

Exam Nonlinear Optics

Wednesday, April 23, 2008, 9.00-12.00 (room 5113.0201)

Give your name on each sheet.

On the first sheet, also give your student number
and the total number of sheets turned in.

Success!

Problem 1 (6 points)

Consider a material consisting of one-dimensional classical anharmonic oscillators with a quadratic force term $-m\alpha x^2$ as nonlinearity. The density of oscillators is denoted \tilde{N} . Assume that a c.w. light beam is incident: $\vec{E} = E \exp[-i\omega t] + \text{c.c.}$

Derive the susceptibilities: $\chi^{(1)}(\omega)$, $\chi^{(2)}(2\omega)$, and $\chi^{(3)}(3\omega)$. Explain the quantities you introduce.

Problem 2 (5 points)

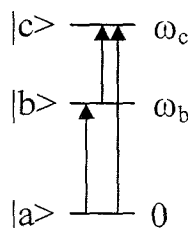
Consider a sum-frequency generation process.

- If both fundamental (incoming) pulses are assumed undepleted, how does the signal intensity then depend on the length L of the sample? (Calculation is allowed, but not necessary.)
- An important factor in this dependence is the wavevector mismatch. How is this quantity defined? Explain physically why it determines the signal intensity?
- Now assume that one of the fundamental pulses is depleted, while the other is not. Draw the intensity of all pulses as a function of propagation distance in the sample for zero wavevector mismatch. How would your drawing change if we increase $\chi^{(2)}$ by a factor of 2?

Problem 3 (8 points)

Consider the molecular energy level diagram below, with three levels of interest. The dephasing rate of the coherence $|a\rangle\langle b|$ is given by γ_{ab} , and analogous for the other coherences. None of the levels has a permanent dipole.

- Show by general consideration of the structure of Feynman diagrams that all three transitions indicated by arrows should be dipole-allowed (i.e., have a nonzero dipole matrix element) in order for this system to have a second-order optical response.
- We are interested in sum-frequency generation, with incoming fields of frequencies ω_1 and ω_2 . Let us assume that $\omega_1 \approx \omega_b$ and $\omega_2 \approx \omega_{cb}$. Show that at least one Feynman diagram exists that gives a double resonance; draw this diagram.
- Calculate the diagram which you have found in b). In case you did not find a diagram with the desired property, give an arbitrary other diagram which contributes to SFG in this system and calculate it.
- Is the assumed arrangement of nonzero transition dipoles allowed for a centrosymmetric system? Motivate your answer.



Problem 4 (7 points)

Consider an ensemble of two-level molecules in a host medium. As a consequence of inhomogeneity in the host, each molecule has a slightly different transition frequency. These frequencies are distributed according to a Gaussian distribution with mean ω_0 and standard deviation D . The coherence between the ground state and the excited state of each molecule decays exponentially with the same homogeneous dephasing rate γ .

- a) Give an expression for the absorption spectrum of the ensemble in the limit $D \gg \gamma$. Same question if $D \ll \gamma$. (Note: No derivation is needed. You do not need to give all prefactors; the frequency dependence is the aspect of interest.)
- b) What is free-induction decay (also known as inhomogeneous dephasing)? What is the physical explanation? What is the timescale for free-induction decay in the above example? (No derivation needed.)
- c) Explain how one can use the two-pulse echo experiment to measure the homogeneous dephasing rate, even if $D \gg \gamma$. You do not need to give a complete calculation; a clear analysis of phase factors and relaxation factors suffices.