\tilde{q}e^{-i\Omega t} + \text{c.c.}), \quad (35)$$

and ignoring the $\ddot{\tilde{q}}$ term because \tilde{q} is assumed to vary slowly, we have

$$-\Omega^2 \tilde{q} - 2i\Omega \dot{\tilde{q}} + \frac{1}{\tau} \dot{\tilde{q}} - \frac{i\Omega}{\tau} \tilde{q} + \Omega_0^2 \tilde{q} = -\frac{1}{2m} \left. \frac{\partial \alpha}{\partial q} \right|_0 E_1^0 E_2^0. \quad (36)$$

At steady state, we have $\dot{\tilde{q}} = 0$, and the above equation becomes

$$\tilde{q} = \frac{1}{\Omega^2 - \Omega_0^2 + i\Omega/\tau} \frac{1}{2m} \left. \frac{\partial \alpha}{\partial q} \right|_0 E_1^0 E_2^0. \quad (37)$$

With the approximation $\Omega \approx \Omega_0$ we have

$$\tilde{q} = \frac{\tau}{i\Omega} \frac{1}{2m} \left. \frac{\partial \alpha}{\partial q} \right|_0 E_1^0 E_2^0. \quad (38)$$

This is the steady-state phonon amplitude.

(e) Next, we find the nonlinear polarizability produced by the Raman interaction. We do this by taking the steady-state phonon amplitude from part (d) and substituting it into $\mu_R = \left(\frac{\partial\alpha}{\partial q}\right) qE = \left(\frac{\partial\alpha}{\partial q}\right) q(E_1(t) + E_2(t))$. Using this expression, find the complex polarizability associated with $\mu_R(\omega_2; \omega_1, -\omega_1, \omega_2)$.

The electric dipole induced by the qE^2 term, under (37), is

$$\mu_R = \left.\frac{\partial\alpha}{\partial q}\right|_0 \cdot \frac{1}{2} \left(\frac{1}{\Omega^2 - \Omega_0^2 + i\Omega/\tau} \frac{1}{2m} \left.\frac{\partial\alpha}{\partial q}\right|_0 E_1^0 E_2^0 e^{-i\Omega t} + \text{c.c.} \right) \cdot \frac{1}{2} (E_1^0 e^{-i\omega_1 t} + E_2^0 e^{-i\omega_2 t} + \text{c.c.}), \quad (39)$$

and therefore the $\mu_R(\omega_2; \omega_1, -\omega_1, \omega_2)$ component is

$$\mu_R(\omega_2; \omega_1, -\omega_1, \omega_2) = \frac{1}{2} \left.\frac{\partial\alpha}{\partial q}\right|_0 \cdot \frac{1}{\Omega^2 - \Omega_0^2 - i\Omega/\tau} \frac{1}{2m} \left.\frac{\partial\alpha}{\partial q}\right|_0 E_1^0 E_2^0 \cdot \frac{1}{2} E_1^0 e^{-i\omega_2 t} + \text{c.c.} \quad (40)$$

The polarizability therefore is

$$\chi_R(\omega_2; \omega_1, -\omega_1, \omega_2) = N\alpha_R = \left(\left.\frac{\partial\alpha}{\partial q}\right|_0\right)^2 \frac{N}{m(\Omega^2 - \Omega_0^2 - i\Omega/\tau)}. \quad (41)$$

Here we treat $E = \text{Re } \tilde{E} e^{-i\omega t}$ as the external field, not just \tilde{E} , so all the 1/2 factors disappear.

3.2 Idealized Raman interaction via Fermi's Golden rule

In this problem, we consider coupling between two quantized electromagnetic modes $(\hat{\mathbf{E}}_p, \hat{\mathbf{E}}_s)$ by a quantized vibrational mode $(\hat{x} = (b^\dagger + b)x_o)$. In the absence of the Raman coupling, the Hamiltonian for the system is $H = \hbar\omega_p (a_p^\dagger a_p + \frac{1}{2}) + \hbar\omega_s (a_s^\dagger a_s + \frac{1}{2}) + \hbar\Omega (b^\dagger b + \frac{1}{2})$, where a_p and a_s are the lowering operators for the optical fields and b is the lowering operator of the phonon mode. The frequencies of these modes are related by $\omega_p = \omega_s + \Omega$, permitting us to view ω_1 as the (blue) pump-field and ω_2 as the (red) Stokes-field. We assume that the Raman interaction is given by $H_{\text{int}} = -\hbar(g a_p^\dagger a_s b + g^* a_p a_s^\dagger b^\dagger)$.

(a) Use number-state representation $|n_p, n_s, n_b\rangle$ to identify the initial and final states that are coupled by H_{int} . [Hint: Your answer should involve processes that create and destroy pump photons.]

If we only consider one scattering incident, then the following transitions may happen because of H_{int} :

$$|n_p, n_s, n_b\rangle \rightarrow |n_p - 1, n_s + 1, n_b + 1\rangle, \quad (42)$$

and

$$|n_p, n_s, n_b\rangle \rightarrow |n_p + 1, n_s - 1, n_b - 1\rangle. \quad (43)$$

When there is no existing Stokes photon in the system only the first transition happens.

(b) Assuming that your final optical state has a uniform density of states, $D(\omega) = D_o$, use Fermi's golden rule to find the rate with which pump photons scatter into the Stokes wave ($R_{p \rightarrow s}$). Similarly, find the rate with which Stokes photons are scattered back into the pump wave ($R_{s \rightarrow p}$).

Using Fermi's golden rule, we have

$$\begin{aligned} R_{p \rightarrow s} &= \frac{2\pi}{\hbar} |\langle n_p - 1, n_s + 1, n_b + 1 | H_{\text{int}} | n_p, n_s, n_b \rangle|^2 \delta(\hbar\omega_p - \hbar\omega_s - \hbar\Omega) \\ &= 2\pi |(-\hbar g^*) \sqrt{n_p} \sqrt{n_s + 1} \sqrt{n_b + 1}|^2 \delta(\omega_p - \omega_s - \Omega) \\ &= 2\pi \hbar^2 |g|^2 n_p (n_s + 1) (n_b + 1) D(\omega_p - \omega_s - \Omega) \\ &= 2\pi \hbar^2 |g|^2 n_p (n_s + 1) (n_b + 1) D_o. \end{aligned} \quad (44)$$

Here in the third line we have replaced the δ -function by the density of states $D(\omega)$, probably because there is some sort of dissipation in the system and there is a finite line width. Similarly

we have

$$\begin{aligned}
R_{s \rightarrow p} &= \frac{2\pi}{\hbar} |\langle n_p + 1, n_s - 1, n_b - 1 | H_{\text{int}} | n_p, n_s, n_b \rangle|^2 \delta(\hbar\omega_p - \hbar\omega_s - \hbar\Omega) \\
&= 2\pi |(-\hbar g) \sqrt{n_p + 1} \sqrt{n_s} \sqrt{n_b}|^2 \delta(\omega_p - \omega_s - \Omega) \\
&= 2\pi \hbar^2 |g|^2 (n_p + 1) n_s n_b D(\omega_p - \omega_s - \Omega) \\
&= 2\pi \hbar^2 |g|^2 (n_p + 1) n_s n_b D_0.
\end{aligned} \tag{45}$$

(c) Use your results from part (b) to formulate a rate-equation for the Stokes photon occupation number (n_s).

We have

$$\begin{aligned}
\frac{dn_s}{dt} &= R_{p \rightarrow s} - R_{s \rightarrow p} \\
&= 2\pi \hbar^2 |g|^2 D_0 (n_p (n_s + 1) (n_b + 1) - (n_p + 1) n_s n_b).
\end{aligned} \tag{46}$$

Interpretation of our results: Note that $R_{p \rightarrow s}$ and $R_{s \rightarrow p}$ contain several terms that represent different types of linear and nonlinear scattering processes. In what follows, we work to understand the significance of our results from parts (b-c).

(d) In the case when $n_s = 0, n_b = 0$, only spontaneous scattering is possible. Identify the rate with which pump photons scatter to the Stokes field from your expression for $R_{p \rightarrow s}$.

When $n_s = 0, n_b = 0$, we have

$$\frac{dn_s}{dt} = 2\pi \hbar^2 |g|^2 D_0 n_p. \tag{47}$$

The RHS is the rate with which pump photons scatter to the Stokes field.

(e) In the case when $n_s \neq 0, n_b = 0$, stimulated Stokes scattering becomes possible. Identify the rate of stimulated Stokes scattering from $R_{p \rightarrow s}$.

In this case we have

$$\frac{dn_s}{dt} = 2\pi \hbar^2 |g|^2 D_0 n_p n_s + 2\pi \hbar^2 |g|^2 D_0 n_p. \tag{48}$$

The second term is spontaneous Stokes scattering; the first term is stimulated Stokes scattering, and the rate is

$$\Gamma_{\text{stimulated Stokes}} = 2\pi \hbar^2 |g|^2 D_0 n_p n_s. \tag{49}$$

(f) In the case when $n_s \neq 0, n_b \neq 0$, it becomes possible for the Stokes photons to scatter back into the pump wave through both stimulated and spontaneous scattering processes. Identify the linear and nonlinear scattering rates that contribute to the depletion of the Stokes wave from $R_{s \rightarrow p}$.

In the most generic case we go back to

$$\frac{dn_s}{dt} = 2\pi \hbar^2 |g|^2 D_0 (n_p (n_s + 1) (n_b + 1) - (n_p + 1) n_s n_b). \tag{50}$$

The minus part of the RHS comes from $R_{s \rightarrow p}$; if the phonon part is seen as external to the degrees of freedom considered, then we have a linear scattering rate

$$\Gamma_{\text{back-scattering, 1}} = 2\pi \hbar^2 |g|^2 D_0 n_b n_s, \tag{51}$$

and a non-linear scattering rate

$$\Gamma_{\text{back-scattering, 1}} = 2\pi \hbar^2 |g|^2 D_0 n_b n_s n_p. \tag{52}$$

(g) In the limit when $n_p \gg 1$ and $n_b = 0$, what does \dot{n}_s reduce to? [Hint: Your result should be suggestive of amplification.] What happens to the Stokes wave amplification in the limit when $n_b \gg 1$?

When $n_p \gg 1$ and $n_b = 0$, we have

$$\frac{dn_s}{dt} = 2\pi \hbar^2 |g|^2 D_0 n_p (n_s + 1), \tag{53}$$

so we can see that n_s undergoes two types of amplification: an exponential amplification because of stimulated Stokes scattering, and a linear growth because of spontaneous Stokes scattering. Since n_b is small, back-scattering is not possible.

When $n_b \gg 1$, we have

$$\frac{dn_s}{dt} = 2\pi\hbar^2|g|^2 D_0 n_b (n_p - n_s). \quad (54)$$

When the number of pump photons is steady, n_s sees a linear growth in the long run, but at first a short exponential damping period may be seen. Intuitively, when the number of phonons is large, the phonon degree of freedom can be seen as a given external field, and H_{int} becomes a linear scattering process, with which a pump photon is turned into a Stokes photon, and vice versa. And thus (54) is consistent with the form of a classical Boltzmann equation describing scattering by disorders, where we see one “incoming” channel that’s proportional to n_p and an “out-going” channel that’s proportional to n_s ; since n_p almost doesn’t deplete, the first channel leads to n_b ’s steady growing, while the second term depletes n_s at $t = 0$.