Exercise 1:

Second-order:

Boyd 1.1.2.

$$\begin{matrix} \omega_1 \rightarrow \\ \omega_2 \rightarrow \end{matrix} \boxed{\chi^{(2)}} \rightarrow \left.\begin{matrix} 2\omega_1 \\ 2\omega_2 \\ \omega_1+\omega_2 \\ \omega_2-\omega_1 \,/\, \omega_1-\omega_2 \end{matrix}\right\} \text{possible frequencies.}$$

→ See next page for work out.

All of these processes can be used in the generation of light but also to study materials.

SHG: Surface spectroscopy
Surface imaging or bulk imaging
SH scattering → particle interfaces
nanoscale structure in bulk liquids.
Mostly probe of electronic structure; resonant or non-resonant

SFG: Often used as surface vibrational spectroscopy to measure a vibrational spectra of interfaces - planar or particle interfaces. Another SFG process is the linear electro optic effect.

SHG / SFG are useful because of their surface specificity. This property follows from the spatial symmetry properties of the material under study.

Third-order processes:

There are many third-order processes; some are used to generate new frequencies, others as switches or to manipulate light, to probe matter, or as a distortion to experiments.

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c.$$

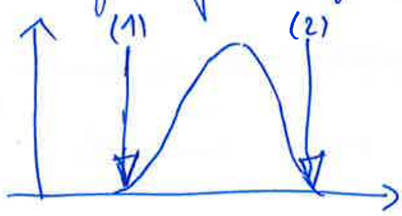
$$p^{(2)} = \epsilon_0 \chi^{(2)} E(t)^2$$

$$p^{(2)} = \varepsilon_0 \chi^{(2)} \cdot \Big[\underbrace{E_1^2 e^{-2i\omega_1 t}}_{\text{SHG}} + \underbrace{E_2^2 e^{-2i\omega_2 t}}_{\text{SHG}} + \underbrace{2E_1E_2 e^{-i(\omega_1+\omega_2)t}}_{\text{SFG}} + \underbrace{2E_1E_2^* e^{-i(\omega_1-\omega_2)t}}_{\text{DFG}} + c.c. \Big] +$$

$$+ 2\varepsilon_0 \chi^{(2)} \left[E_1E_1^* + E_2E_2^* \right]$$

OR (optical rectification)

= DFG of the pulse with itself $(2)-(1)$ ⇒ get very low frequencies (THz)



Exercise 1, continued:

Examples:

- Third harmonic generation
- Intensity dependent refractive index
- Kerr effect
- Electric field induced second harmonic generation.

To Exercise 1

Example:

FH signal ~~400~~ 190 – ~~400~~ 300

4H signal : 310 – 540

signal

1086 ···> 513 ---> 614 – 1080
980 -- 2700 → idler

(980 + 1080 = 518)
(614 + 2700 = 514)

signal – idler
4500 – 20000 nm
(KTA ; gase)

2H idler : 490 – 630

4H idler 280 – 380

White light

WL
+
513

BBO ?

Exercise 2

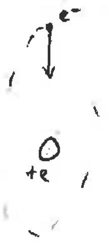
2a.

Attraction of nucleus with electron ((Coulombic): centripetal (F_p)

Reaction force due to rotating motion in fixed reference frame: centrifugal (F_f)

b $m_e \cong 10^{-31}$ kg $m_{H^+} \cong 10^{-27}$ kg . the electron moves, the nucleus remains fixed

c.



$\sin\theta = \dfrac{l}{R}$

$F_e = \dfrac{-e^2}{4\pi\varepsilon_0 \varepsilon R^2}$

$F_{ind} = -e \cdot E = \dfrac{-e^2 \cdot \sin\theta}{4\pi\varepsilon_0\varepsilon R^2} = \dfrac{-e^2 \cdot l}{4\pi\varepsilon_0\varepsilon R^3} \Rightarrow \dfrac{e \cdot P_{ind}}{4\pi\varepsilon_0\varepsilon R^3} = eE$

$P_{ind} = -e \cdot l$

$= \bar{A} K_0 E$

$\uparrow$

$e: -;\ E: -$

or

$P_{ind} = \underbrace{4\pi\varepsilon_0\varepsilon R^3}_{\alpha_0} \cdot E$

d. We have $P^{(1)} = N \cdot \langle p^{(1)} \rangle$ orientational avg. for a gas this is simply $N \cdot p^{(1)}$

$P^{(1)} = \varepsilon_0 \chi^{(1)} E = N\, p^{(1)} = N \cdot 4\pi\varepsilon_0\varepsilon R^3 \cdot E$

$\Rightarrow \chi^{(1)} = 4\pi\varepsilon R^3 \cdot N$ and $\varepsilon = 1 + \chi^{(1)}$

$\qquad m^3 \quad \dfrac{\text{Molecules}}{m^3}$

the gas

$R^3 = (5.29 \cdot 10^{-11})^3 = 1.48 \cdot 10^{-31}$ m3

$\chi^{(1)} = 0.04$

$\varepsilon = 1.04$

$d = 70.85 \ \dfrac{kg}{m^3}$

$M = 2$ g/mol $= 2 \cdot 10^{-3}$ kg/mol

$N = 70.85 \cdot 500 \cdot 6.02 \cdot 10^{23} = 2.13 \cdot 10^{28} \ \dfrac{\text{molec.}}{m^3}$

$\dfrac{kg}{m^3} \cdot \dfrac{mol}{kg} \cdot \dfrac{molec}{mol}$

$p = e \cdot l$ dipole moment $[C \cdot m]$

$\;\;= \alpha E$

$P = N \underbrace{\alpha E}_{p}$ polarizability (macroscopic) (density of dipoles) $\left[\frac{C \cdot m}{m^3}\right]$

Exercise 3

3a. $E = \frac{e}{4\pi\varepsilon_0 \varepsilon R^2}$ $\rightarrow$ $E = 5 \cdot 10^{11} \frac{V}{m}$

$a_0 = 5.29 \cdot 10^{-11}$ m

b. $I = 2n\varepsilon_0 c |E|^2 = 1.3 \cdot 10^{17}$ W/cm²

c. $P_{peak} = \frac{P_{avg}}{f \cdot \tau}$ (rep rate ; pulse dur) $\frac{P_{avg}}{f}$ = pulse energy

$I_{peak} = \frac{P_{peak}}{A} = \frac{P_{avg}}{f \cdot \tau \cdot A}$

$\uparrow$ area

$0.013 \cdot f = 1$

$f = 77$ Hz ; so not with 1 kHz - 1 MHz

(due to the small focus)

Note that with this type of ~~intensity~~ pulse energy (13 mJ) there is typically already white light generation with this narrow focus.