PHYS2202 Nonlinear Optics
Problem Set 3
Due at the beginning of class on Tuesday, March 31, 2020

1. **(10 points) Second order density matrix**
   In class, we walked through the calculation of the second order density matrix ($\rho^{(2)}$).
   However, we made a mistake (OK, I made a mistake) in the moment and got zero, which
   I said was the wrong result. The error that I made was simply that I didn't look at every
   density matrix element separately; I basically added them together. Here, you will correct
   that mistake and find the exact density matrix $\rho^{(2)}(t)$.

   As in class, we will make this as simple as possible:

   (a) We will consider a two-level system of sub-wavelength spatial extent (so that we can
   ignore propagation effects).

   (b) We will excite the system impulsively (so that our time integrals are as simple as
   possible).

   (c) The excitation will be by a pair of co-linearly propagating pulses. (OK, we could
   have made this simpler and used only a single pulse, but that would be a little too
   simple.)

   (d) We will also neglect all damping.

   In other words, our system has only two eigenstates of the unperturbed Hamiltonian, $\hat{H}_0$,
   which we will label $|1\rangle$, the ground state, and $|2\rangle$, the excited state. We will call the
   unperturbed ground state energy E_1, and the excited state energy E_2. The excitation
   field at the material system is given by $\vec{E}(t) = \hat{z}\mathscr{E}\left[\delta\left(t - \tau_a - \frac{\vec{k}\cdot\vec{x}}{\omega_{21}}\right) + \delta\left(t - \tau_b - \frac{\vec{k}\cdot\vec{x}}{\omega_{21}}\right)\right]$.
   (This may look like an unusual wave, but while it is somewhat artificial, you should see
   that it is just a pair of delta function pulses propagating in the same direction.)

   Please remember that, unless we specify otherwise, we work in the dipole approximation,
   and our interaction Hamiltonian has no diagonal elements in the basis of the eigenstates
   of the unperturbed Hamiltonian.

   Assume that initially, the system is in its ground state, i.e., $\rho(t < 0) = |1\rangle\langle 1|$ or

   $$\begin{pmatrix} \rho_{11}(t) & \rho_{12}(t) \\ \rho_{21}(t) & \rho_{22}(t) \end{pmatrix} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}. \tag{1}$$

   Your tasks are the following.

   (a) Write the density matrix (i.e., write the matrix representation of the density operator
   in the $(1,0), (0,1)$ basis) for all times $t > \tau$. Just write the part that depends on the
   time difference $\tau_b - \tau_a$, i.e., the part that depends on interactions with both of the
   pulses making up the exciting field.

   (b) Write the value of the expectation value of the dipole moment operator at $t > \tau$?

2. **(20 points) The third-order density matrix** We now go two steps further than in the
   previous problem. We will consider the next order in the density matrix, and we will excite
   the system with three electromagnetic pulses that are traveling in different directions:

   $$\vec{E}(t) = \hat{z}\mathscr{E}_0\left[\delta\left(t - \tau_a - \frac{\vec{k}_a \cdot \vec{x}}{\omega_{21}}\right) + \delta\left(t - \tau_b - \frac{\vec{k}_b \cdot \vec{x}}{\omega_{21}}\right) + \delta\left(t - \tau_c - \frac{\vec{k}_c \cdot \vec{x}}{\omega_{21}}\right)\right].$$

We are interested in the third-order contribution to the optical response. This will require that we calculate the third-order density matrix. As we saw in our treatment of a classical oscillator, when we have more than just a single monochromatic wave, the number of new frequencies can rapidly expand. If we include pulses in different directions, the same happens: we get new pulses characterized with wave vectors in different directions than any that were present in the input pulses. However, different frequencies and wave vectors are easy to distinguish. To separate frequencies we can use spectrometers. To separate wave vectors characterized by different directions, we can just place our detector in the path of the direction of propagation that interests us. Here, we will focus on a single direction, that given by $\vec{k}_c + \vec{k}_b - \vec{k}_a$. This will dramatically reduce the number of processes that we have to consider, since combined with the impulsive nature of our excitation it automatically chooses a small subset of interactions with the various delta function pulses.

Your tasks are the following:

(a) Calculate the entire density matrix (but only that part characterized by the wave vector specified above) at times $t > \tau_c + \frac{\vec{k}_c \cdot \vec{x}}{\omega_{21}}$.

(b) Write down the density matrix at time t given by $t - \tau_c = \tau_b - \tau_a$.

(c) Suppose now that we have an ensemble of almost (but not quite) identical oscillators characterized by a continuous distribution, $g(\omega_{21})$, of resonant frequencies ω_{21}, where the width of the distribution $g(\omega_{21})$ is $\Delta\omega_{21}$. The density matrix must now be determined by averaging over all the frequencies.

   i. Write down the density matrix at time $(t - \tau_c) - (\tau_b - \tau_a) >> \frac{1}{\Delta\omega_{21}}$.

   ii. Write the density matrix at time $t - \tau_c = \tau_b - \tau_a$.

   Note that you do not need to know the exact distribution $g(\omega_{21})$. We are not looking for a precise answer, just a reasonable estimate. If you really want a distribution, you can assume a Gaussian distribution.