Problem 1. Two optical beams $E_1$ and $E_2$ with wavelengths of 1.0 and 0.6 $\mu$m respectively are incident on a nonlinear material.

(a) Assuming a $\chi^{(2)}$ nonlinearity, what new wavelengths can possibly be generated in this material?

The above nonlinear material is now replaced with a centro-symmetric material for the remaining part of this problem.

(b) What is the dominant nonlinear susceptibility?

(c) Assuming $\chi^{(3)}$ nonlinearity, what new wavelengths $\lambda_j$ can possibly be generated in this material that simultaneously involve the interaction of both $E_1$ and $E_2$ beams? Write down the corresponding nonlinear polarization $P(\lambda_j)$ including their $\chi^{(3)}(\lambda_j; \lambda_k, \lambda_q, \lambda_p)$ terms (ignore Cartesian indices).

(d) If $|E_1| >> |E_2|$, identify the most dominant terms in part (c).

(e) Write down the nonlinear polarization terms associated with self- and cross phase modulation of each beam (identify $\chi^{(3)}(\lambda_{ij}; \lambda_k, \lambda_q, \lambda_p)$ terms)

(f) Under what condition the simultaneous presence of both beams leads to a nonlinear attenuation (absorption) of both beam? Describe this process, the required energy resonance (use diagrams), and the nature of the complex susceptibility $\chi^{(3)}(\lambda_{ij}; \lambda_k, \lambda_q, \lambda_p)$ (with respect to part e).

(g) Under what condition the simultaneous presence of both beams leads to a nonlinear attenuation (absorption) of one beam (which?) and gain in the other (which?)? Describe this process, the required energy resonance (use diagrams), and the nature of the complex susceptibility $\chi^{(3)}(\lambda_{ij}; \lambda_k, \lambda_q, \lambda_p)$ (with respect to part e and f).
Problem 2. Two-Photon Spectroscopy:

The 1S-2S transition in atomic Hydrogen (E=10.206 eV) is investigated using two-photon spectroscopy with two narrow-band CW laser sources. A pump laser with fixed wavelength $\lambda_1=200\text{nm}$ and a tunable laser ($\lambda=250$-350 nm) are used in a counter propagating arrangement as shown.

(a) Qualitatively plot the transmission of the probe beam as a function of its tunable wavelength $\lambda_2$.

(b) Will the result in (a) be any different if they two beams were co-propagating? (Hint: think Doppler!)
a) \[ \lambda_1 = 1 \text{ nm} \]
\[ \lambda_2 = 0.64 \text{ nm} \]

\[ 2 \omega_1 \Rightarrow \frac{\lambda_1}{2} = 0.5 \text{ nm} \]
\[ 2 \omega_1 = \frac{\lambda_1}{2} = 0.32 \text{ nm} \]

\[ w_3 = w_2 - w_1 \Rightarrow \lambda_3 = (\frac{1}{\lambda_2} - \frac{1}{\lambda_1})^{-1} = \frac{0.6 \times 1}{1 - 0.6} = 1.5 \text{ nm} \]

\[ w_4 = w_2 + w_1 \Rightarrow \lambda_4 = (\frac{1}{\lambda_2} + \frac{1}{\lambda_1})^{-1} = \frac{0.6 \times 1}{1 + 0.6} \]

b) \( x^{(3)} \)

c) \[ w_3 = 2 \omega_1 + w_2 \]
\[ \lambda_3 = (\frac{1}{\lambda_2} - \frac{1}{\lambda_1})^{-1} = \frac{0.5 \times 0.6}{0.5 + 0.6} = 0.273 \text{ nm} \]
\[ w_4 = 2 \omega_2 - w_1 \]
\[ \lambda_4 = (\frac{1}{\lambda_2} + \frac{1}{\lambda_1})^{-1} = \frac{0.2 \times 1}{1 + 0.3} = 0.231 \text{ nm} \]
\[ w_5 = 2 \omega_2 - w_1 \]
\[ \lambda_5 = (\frac{1}{\lambda_2} - \frac{1}{\lambda_1})^{-1} = \frac{0.5 \times 0.6}{0.5 - 0.2} = 3.8 \text{ nm} \]
\[ P(w_3) = 3 \times 5 \times 3 \times (w_3; w_1, w_1, w_1)E_1 E_2 \]
\[ P(w_4) = 3 \times 5 \times 3 \times (w_4; w_2, w_2, w_2)E_1 E_1 \]
\[ P(w_5) = 3 \times 5 \times 3 \times (w_5; w_2, w_2, w_2)E_2 E_2 \]
\[ P(w_6) = 3 \times 5 \times 3 \times (w_6; w_2, w_2, w_2)E_1 E_2 \]

d) \( |E_1| >> |E_2| \)

\[ w_2 = (2 \omega_1 + w_2) \] and \( w_6 = (2 \omega_2 - w_2) \)

one dominant
e) SPM & XPM

\[ P_1(w_1) = 3E_0 \left[ X^{(2)}(w_1, w, w, w_1) |E_1|^2 E_1 + 2 X^{(3)}(w, w_1, w_1, w_2) |E_1|^2 \right] \]

SPM

\[ P_2(w_2) = 3E_0 \left[ X^{(3)}(w_2, w_2, w_2, w_2) |E_2|^2 + 2 X^{(3)}(w_2, w_2, w_1, -w_1) |E_2|^2 \right] \]

XPM.

Similarly:

\[ P_3 = \frac{X^{(3)}(w_3, w_2, w_2, w_2) |E_3|^2}{|E_1|^2} \]

f) Nonlinear absorption in both beams occurs

render nondegenerate "two-photon absorber" circumstances

\[ w_1 + w_2 = w_3 \]

This corresponds to imaginary part of

\[ X^{(3)}(w_1, w_1, w_2, -w_2) \]

\[ X^{(3)}(w_2, w_2, w_1, -w_1) \]
g) **BONUS**

This is a Raman-type process where

\[ W_2 - W_1 = W_{gu} \]

in which \( W_2 \) is absorbed and \( W_1 \) is amplified.

\[ E_{ag} = \frac{1}{2} W_2 - \frac{1}{2} W_1 \]

\[ X^{(3)} I^2 | W_1, W_2, W_2, W_2, W_2, W_1 \rangle \]

\[ \frac{dI_2}{dz} = 0 \]

\[ \Theta \times \text{Im} \{ X^{(3)} \} \]

\[ \frac{dI_1}{dz} = +q \]

Another process that may result in absorption of \( W_2 \), and gain in \( W_1, 2 \) is the 4-photon **parametric process**.

\[ 2w_2 = 2w_2 - w_1 \]

\[ w_1 = 2w_1 - w_2 \]

\[ w_2 (\text{gain}) \]

\[ w_2 (\text{pum}) \]

\[ w_1 (\text{pum}) \]

\[ w_2 (\text{foucault}) \]
Problem 2. Two-Photon Spectroscopy:
(a) Qualitatively, the transmission of the probe should look like this. The x-axis is quantitative.

(b) For counter propagating beams, the Doppler shifts from each photon pair cancel each other:

\[ \nu_1 \left( 1 \pm \frac{v_z}{c} \right) + \nu_2 \left( 1 \mp \frac{v_z}{c} \right) \sim E_{2S-1S} \]

Thus, two-photon spectroscopy can result in measurements of nearly Doppler-free linewidths. For purely Doppler-free measurement, the experiment should involve degenerate two-photon spectroscopy where \( \nu_1 = \nu_2 = E_{2S-1S}/h \) (as shown in the seminal paper by Hansch et al): “Doppler-Free Two-Photon Spectroscopy of Hydrogen 1S–2S”, by T. W. Hänsch, S. A. Lee, R. Wallenstein, and C. Wieman, Phys. Rev. Lett. 34, 307 (1975)

However, in co-propagating arrangement, there is no such cancellation and the resonance condition is:

\[ \nu_1 \left( 1 \pm \frac{v_z}{c} \right) + \nu_2 \left( 1 \pm \frac{v_z}{c} \right) = E_{2S-1S}/h \]

Resulting in a transmission spectrum that is Doppler broadened (qualitatively speaking):