

# PHYS2202 Nonlinear Optics

## Problem Set 4 solutions

### 1. (25 points) Electromagnetically induced transparency

Consider the three-level system represented by the energy diagram below. The excited state,  $b$ , is much higher in energy relative to the ground state than the thermal energy ( $\hbar\omega_{ba} \gg k_B T$ ) so that we can assume that at equilibrium  $\hat{\rho}^{(0)} = |a\rangle\langle a|$ . The system is characterized by homogeneous damping rates  $\Gamma_{ba}$ ,  $\Gamma_{ca}$ , and  $\Gamma_{cb}$ . Suppose that we use a weak probe pulse at frequency  $\omega_p = \omega_{ca} - \delta$  and a strong saturating pump pulse at frequency  $\omega_s = \omega_{cb} - \Delta$ . (In other words, the probe and pump beams are detuned from resonance by  $\delta$  and  $\Delta$  respectively.) Suppose, too, that the detunings are small compared to  $\omega_{ba}$ :  $\delta, \Delta \ll \omega_{ba}$ .

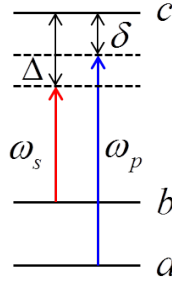


Figure 1: Energy level diagram showing probe (blue) and saturating pump (blue) detuned from the  $\omega_{ca}$  (probe) and  $\omega_{cb}$  (pump) by  $\delta$  and  $\Delta$ , respectively.

- (a) Using the density-matrix formalism, find the susceptibility associated with the first order *in the probe* response. This requires that we consider the response to infinite order in the pump intensity!

Hint: although we are discussing the frequency domain and so cannot say that one field is present before another, we are interested in the maximally resonant response. If using a diagrammatic approach, think about what that must mean for the ordering of the actual interactions (not simply when the fields are present but in what order they must interact). If using a non-diagrammatic approach, pay attention to which terms are fully resonant and which are not.

**Solution:** We are looking for the susceptibility that is linear in  $\mathcal{E}(\omega_p)$ :

$$\chi(\omega_p, \mathcal{E}(\omega_s)). \quad (1)$$

For dilute oscillators (e.g., a dilute atomic vapor), this is just

$$\chi^{(1)}(\omega_p, \mathcal{E}(\omega_s)) = \frac{P(\omega_p)}{\epsilon_0 \mathcal{E}(\omega_p)} = N \frac{\langle \vec{p}(\omega_p) \rangle}{\epsilon_0 \mathcal{E}(\omega_p)} = N \frac{\text{Tr}(\hat{\mathbf{p}} \hat{\rho}^{(1)}(\omega_p))}{\epsilon_0 \mathcal{E}(\omega_p)}. \quad (2)$$

The only element of the density matrix that will oscillate near its resonance frequency when driven at  $\omega_p$  is  $\rho_{ca}$ . Therefore, we are looking to calculate

$$\chi^{(1)}(\omega_p, \mathcal{E}(\omega_s)) = N \frac{\mu_{ac} \rho_{ca}^{(1)}(\omega_p)}{\epsilon_0 \mathcal{E}(\omega_p)}. \quad (3)$$

## The diagrammatic approach

We begin with the diagrammatic approach.

As usual, we will look first for the maximally resonant response, ideally a response that is resonant at every step. If there is a response that is resonant at every step, then we are done, since other terms are less strongly resonant and so should be much smaller and so can be neglected. Otherwise, we will have to look for a response that is resonant at every step but one, and so on.

Since the system starts at equilibrium in the ground state, the first interaction must be with the probe at  $\omega_p$ , since the field at  $\omega_s$  is not resonant with a transition involving the ground state. Since we are looking for a response that is linear in  $\mathcal{E}(\omega_p)$ , that first interaction will be the only interaction with the probe. It leaves the system in the  $\rho_{ca}$  coherence. (Since we are looking at the response at  $\omega_p$ , the interaction must involve either “absorption” on the left side of the density operator or “emission” on the right side, but the latter is non-resonant since the material system is starting in the ground state and so has nowhere lower in energy to go—again, in a resonant process.)

Once the system is in the  $\rho_{ca}$  coherence, the only interactions will be with the pump at  $\omega_s$ , but this field is only resonant with the  $c \rightarrow b$  transition. Therefore, the diagrammatic perturbation expansion of the response of interest (linear in  $\mathcal{E}(\omega_p)$ ) looks as in Figure 2.

$$P_{ca}(\omega_p) = \left( \text{Diagram 1} + \text{Diagram 2} + \dots \right) = \left( \text{Diagram 3} \right) \times \left( \text{Diagram 4} + \text{Diagram 5} + \dots \right) = \left( \text{Diagram 3} \right) \times \left( \text{Diagram 6} \right)^{-1}$$

Figure 2: Feynman diagrams describing electromagnetically induced transparency for the three-level system in Fig. 1.

The key point in the diagrammatic expansion is that we see that the only way to get full resonance is for all the interactions to occur on the left side of the density operator and for the pair of interactions with  $\mathcal{E}(\omega_s)$  and  $\mathcal{E}(-\omega_s) = \mathcal{E}^*(\omega_s)$  to occur repeatedly, each time leaving the system back in the  $\rho_{ca}$  coherence. This infinite sum can be calculated as long as the repeated element is less than unity. We just pull out the common first step in all the diagrams leaving us with a sum of the form  $1 + x + x^2 + \dots$ , which is simply the expansion of  $\frac{1}{1-x}$ .

The diagrams are easily translated. All interactions are associated with a factor of

$$-i/\hbar. \text{ After each interaction, there is a factor of } \left\{ -i \left[ \left( \sum_j \omega_j \right) - \omega_{nm} + i\Gamma_{mn} \right] \right\}^{-1},$$

where the sum is a sum of all the interactions that have occurred up to that point in the diagram,  $\omega_{nm}$  is the resonant frequency of the  $\rho_{nm}$  density matrix element, and  $\Gamma_{mn}$  is the decay rate of that element. (Alternatively, we can associate no factor, i.e.,

$$\text{a factor of 1, with each interaction and a factor of } \left\{ \hbar \left[ \left( \sum_j \omega_j \right) - \omega_{nm} + i\Gamma_{mn} \right] \right\}^{-1}$$

after each interaction) For every interaction on the right hand side of the density operator there is a factor of  $-1$ . All of these rules come straight from the Liouville

equation. Translation of the final formulation of the diagrams then yields

$$\rho_{ca}(\omega_p) = \left(-\frac{i}{\hbar}\right) \frac{1}{-i(\omega_p - \omega_{ca} + i\Gamma_{ca})} H_{I,ca}(\omega_p) \rho_{aa}^{\text{eq}} \quad (4)$$

$$\times \left[ 1 - \left(-\frac{i}{\hbar}\right)^2 \frac{1}{-i(\omega_p - \omega_{ca} + i\Gamma_{ca})} H_{I,cb}(\omega_s) \frac{1}{-i(-\omega_s + \omega_p - \omega_{ba} + i\Gamma_{ba})} H_{I,bc}(-\omega_s) \right]^{-1}$$

At equilibrium,  $\rho_{mn}^{\text{eq}} = \delta_{ma}\delta_{na}$ , i.e., only  $\rho_{aa} = 1$  is non-zero. Defining  $\delta = \omega_p - \omega_{ca}$  (the opposite sign of the problem statement) and  $\Delta = \omega_s - \omega_{cb}$  (again the opposite sign from the problem statement) and noting that  $\omega_p - \omega_s - \omega_{ba} = \omega_{ca} + \delta - (\omega_{cb} + \Delta) - \omega_{ba} = \delta - \Delta$ , we can write

$$\rho_{ca}(\omega_p) = -\frac{\Omega_{ca}(\omega_p)}{\delta + i\Gamma_{ca}} \left[ 1 - \frac{|\Omega_{cb}(\omega_s)|^2}{(\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})} \right]^{-1} \quad (5)$$

$$= \frac{(\delta - \Delta + i\Gamma_{ba})}{|\Omega_{cb}(\omega_s)|^2 - (\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})} \Omega_{ca}(\omega_p). \quad (6)$$

The susceptibility at  $\omega_p$  associated with the linear response to  $\mathcal{E}(\omega_p)$  is then

$$\chi^{(1)}(\omega_p, \mathcal{E}(\omega_s)) = -\frac{N |\vec{\mu}_{ac}|^2}{\epsilon_0 \hbar (\delta + i\Gamma_{ca})} \left[ 1 - \frac{|\Omega_{cb}(\omega_s)|^2}{(\delta_p + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})} \right]^{-1} \quad (7)$$

$$= \frac{N |\vec{\mu}_{ac}|^2}{\hbar \epsilon_0} \frac{(\delta - \Delta + i\Gamma_{ba})}{|\Omega_{cb}(\omega_s)|^2 - (\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})}. \quad (8)$$

## The Liouville equation approach

This approach is fundamentally the same, since it si from the Liouville equation that we developed the double sided Feynman diagrams. We will call this the “Liouville equation approach” simply because it makes a more explicit step-by-step approach invoking the Liouville equation at each step. This is a much more tedious approach, and we only get the big picture at the end.

The Liouville equation is

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar} [\hat{H}_0, \hat{\rho}] - \frac{i}{\hbar} [\hat{H}_I, \hat{\rho}] + \left( \frac{\partial \hat{\rho}}{\partial t} \right)_{\text{random}}. \quad (9)$$

Expressed in terms of Fourier components, the corresponding equations of motion for the elements of the density matrix are

$$-i\omega \tilde{\rho}_{mn}(\omega) = -\frac{i}{\hbar} [\hat{H}_0, \hat{\rho}(\omega)]_{mn} - \frac{i}{\hbar} \sum_j [\hat{H}_I(\omega_j), \hat{\rho}(\omega - \omega_j)]_{mn} - \Gamma_{mn} \tilde{\rho}_{mn}(\omega), \quad (10)$$

where the sum is over all frequencies (positive and negative) present in the interaction Hamiltonian. This can be rewritten as

$$-i\hbar(\omega - \omega_{mn} + i\Gamma_{mn}) \tilde{\rho}_{mn}(\omega) = -\frac{i}{\hbar} \sum_j [\hat{H}_I(\omega_j), \hat{\rho}(\omega - \omega_j)]_{mn}, \quad (11)$$

from which the rules of translation for our Feynman diagrams should be clear.

We need to calculate  $\rho_{ca}(\omega_p)$ . The only frequencies in our input beams are  $\pm\omega_p$  and  $\pm\omega_s$ . Since we are looking for  $\rho_{ca}(\omega_p)$  that is linear in  $\mathcal{E}(\omega_p)$ , the term  $\mathcal{E}(-\omega_p) = \mathcal{E}^*(\omega_p)$  (i.e.,  $H_I(-\omega_p)$ ) cannot appear; there will be no terms involving  $-\omega_p$ . The equation of motion for  $\rho_{ca}(\omega_p)$  is

$$\begin{aligned} & -i(\omega_p - \omega_{ca} + i\Gamma_{ca}) \rho_{ca}(\omega_p) \\ &= -\frac{i}{\hbar} \{ [H_{I,ca}(\omega_p)\rho_{aa}(0) + H_{I,ca}(\omega_s)\rho_{aa}(\omega_p - \omega_s) + H_{I,ca}(-\omega_s)\rho_{aa}(\omega_p + \omega_s) \\ & \quad + H_{I,cb}(\omega_p)\rho_{ba}(0) + H_{I,cb}(\omega_s)\rho_{ba}(\omega_p - \omega_s) + H_{I,cb}(-\omega_s)\rho_{ba}(\omega_p + \omega_s)] \\ & \quad - [\rho_{cc}(0)H_{I,ca}(\omega_p) + \rho_{cc}(\omega_p - \omega_s)H_{I,ca}(\omega_s) + \rho_{cc}(\omega_p + \omega_s)H_{I,ca}(-\omega_s) \\ & \quad + \rho_{cb}(0)H_{I,ba}(\omega_p) + \rho_{cb}(\omega_p - \omega_s)H_{I,ba}(\omega_s) + \rho_{cb}(\omega_p + \omega_s)H_{I,ba}(-\omega_s)] \}. \end{aligned} \quad (12)$$

We can immediately disregard many of these terms because they are nonresonant.  $\rho_{aa}$  and  $\rho_{cc}$  are populations and so “oscillate” at zero frequency. **Therefore, we can disregard all terms  $\rho_{aa}(\omega)$  and  $\rho_{cc}(\omega)$  where  $\omega \neq 0$  as non-resonant.** Similarly, all coherences should oscillate at non-zero frequency, so we can disregard the terms  $\rho_{ba}(0)$  and  $\rho_{ca}(0)$  as non-resonant.  $\rho_{ba}$  has a natural (resonant) frequency at  $\omega_{ba}$ , but  $\omega_p + \omega_s$  is far from  $\omega_b$ , so we can disregard  $\rho_{ba}(\omega_p + \omega_s)$ . For the same reason, we can disregard  $\rho_{cb}(\omega_p - \omega_s)$  and  $\rho_{cb}(\omega_p + \omega_s)$ , since  $\rho_{cb}$  is resonant at  $\omega_c \sim \omega_p$ . The fully resonant terms in the equation of motion for  $\rho_{ca}(\omega_p)$  reduce to

$$\begin{aligned} \hbar(\omega_p - \omega_{ca} + i\Gamma_{ca}) \rho_{ca}(\omega_p) &= H_{I,ca}(\omega_p)\rho_{aa}(0) + H_{I,cb}(\omega_s)\rho_{ba}(\omega_p - \omega_s) \\ & \quad - \rho_{cc}(0)H_{I,ca}(\omega_p). \end{aligned} \quad (13)$$

There is one other very important *set* of constraints on the terms that can appear in our equations of motion:

- i. The process must be resonant at every interaction step;
- ii. The system starts in the ground state, i.e.,  $\rho_{mn}^{\text{eq}} = \delta_{ma}\delta_{na}$ , where  $\delta_{ij}$  is the Kronecker delta function;
- iii. The only resonant interaction involving the ground state  $a$  is via  $\omega_p$ , which can only appear once in the entire process.
- iv. We are looking for a process at frequency  $\omega_p$ .

The preceding constraints mean that the one interaction at  $\omega_p$  occurs on the left side of the density operator and that the right side of the density operator must always remain as  $a$ , i.e., the only non-zero terms at any frequency are  $\rho_{aa}$ ,  $\rho_{ba}$ , and  $\rho_{ca}$ . Therefore, the equation of motion for  $\rho_{ca}(\omega_p)$  becomes a little simpler, since we will neglect the  $\rho_{cc}(0)$  term:

$$\hbar(\omega_p - \omega_{ca} + i\Gamma_{ca}) \rho_{ca}(\omega_p) = H_{I,ca}(\omega_p)\rho_{aa}(0) + H_{I,cb}(\omega_s)\rho_{ba}(\omega_p - \omega_s). \quad (14)$$

To solve for  $\rho_{ca}(\omega_p)$  we see that we first need to solve the equations of motion for  $\rho_{aa}(0)$  and  $\rho_{ba}(\omega_p - \omega_s)$ .

$\rho_{aa}(0)$  is easy. Since only  $\omega_p$  is near resonance with the  $ac$  transition and only one interaction at frequency  $\omega_p$  takes place,  $\rho_{aa}(0)$  can only be the equilibrium value  $\rho_{aa}(0) = \rho_{aa}^{\text{eq}} = 1$ .

Let's calculate  $\rho_{ba}(\omega_p - \omega_s)$ . The equation of motion is

$$-i[(\omega_p - \omega_s) - \omega_{ba} + i\Gamma_{ba}] \rho_{ba}(\omega_p - \omega_s) = -\frac{i}{\hbar} \sum_j \left[ \hat{H}_I(\omega_j), \hat{\rho}(\omega_p - \omega_s - \omega_j) \right]_{ba} \quad (15)$$

or

$$\begin{aligned}
\hbar(\omega_p - \omega_s - \omega_{ba} + i\Gamma_{ba})\rho_{ba}(\omega_p - \omega_s) &= \sum_j \left[ \hat{H}_I(\omega_j), \hat{\rho}(\omega_p - \omega_s - \omega_j) \right]_{ba} \\
&= H_{I,bc}(\omega_p) \rho_{ca}(-\omega_s) + H_{I,bc}(\omega_s) \rho_{ca}(\omega_p - 2\omega_s) + H_{I,bc}(-\omega_s) \rho_{ca}(\omega_p) \\
&\quad + H_{I,ba}(\omega_p) \rho_{aa}(-\omega_s) + H_{I,ba}(\omega_s) \rho_{aa}(\omega_p - 2\omega_s) + H_{I,ba}(-\omega_s) \rho_{aa}(\omega_p) \\
&\quad - \underline{\rho_{bb}(-\omega_s)} H_{I,ba}(\omega_p) - \underline{\rho_{bb}(\omega_p - 2\omega_s)} H_{I,ba}(\omega_s) - \underline{\rho_{bb}(\omega_p)} H_{I,ba}(-\omega_s) \\
&\quad - \underline{\rho_{bc}(-\omega_s)} H_{I,ca}(\omega_p) - \underline{\rho_{bc}(\omega_p - 2\omega_s)} H_{I,ca}(\omega_s) - \underline{\rho_{bc}(\omega_p)} H_{I,ca}(-\omega_s) \\
&= H_{I,bc}(-\omega_s) \rho_{ca}(\omega_p).
\end{aligned} \tag{16}$$

Terms in red are non-resonant, and underlined terms are discarded because they are not of the form  $\rho_{na}$  (they are also all non-resonant except for  $\rho_{bc}(-\omega_s)$ ). Thankfully, no new terms were introduced. We already found an equation for  $\rho_{ca}(\omega_p)$ , so we now have a closed set of equations, which we summarize here:

$$\rho_{ca}(\omega_p) = \frac{H_{I,ca}(\omega_p) \rho_{aa}(0) + H_{I,cb}(\omega_s) \rho_{ba}(\omega_p - \omega_s)}{\hbar(\omega_p - \omega_{ca} + i\Gamma_{ca})} \tag{18}$$

$$\rho_{aa}(0) = \rho_{aa}^{\text{eq}} = 1 \tag{19}$$

$$\rho_{ba}(\omega_p - \omega_s) = \frac{H_{I,bc}(-\omega_s) \rho_{ca}(\omega_p)}{\hbar(\omega_p - \omega_s - \omega_{ba} + i\Gamma_{ba})} \tag{20}$$

These yield a closed solution for  $\rho_{ca}(\omega_p)$ :

$$\rho_{ca}(\omega_p) = \frac{1}{\hbar(\omega_p - \omega_{ca} + i\Gamma_{ca})} \left[ H_{I,ca}(\omega_p) \rho_{aa}^{\text{eq}} + \frac{|H_{I,cb}(\omega_s)|^2 \rho_{ca}(\omega_p)}{\hbar(\omega_p - \omega_s - \omega_{ba} + i\Gamma_{ba})} \right] \tag{21}$$

$$= \frac{1}{(\omega_p - \omega_{ca} + i\Gamma_{ca})} \left[ -\Omega_{ca}(\omega_p) \rho_{aa}^{\text{eq}} + \frac{|\Omega_{cb}(\omega_s)|^2 \rho_{ca}(\omega_p)}{(\omega_p - \omega_s - \omega_{ba} + i\Gamma_{ba})} \right], \tag{22}$$

where  $\Omega_{nm}(\omega) \equiv \vec{\mu}_{mn} \cdot \mathcal{E}(\omega)/\hbar$  is the Rabi frequency associated with the field at  $\omega$  and the  $m \leftrightarrow n$  transition. Changing to a standard sign convention,  $\delta = \omega_p - \omega_{ca}$  and  $\Delta = \omega_s - \omega_{cb}$ , we can write  $\omega_p - \omega_s - \omega_{ba} = \omega_{ca} + \delta - (\omega_{cb} + \Delta) - \omega_{ba} = \delta - \Delta$ , and

$$\rho_{ca}(\omega_p) = \frac{1}{(\delta + i\Gamma_{ca})} \left[ -\Omega_{ca}(\omega_p) \rho_{aa}^{\text{eq}} + \frac{|\Omega_{cb}(\omega_s)|^2 \rho_{ca}(\omega_p)}{(\delta - \Delta + i\Gamma_{ba})} \right]. \tag{23}$$

This is rewritten by bringing both  $\rho_{ca}(\omega_p)$  terms to the same side, so that:

$$\rho_{ca}(\omega_p) = -\frac{\Omega_{ca}(\omega_p)}{\delta + i\Gamma_{ca}} \left[ 1 - \frac{|\Omega_{cb}(\omega_s)|^2}{(\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})} \right]^{-1} \tag{24}$$

$$= \frac{(\delta - \Delta + i\Gamma_{ba}) \Omega_{ca}(\omega_p)}{|\Omega_{cb}(\omega_s)|^2 - (\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})}, \tag{25}$$

Exactly as found in Eq. 5.

- (b) Assume that  $\Gamma_{ba} = 0.01\Gamma_{ca} = 0.01\Gamma_{cb}$ . Plot the linear absorption coefficient as a function of probe frequency,  $\omega_s$  for  $\Delta = 0$  in the cases

- i.  $\Omega_s = 0$  (this is just the normal linear absorption coefficient)

The linear absorption coefficient is given by twice the imaginary part of the wave vector:

$$\alpha(\omega_p) = 2\text{Im}(\chi(\omega_p)) = 2\frac{\omega}{c}\text{Im}(\sqrt{\epsilon_r}), \quad (26)$$

where  $\epsilon_r(\omega_p) = 1 + \chi(\omega_p)$  is the relative susceptibility. For  $\chi(\omega_p) \ll 1$ ,

$$\sqrt{\epsilon_r} \approx 1 + \frac{1}{2}\chi(\omega_p). \quad (27)$$

The absorption coefficient is then

$$\alpha(\omega_p) = \frac{\omega}{c}\text{Im}[\chi(\omega_p)] \quad (28)$$

In the case that  $\Omega_{cb}(\omega_s) = 0$ ,

$$\begin{aligned} \chi^{(1)}(\omega_p, \mathcal{E}(\omega_s)) &= -\frac{N |\vec{\mu}_{ac}|^2}{\hbar\epsilon_0} \frac{(\delta - \Delta + i\Gamma_{ba})}{(\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})} \\ &= -\frac{N |\vec{\mu}_{ac}|^2}{\hbar\epsilon_0} \frac{1}{(\delta + i\Gamma_{ca})}. \end{aligned} \quad (29)$$

The absorption coefficient is then just

$$\alpha(\omega_p) = \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{\Gamma_{ca}}{\delta^2 + \Gamma_{ca}^2}. \quad (30)$$

The absorption coefficient is just a Lorentzian line (we are dealing with a homogeneously broadened system) of half-width at half-maximum (HWHM)  $\Gamma_{ca}$ .

- ii.  $\Omega_s \equiv \left| \frac{\vec{\mu}_{cb} \cdot \vec{E}_s}{\hbar} \right| = 0.5\Gamma_{ca}$

**Solution:** In the more general case, it is easiest to just plot the result with software. However, the general result for the susceptibility is

$$\begin{aligned} \chi^{(1)}(\omega_p, \mathcal{E}(\omega_s)) &= \frac{N |\vec{\mu}_{ac}|^2}{\hbar\epsilon_0} \frac{(\delta - \Delta + i\Gamma_{ba})}{|\Omega_{cb}(\omega_s)|^2 - (\delta + i\Gamma_{ca})(\delta - \Delta + i\Gamma_{ba})} \\ &= \frac{N |\vec{\mu}_{ac}|^2}{\hbar\epsilon_0} \frac{(\delta - \Delta + i\Gamma_{ba})}{\left[ |\Omega_{cb}(\omega_s)|^2 - \delta(\delta - \Delta) + \Gamma_{ca}\Gamma_{ba} \right] - i[(\delta - \Delta)\Gamma_{ca} + \delta\Gamma_{ba}]} \\ &= \frac{N |\vec{\mu}_{ac}|^2}{\hbar\epsilon_0} \frac{(\delta - \Delta + i\Gamma_{ba})}{\left[ |\Omega_{cb}(\omega_s)|^2 - \delta(\delta - \Delta) + \Gamma_{ca}\Gamma_{ba} \right]^2 + [(\delta - \Delta)\Gamma_{ca} + \delta\Gamma_{ba}]^2} \\ &\quad \times \left\{ \left[ |\Omega_{cb}(\omega_s)|^2 - \delta(\delta - \Delta) + \Gamma_{ca}\Gamma_{ba} \right] + i[(\delta - \Delta)\Gamma_{ca} + \delta\Gamma_{ba}] \right\}. \end{aligned} \quad (31)$$

The absorption coefficient is then

$$\begin{aligned} \alpha(\omega_p) &= \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{(\delta - \Delta) [(\delta - \Delta)\Gamma_{ca} + \delta\Gamma_{ba}] + \Gamma_{ba} \left[ |\Omega_{cb}(\omega_s)|^2 - \delta(\delta - \Delta) + \Gamma_{ca}\Gamma_{ba} \right]}{\left[ |\Omega_{cb}(\omega_s)|^2 - \delta(\delta - \Delta) + \Gamma_{ca}\Gamma_{ba} \right]^2 + [(\delta - \Delta)\Gamma_{ca} + \delta\Gamma_{ba}]^2} \\ &= \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{\Gamma_{ba} \left[ |\Omega_{cb}(\omega_s)|^2 + \Gamma_{ca}\Gamma_{ba} \right] + (\delta - \Delta)^2 \Gamma_{ca}}{\left[ |\Omega_{cb}(\omega_s)|^2 - \delta(\delta - \Delta) + \Gamma_{ca}\Gamma_{ba} \right]^2 + [(\delta - \Delta)\Gamma_{ca} + \delta\Gamma_{ba}]^2} \end{aligned} \quad (32)$$

To get an idea of what happens as we tune  $\delta$ , consider the case of  $\Gamma_{ba}, \Omega_{cb} \ll \Gamma_{ca}$ . In that case, if  $\delta - \Delta \gtrsim \Omega_{cb}, \Gamma_{ba}$ ,

$$\begin{aligned}\alpha(\omega_p) &\approx \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{(\delta - \Delta)^2 \Gamma_{ca}}{[\delta(\delta - \Delta)]^2 + [(\delta - \Delta)\Gamma_{ca}]^2} \\ &= \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{\Gamma_{ca}}{\delta^2 + \Gamma_{ca}^2}.\end{aligned}$$

The absorption spectrum then looks just like the absorption spectrum in the absence of the pump. However, as we decrease the magnitude of  $\delta - \Delta$ , we find that the absorption is reduced. At  $\delta = \Delta$  we find the absorption coefficient in the general case to be

$$\begin{aligned}\alpha(\omega_p) &= \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{\Gamma_{ba} \left[ |\Omega_{cb}(\omega_s)|^2 + \Gamma_{ca} \Gamma_{ba} \right]}{\left[ |\Omega_{cb}(\omega_s)|^2 + \Gamma_{ca} \Gamma_{ba} \right]^2 + [\delta \Gamma_{ba}]^2} \\ &= \frac{N |\vec{\mu}_{ac}|^2 \omega_p}{\hbar c \epsilon_0} \frac{\left[ \frac{|\Omega_{cb}(\omega_s)|^2}{\Gamma_{ba}} + \Gamma_{ca} \right]}{\left[ \frac{|\Omega_{cb}(\omega_s)|^2}{\Gamma_{ba}} + \Gamma_{ca} \right]^2 + \delta^2}.\end{aligned}\quad (33)$$

Two features stand out:

- A. The absorption is *reduced* from its value in the absence of the pump. At  $\delta = 0$ , instead of a value of proportional to  $\Gamma_{ca}^{-1}$ , it takes on a value  $\left( \frac{|\Omega_{cb}(\omega_s)|^2}{\Gamma_{ba}} + \Gamma_{ca} \right)^{-1}$ . For  $\Omega_{cb}^2(\omega_s) \gg \sqrt{\Gamma_{ca} \Gamma_{cb}}$ , the absorption is greatly reduced.
- B. The resulting dip in absorption is characterized by a width that increases with  $\Omega_{cb}(\omega_s)$ .

For  $\Gamma_{ba} = 0.01\Gamma_{ca}$  and  $\Omega_{cb}(\omega_s) = 0.5\Gamma_{ca}$ , we see that for  $\delta = 0$ ,

$$\alpha(\omega_p) = \frac{\omega}{c} \frac{N |\vec{\mu}_{ac}|^2}{\epsilon_0} \frac{0.01}{0.26\Gamma_{ca}} = \frac{1}{26} \alpha(\omega_p, \Omega_{cb}(\omega_s) = 0). \quad (34)$$

iii.  $\Omega_s \equiv \left| \frac{\vec{\mu}_{cb} \cdot \vec{E}_s}{\hbar} \right| = 5\Gamma_{ca}$

**Solution:** In this case the absorption at  $\delta = 0$  is only

$$\alpha(\omega_p) = \frac{\omega}{c} \frac{N |\vec{\mu}_{ac}|^2}{\epsilon_0} \frac{0.01}{25.01\Gamma_{ca}} = 4 \times 10^{-4} \alpha(\omega_p, \Omega_{cb}(\omega_s) = 0). \quad (35)$$

Moreover, the absorption is low over a wide spectral range (wide compared to the homogeneous linewidths  $\Gamma_{ca}, \Gamma_{ba}$ ).

The absorption spectra for the cases of  $\Delta = 0$  for  $\Omega_{cb}(\omega_s) = 0, 0.5\Gamma_{ca}$ , and  $5\Gamma_{ca}$  are shown in Fig. 3.

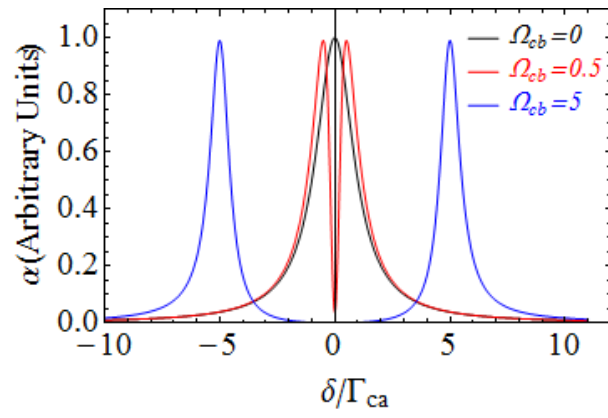


Figure 3: Absorption spectra in arbitrary units for  $\Omega_{cb} = 0$  (black),  $0.5\Gamma_{ca}$  (red), and  $5\Gamma_{ca}$  (blue).