NONLINEAR OPTICS (PHYC/ECE 568)

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Homework #7, Due Monday, Nov. 13

1. Thermal n_2^{eff}

(a) Calculate the nonlocal n_2^{eff} (defined as $<\Delta n>/I_0$) due to laser heating of a liquid characterized by its absorption coefficient α (cm^{-1}), density ρ ($gr./cm^3$), heat capacity C_v(J/K/gr.) and thermo-optic coefficient dn/dT (K^{-1}). The laser intensity is $I(t)=I_0 f(t/t_p)$ where I_0 is the peak intensity and $f(t/t_p)$ denotes the normalized temporal profile of the pulse. Thermal diffusion can be ignored in this problem if we assume that the diffusion time is much longer than t_p while being much shorter than the inter-pulse spacing. The latter requirement is for avoiding heat accumulation from pulse to pulse.

(b) Evaluate n_2^{eff} for liquid CS_2 and a pulsed CO_2 laser (λ=10.6 μm) having a square temporal profile (t_p=100 ns). The CS_2 parameters are $\alpha=0.2\ cm^{-1}$, $\rho C_v=1.3\ J/K/cm^3$, and $dn/dT=-8\times10^{-4}\ K^{-1}$.

(c) If the sound velocity (v_s) in CS_2 is 1.5×10^5 cm/sec., what is the largest laser spot-size (w_0) for which the n_2^{eff} obtained in (b) is valid? What happens as the spot size becomes larger than this value?

2. n_2^{eff} due to photo-generation of charge-carriers in semiconductors:

(a) Calculate the n_2^{eff} due to resonant interband charge-carrier generation in semiconductors. The known parameters for the semiconductor are: the band-gap energy E_g, the electron effective mass m^* (for both conduction and valence bands), the valence-to-conduction band absorption coefficient α, and the carrier recombination time τ. This requires a calculation of the electronic density change ΔN (in both bands) due to linear absorption followed by the calculation of the resultant index change Δn from both bands using a harmonic classical electron oscillator (CEO) model. In this simple approach, the electrons in the valence band are considered bound with a resonant frequency $\omega_0=\omega_g=E_g/\hbar$, while the conduction electrons are considered free ($\omega_0=0$). Ignore damping in the CEO models. The governing equation for ΔN is:

$$\frac{d\Delta N}{dt} = \frac{\alpha I(t)}{\hbar\omega} - \frac{\Delta N}{\tau}$$

where $\hbar\omega$ is the incident photon energy and $I(t)=I_0 f(t/t_p)$ is the instantaneous laser intensity. Consider two extreme cases of $t_p>>\tau$ and $t_p<<\tau$. (You may assume a rectangular pulse).

(b) Evaluate n_2^{eff} (cm/W) and the effective $\chi^{(3)}$ (esu or m^2/V^2) for GaAs with E_g=1.4 eV, $m^*=0.1\ m_0$, $\alpha=100\ cm^{-1}$, τ=1 ns. The laser wavelength is λ=900 nm and t_p=10 ps.

(c) Do you expect an (nonlinear) absorptive component $\Delta\alpha$ associated with the above index change Δn? Explain (briefly).

Problem 3. Consider an *isotropic* nonlinear material (e.g. silica glass). We are concerned here with the self-phase modulation case where $\chi^{(3)}_{ijkl}(\omega;\omega,\omega,-\omega)$ is involved.

(a) Show that $\chi^{(3)}_{xxxx}=\chi^{(3)}_{xxyy}+\chi^{(3)}_{xyyx}+\chi^{(3)}_{xyxy}$

(b) Let the incident field be represented by $\vec{E}=E_0 e^{-i\omega t}(\hat{x}+e^{i\phi}\hat{y})/\sqrt{2}$ where ϕ is the phase difference between *x* and *y* components. Note that $\phi=m\pi$ and $\phi=(m+1/2)\pi$ describe the linear and circular polarization receptively. Let us now define an effective nonlinear susceptibility through:

$$\vec{P}^{(3)}(\omega)=3\varepsilon_0\chi^{(3)}_{eff}\,|E_0|^2\,\vec{E}$$

(i) Find $\chi^{(3)}_{eff}$ as a function of the $\chi^{(3)}$ tensor elements and the phase angle ϕ. Note, this corresponds to conditions where P is in phase as well as along E. Can we define such a χ_{eff} for all values of ϕ (i.e. elliptical polarization)? *Read Sec. 4.2 (Boyd, 3rd. ed.). You may start with Eq. 4.2.7*

(ii) Find the circular/linear dichroism defined as $\eta=\chi^{(3)}_{eff}(\phi=\pi/2)/\chi^{(3)}_{eff}(\phi=0)$

NLO HW # ~~7~~ Solution

problem ① - ⓐ

$$n_2^{eff} = \frac{\langle \Delta n \rangle}{I_0} = \left\langle \frac{\partial n}{\partial T} \cdot \Delta T \right\rangle \Big/ I_0 = \frac{\partial n}{\partial T} \cdot \frac{\langle \Delta T \rangle}{I_0} .$$

$$\frac{dT}{dt} = \frac{1}{\rho C_v} \alpha I(t) \rightarrow \Delta T(t) = \frac{\alpha}{\rho C_v} \int_{-\infty}^{+\infty} I(t')\, dt'$$

$$= \frac{\alpha I_0}{\rho C_v} \int_{-\infty}^{+\infty} f\left(\frac{t'}{t_p}\right) dt'$$

- for a square pulse: $\Delta T(t) = \dfrac{\alpha I_0 t}{\rho C_v}$

$$\Rightarrow \langle \Delta T \rangle = \frac{\alpha I_0}{\rho C_v} \frac{t_p}{2}$$

* In General,

- for an arbitrary pulse shape $f(t)$ (i.e. $I(t) = I_0 f(t)$)

the averaging is performed as

$$\langle \Delta T \rangle = \frac{\int \Delta T(t) f(t)\, dt}{\int f(t)\, dt} = \frac{\alpha I_0}{\rho C_v} \cdot \frac{\int_{-\infty}^{+\infty} f(t)\, dt \int_{-\infty}^{+\infty} f(t')\, dt'}{\int_{-\infty}^{+\infty} f(t)\, dt}$$

Since, $t_p = \int_{-\infty}^{+\infty} f(t)\, dt$ → One can show that independent of the slope of $f(t)$,

$$\rightarrow \frac{\int_{-\infty}^{+\infty} f(t)\, dt \int_{-\infty}^{+\infty} f(t')\, dt'}{\int_{-\infty}^{+\infty} f(t)\, dt} = \frac{t_p}{2}$$

$$\Rightarrow n_2^{eff} = \frac{\partial n}{\partial T} \cdot \frac{\alpha I_0 t_p}{2 \rho C_v} \times \frac{1}{I_0} \Rightarrow n_2^{eff} = \frac{\alpha t_p}{2 \rho C_v} \cdot \frac{\partial n}{\partial T}$$

problem ①-ⓑ

$$n_2^{eff} = \frac{0.2 \times 100 \times 10^{-9}}{1.3 \times 2} \times -8 \times 10^{-4} \simeq -6 \times 10^{-12} \ cm^2/W$$

problem ①_ⓒ

- the rise ~~time~~ of the nonfocal thermal lens is the accoustic transit time across the beam:

$$\tau_{ac} \simeq \frac{w_0}{v_s}$$

→ For n_2^{eff} derived in part "a" to be valid,

$t_p > \tau_{ac} \Rightarrow w_0 < v_s t_p = 1.5 \times 10^{-2}$ cm $= 150\ \mu m$

for $w_0 > 150\ \mu m$ → n_2^{eff} reduces.

problem (2) - (a)

$$\rightarrow \quad n \simeq 1 + \frac{2\pi(e^2/m^*)N_b}{\omega_g^2 - \omega^2} + \frac{2\pi(e^2/m)N_f}{-\omega^2}$$

- Initially, all the electrons are bound. (in the valence band) and N_f (density of electron in Conduction band) = 0

- after excitation of ΔN, n can be obtained by replacing $N_b \rightarrow N_b - \Delta N$ and $N_f \rightarrow \Delta N$!

Thus,

$$\Delta n = \frac{2\pi e^2 \Delta N}{m^*}\left(\frac{-1}{\omega_g^2 - \omega^2} + \frac{+1}{-\omega^2}\right) = \frac{-2\pi e^2 \Delta N}{m^* \omega^2} \cdot \frac{\omega_g^2}{\omega_g^2 - \omega^2}$$

Now, we evaluate ΔN.

$$\frac{d(\Delta N)}{dt} = \frac{\alpha I(t)}{\hbar\omega} - \frac{\Delta N}{\tau} \implies \text{let } \Delta N = \Delta N' e^{-t/\tau}$$

$$\implies \frac{d\Delta N'}{dt} = \frac{\alpha I(t) e^{t/\tau}}{\hbar\omega} \implies \boxed{\Delta N(t) = \frac{\alpha I_0}{\hbar\omega}\int_{-\infty}^{t} f\left(\frac{t'}{t_p}\right) e^{\frac{t'-t}{\tau}} dt'}$$

* Case 1: $t_p \gg \tau$

then letting $t > \tau$, $\quad \displaystyle\int_{-\infty}^{t} f\left(\frac{t'}{t_p}\right) e^{\frac{t'-t}{\tau}} dt' \simeq \tau$

$$\rightarrow \quad \Delta N(t) = \frac{\alpha I_0 \tau}{\hbar\omega}$$

$$\rightarrow \quad \langle \Delta N \rangle = \frac{\alpha I_0 \tau}{\hbar\omega}$$

problem ② - ⓐ

* Case 2 : $t_p \ll \tau$

$$-\int_{-\infty}^{t} f\left(\frac{t'}{t_p}\right) e^{\frac{t'-t}{\tau}}\, dt' \simeq \int_{-\infty}^{\tau} f\left(\frac{t'}{t_p}\right) dt'.$$

- similar to problem ①,

$$\langle \Delta N \rangle = \frac{\alpha I_0 t_p}{2\hbar\omega}.$$

Thus,

$$n_2^{eff} = \frac{\langle \Delta n \rangle}{I_0} = \frac{K \langle \Delta N \rangle}{I_0}$$

Case ① $\rightarrow$ $$n_2^{eff} = -\frac{2\pi e^2}{m^*} \cdot \frac{\alpha \tau}{\hbar\omega} \cdot \frac{1}{\omega^2} \left(\frac{\omega_g^2}{\omega_g^2 - \omega^2} \right)$$

Case ② $\rightarrow$ $$n_2^{eff} = -\frac{2\pi e^2}{m^*} \frac{\alpha t_p}{2\hbar\omega} \cdot \frac{1}{\omega^2} \left(\frac{\omega_g^2}{\omega_g^2 - \omega^2} \right)$$

problem ② - ⓑ

$\rightarrow t_p (= 10\ ps) \ll \tau (= 1\ ns)$ $\Rightarrow$ Case ② applies!

$\rightarrow$ Note :

$$n_2^{eff}(mks) = -\frac{e^2 \alpha t_p}{4 m^* \hbar\omega\, \epsilon_0} \cdot \frac{1}{\omega^2} \cdot \left(\frac{\omega_g^2}{\omega_g^2 - \omega^2} \right)$$

problem (2)-(c)

→ Obviously, due to kramers-kroning relations, (principle of - Casuality), any changing in index Δn is associated (accompanied) by a change of absorption (at resonance).

- For the above problem, this change of absorption is due by absorption

In a two level system,

$$\alpha = (N_1 - N_2)\sigma \qquad \begin{cases} N_1 \to N_0 - \Delta N \\ N_2 \to \Delta N \end{cases}$$

$$\to \Delta\alpha = -2.\Delta N.\sigma = -2\,\frac{\alpha t_p \sigma}{2\hbar\omega}.I$$

$$\Rightarrow \Delta\alpha = -kI$$

problem (3)

$$P_i = 3\epsilon_0 \sum \chi^{(3)}_{ijkl} E_j E_k E_l^* .$$

So,

$$P_x = 3\epsilon_0 \left\{ \chi_{xxxx} E_x E_x E_x^* + (\chi_{xxyy} + \chi_{xyxy}) E_x E_y E_y^* + \chi_{xyyx} E_y E_y E_x^* \right\}$$

$$P_y = 3\epsilon_0 \left\{ \chi_{xxxx} E_y E_y E_y^* + (\chi_{xxyy} + \chi_{xyxy}) E_y E_x E_x^* + \chi_{xyyx} E_x E_x E_y^* \right\}$$

↳ when the symmetry property, that $\begin{cases} \chi_{xxxx} = \chi_{yyyy} \\ \chi_{xxyy} = \chi_{yyxx} \end{cases}$, etc. has been used.

(a) → Consider two cases: (1) → $\vec{E} = E_x$ only $(= E_0 e^{-i\omega t})$

(2) → $\vec{E} = \frac{E_0}{\sqrt{2}} (\hat{x} + \hat{y}) e^{-i\omega t}$.

- Case (2) is obtained by rotating the isotropic material by 45° !

- Case (1) : $\vec{P} = P_x = 3\epsilon_0 \chi_{xxxx} E_0 |E_0|^2 e^{-i\omega t}$.

- Case (2) : $\vec{P} = P_x \hat{x} + P_y \hat{y} = 3\epsilon_0 (\chi_{xxxx} + \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx})$

$$\times \frac{1}{2\sqrt{2}} |E_0| E_0 e^{-i\omega t} \frac{(\hat{x} + \hat{y})}{\sqrt{2}}$$

problem ③

- for isotropic materials, $\vec{P}$ should be the same, regardless of the axes orientations; thus,

$$\frac{\chi_{xxxx} + \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}}{2} = \chi_{xxxx}$$

or $\boxed{\chi_{xxxx} = \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}}$

problem ③ - ⓑ

$\rightarrow \vec{E} = \frac{E_0}{\sqrt{2}} (\hat{x} + \hat{y} e^{i\varphi}) e^{-i\omega t}$

- substitute into P_x & P_y in part (a):

$$\begin{cases} P_x = 3\epsilon_0 \left\{ \left(\chi_{xxxx} - \frac{\chi_{xyyx}}{2} \right) + \chi_{xyyx} \frac{e^{2i\varphi}}{2} \right\} |E_0|^2 \frac{E_0}{\sqrt{2}} e^{-i\omega t} \\ P_y = 3\epsilon_0 \left\{ \left(\chi_{xxxx} - \frac{\chi_{xyyx}}{2} \right) + \chi_{xyyx} \frac{e^{-2i\varphi}}{2} \right\} |E_0|^2 \frac{E_0}{\sqrt{2}} e^{i\varphi} e^{-i\omega t}. \end{cases}$$

$\rightarrow$ Note,

$\vec{P} = P_x \hat{x} + P_y \hat{y}$ $\rightarrow$ will be along $\vec{E}$ only if:

$$e^{2i\varphi} = e^{-2i\varphi}$$

- This corresponds to:

(i) Linear Polarization $\rightarrow$ $\varphi = m\pi$ $\Rightarrow$ $e^{2i\varphi} = e^{-2i\varphi} = 1$

(ii) Circular Polarization $\rightarrow$ $\varphi = (2m+1)\frac{\pi}{2}$ $\Rightarrow$ $e^{2i\varphi} = e^{-2i\varphi} = -1$

- Thus,

$$\chi^{(3)}_{eff} = \begin{cases} \chi_{xxxx} & ; \text{ for LP} \\ (\chi_{xxxx} - \chi_{xyyx}) & ; \text{ for CP} \end{cases}$$

- Circular Dichroism:

$$\eta = \frac{\chi^{(3)}_{eff}\ (\text{Circular})}{\chi^{(3)}_{eff}\ (\text{linear})} = 1 - \frac{\chi_{xyyx}}{\chi_{xxxx}}$$

For Kleinman System: $\eta = \frac{2}{3}$.