

## Problem Set PS06

ISSUED: 2/17/99 Due: 2/24/00

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Name \_\_\_\_\_

**Instructions.** Complete all questions before class on the due date. You are encouraged to work together. Be sure to struggle with the problem before seeking help. Many of the exercises are very similar to problems in the book. Understanding the solution to these problems will be helpful in completing the assigned exercises.

### Exercises

1. Three distinct laser beams are used to perform a four wave mixing experiment. For beam 1,  $\omega = 20000\text{cm}^{-1}$ ; for beam 2,  $\omega = 20000\text{cm}^{-1}$ ; and for beam 3,  $\omega = 19000\text{cm}^{-1}$ . What is the signal frequency along the following signal wavevectors. Identify the signals that would be considered nearly degenerate four wave mixing (that is, the signal frequency is close to the fundamental frequencies).

(a)  $\vec{k}_{sig} = \vec{k}_1 + \vec{k}_2 + \vec{k}_3$

(b)  $\vec{k}_{sig} = \vec{k}_1 - \vec{k}_2 + \vec{k}_3$

(c)  $\vec{k}_{sig} = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$

(d)  $\vec{k}_{sig} = -\vec{k}_1 + \vec{k}_2 + \vec{k}_3$

2. The coherent Raman scattering (CRS) spectroscopies are an important class of nearly degenerate four wave mixing processes. In a typical CRS experiment two fundamental beams (beams 1 and 2) are identical and a third beam (beam 3) is chosen such the difference between frequencies of beam 1 or 2 and that of beam 3 matched a vibrational transition of the sample. When the beams are in the BOX beam geometry, the CRS signal emerges along the wavevector  $\vec{k}_{sig} = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$ .

(a) Draw the beam configuration for a CRS experiment.

(b) There are two types of CRS experiments i) CARS (coherent anti-Stokes Raman scattering) and CSRS (coherent Stokes Raman scattering). Based on what you know to be the definition of Stokes scattering, How do these two experiments differ? Draw a spectrum which shows the fundamental and signal beam frequencies.

(c) Say you wanted to do a CARS experiment to study the ring breathing mode of benzene ( $992\text{cm}^{-1}$ ). Which dyes would you use for your laser beams (see figure below)? Where in frequency would your signal come out?

(d) Say you wanted to do a CARS experiment to study the C-H symmetric stretching mode of benzene ( $3063\text{cm}^{-1}$ ). Which dyes would you use for your laser beams (see figure below)? Where in frequency would your signal come out?

(e) Say you wanted to do a CSRS experiment to study the ring breathing mode of benzene ( $992\text{cm}^{-1}$ ). Which dyes would you use for your laser beams (see figure below)? Where in frequency would your signal come out?

- (f) Say you wanted to do a CSRS experiment to study the C–H symmetric stretching mode of benzene ( $3063\text{cm}^{-1}$ ). Which dyes would you use for your laser beams (see figure below)? Where in frequency would your signal come out?

3. Read sections 1.9 and 1.11 of Laidler & Meiser and work problems 1.31, 1.36, 1.37, 1.39, 1.41 and 1.45

### Conceptual Problems

4. How does a laser work?
5. Use the Boltzmann distribution to show that no matter how hot it gets an ensemble of two level systems in thermal equilibrium will never have a larger population in the excited state than in the ground state. (This is a very important concept to understand because we must get around this to make a laser work).
6. In the previous problem you showed that an ensemble of two level systems in thermal equilibrium can never have what is called *population inversion* (a higher population in the excited state than in the ground state). One sometimes sees in the literature authors saying that an ensemble has “negative temperature” (a temperature below absolute zero) when a population inversion situation exists. Use the Boltzmann distribution to explain why they might use this phrase.

