Lecture 19

Third harmonic generation. Nonlinear absorption.

Parametric processes due to 4-wave mixing.

Third harmonic generation

Third harmonic generation (THG)

Fourier component

Assume that the input field has only one frequency component: ω $\qquad E(t) = \frac{1}{2}E_1 e^{i\omega t} + c.c.$

Also assume a scalar version of the 3-wave interaction : all fields are along one axis (e.g. x-axis)

We will now look for the nonlinear polarization component at angular frequency $\pm 3\omega$ in the form

$$P^{(3)}(t) = \frac{1}{2}(P(3\omega)e^{i3\omega t} + c.c.)$$

NL polarization:

$$P_i^{(3)} = \epsilon_0 \sum_{j,k,l} \chi_{ijkl} E_j E_k E_l$$

→ scalar version $\quad P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$

$\chi^{(3)} = \chi^{(3)}_{xxxx}$

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} (\tfrac{1}{2} E_1 e^{i\omega t} + c.c.)^3 = = \frac{1}{8}\epsilon_0 \chi^{(3)} (E_1^3 e^{3i\omega t} + 3(E_1 E_1^*) E_1 e^{i\omega t} + c.c.) =$$

pick only components with $\pm 3\omega$

$$= \frac{1}{4}\epsilon_0 \chi^{(3)} (\frac{1}{2} E_1^3 + c.c)$$

$$\rightarrow \qquad P(3\omega) = \frac{1}{4}\epsilon_0 \chi^{(3)} E_1^3 \qquad (19.1)$$

Third harmonic generation (THG)

Now use Slowly Varying Envelope Approximation (SVEA) from Lecture 2 to calculate the THG output :

$$\frac{\partial E(z)}{\partial z} = -\frac{i\omega c}{2n}\mu_0 P \qquad (2.3)$$

perturbation polarization

$$P(3\omega) = \frac{1}{4}\epsilon_0 \chi^{(3)} E_1^3(\omega)$$

In the slowly varying envelope (SVEA) approximation

$$\frac{\partial E(3\omega)}{\partial z} = -\frac{i\omega}{2nc\epsilon_0} P_{NL}$$

$$E(3\omega) = E_3(z)$$

phase mismatch

$$\Delta k = k_3 - 3k_1$$

In the low conversion limit:
$E_3 \ll E_1,\ E_1 = const$

$$\frac{\partial E_3(z)}{\partial z} = -\frac{i\omega}{8nc}\chi^{(3)} E_1^3(\omega) e^{-i\Delta k z}$$

Third harmonic generation (THG)

Easy to integrate if Δk=0:

$$E_3(3\omega) = -i\frac{\omega}{8nc}\chi^{(3)}E_1^3\,L$$

TH field grows linearly with length

$$|E_3(3\omega)|^2 = (\frac{\omega}{8nc}\chi^{(3)})^2|E_1|^6L^2$$

Now recall that the intensity $I = \frac{1}{2}c\epsilon_0 n|E|^2$ → $|E|^2 = 2I/c\epsilon_0 n$

$$I_{3\omega} = \left(\frac{\omega}{8nc}\chi^{(3)}\right)^2(\frac{2}{c\epsilon_0 n})^2 {I_\omega}^3L^2 = (\frac{\omega}{4\epsilon_0 n^2c^2}\chi^{(3)})^2{I_\omega}^3L^2 \quad (19.2)$$

Power $P = I \times A_{eff}$ → $I = \dfrac{P}{A_{eff}}$

$$\frac{P_{3\omega}}{A_{eff}} = (\frac{\omega}{4\epsilon_0 n^2c^2}\chi^{(3)})^2(\frac{P_\omega}{A_{eff}})^3L^2$$

Power

$$\boxed{P_{3\omega} = (\frac{\omega}{4\epsilon_0 n^2c^2}\chi^{(3)})^2\frac{P_\omega^3L^2}{{A_{eff}}^2}} \quad (19.3)$$

– scales as as ${P_\omega}^3$ and grows quadratically with length

Third harmonic generation (THG)

Refractive indices, phase-matching directions and third order nonlinear coefficients of rutile TiO_2 from third harmonic generation

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Abstract: Experiments of third harmonic generation in rutile TiO_2 allowed us to determine the phase-matching angles and the refractive indices of the crystal up to 4500 nm. We also showed that χ_{16} and χ_{18} coefficients of the third order electric susceptibility tensor exhibit opposite signs, and that $|\chi_{18}(616.7\text{nm})| = 9.7 \times 10^{-20}$ m^2V^{-2}.

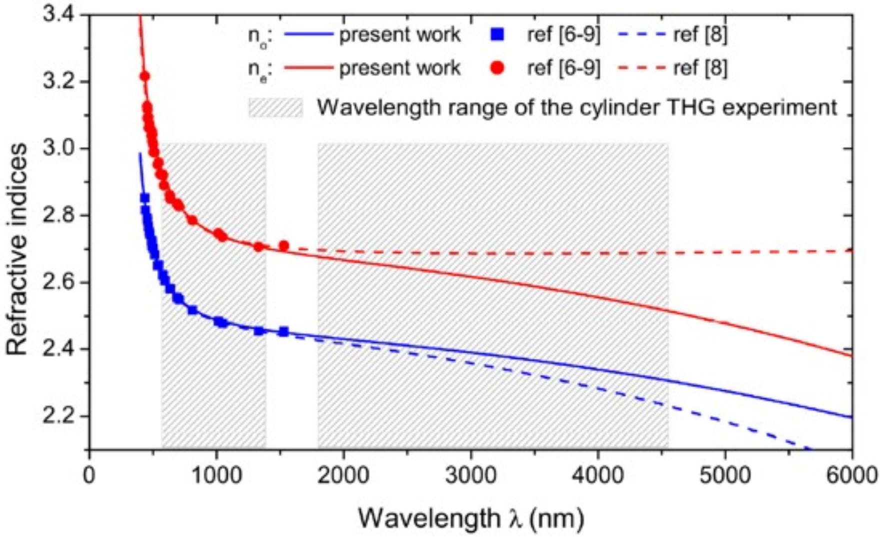

Fig. 3. Ordinary n_o and extraordinary n_e principal refractive indices of TiO_2 as a function of wavelength from present work in solid lines, and from [6–9] in symbols (experiments) and dashed lines (calculations).

$$\omega^{(o)} + \omega^{(e)} + \omega^{(e)} \rightarrow 3\omega^{(o)}$$

phase matching

Third harmonic generation (THG)

For $\Delta k \neq 0$

and low conversion limit:
$E_3 \ll E_1, \quad E_1 \approx const$

$$\frac{\partial E_3(z)}{\partial z} = -\frac{i\omega}{8nc}\chi^{(3)}E_1^3(\omega)e^{-i\Delta kz}$$

$$E_3(3\omega) = -\frac{i\omega}{8nc}\chi^{(3)}E_1^3(\omega)\underbrace{\int_0^L e^{-i\Delta kz}dz} = -\frac{\omega}{4nc\Delta k}\chi^{(3)}E_1^3(\omega)$$

$$-\frac{i}{\Delta k}(1 - e^{-i\Delta kL}) = -\frac{2i}{\Delta k}$$ (at L=L_c=$\pi/\Delta k$)

$$|E_3|^2 = (\frac{\omega}{4nc\Delta k}\chi^{(3)})^2|E_1|^6$$ field intensity

Now recall that the intensity $I = \frac{1}{2}c\epsilon_0 n|E|^2$ → $|E|^2 = 2I/c\epsilon_0 n$

Third harmonic generation (THG)

for intensities:
$$2I_{3\omega}/c\epsilon_0 n = (\frac{\omega}{4nc\Delta k}\chi^{(3)})^2(2I_\omega/c\epsilon_0 n)^3$$

$$I_{3\omega} = (\frac{\omega}{2\epsilon_0 n^2 c^2 \Delta k}\chi^{(3)})^2 {I_\omega}^3 \qquad (19.4)$$

Power $P = I \times A_{eff}$ → $I = \frac{P}{A_{eff}}$

$$\frac{P_{3\omega}}{A_{eff}} = (\frac{\omega}{2\epsilon_0 n^2 c^2 \Delta k}\chi^{(3)})^2(\frac{P_\omega}{A_{eff}})^3$$

$$P_{3\omega} = (\frac{\omega}{2\epsilon_0 n^2 c^2 \Delta k}\chi^{(3)})^2 \frac{P_\omega^3}{{A_{eff}}^2} \qquad (19.5)$$

– grows as ${P_\omega}^3$ over one coherence length

Third harmonic generation in a fiber

Now let us take a concrete example: THG in a fiber 1.56 μm -> 0.52 μm (green)

Fused silica: $\chi^{(3)} = 2.5\times10^{-22}\, m^2/V^2$

$$\Delta k = 2\pi\left(\frac{n3}{\lambda 3} - 3\,\frac{n1}{\lambda 1}\right) = 2\pi\left(\frac{1.4613}{0.52} - 3\,\frac{1.4439}{1.56}\right) = 0.21\ \mu m^{-1} = 2.1e5\ m^{-1}$$

(coh. length $\frac{\pi}{\Delta k}$ ~ 15 μm)

Assume a fused silica fiber with A_{eff}=(10μm)^2 → 10^{-10} m^2

Assume 1-W CW pump laser at frequency ω (1.56 μm)

$$P_{3\omega} = \left(\frac{\omega}{2\epsilon_0 n^2 c^2 \Delta k}\chi^{(3)}\right)^2 \frac{P_\omega^3}{{A_{eff}}^2} =$$

=(1.2e15*2.5e-22 /2 /8.85e-12/ 1.46^2 /3e8^2/2.1e5)^2 *1/ 1e-10^2=

= 1.7 x10^{-17} W → 45 photons per second

is it practical ?

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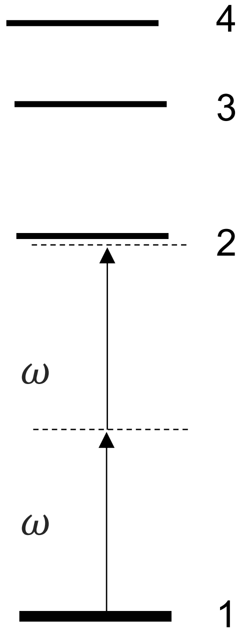
* green photon at 0.52 μm is: $\hbar\omega$ = 3.8 x10^{-19} J

Two-photon absorption

Nonlinear susceptibility $\chi^{(3)}$, quantum mechanical model

see e.g. Boyd, Stegeman

$\chi^{(3)}$ - is the sum of many different terms, but let us take the term that is **close to resonance 1-3**:



$$\chi^{(3)} \approx \frac{N}{\epsilon_0 \hbar^3} \frac{\mu_{14}\mu_{12}\mu_{23}\mu_{34}}{(\omega_{31}-2\omega)(\omega_{21}-2\omega)(\omega_{21}-\omega)}$$

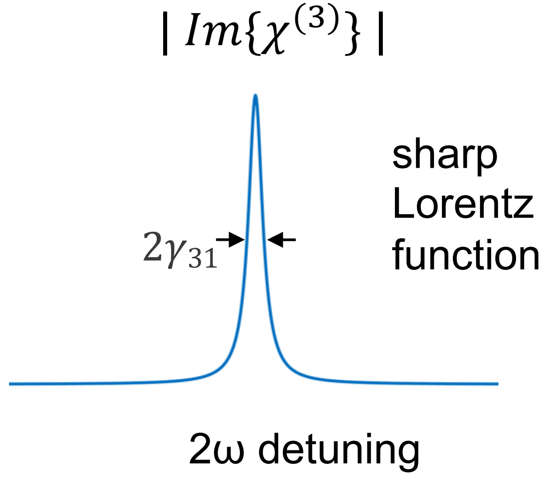
$$\rightarrow \quad \chi^{(3)} \approx \frac{N}{\epsilon_0 \hbar^3} \frac{\mu_{14}\mu_{12}\mu_{23}\mu_{34}}{(\omega_{31}-2\omega)(\omega_{21}-2\omega+i\gamma_{21})(\omega_{21}-\omega)} = -i \frac{N}{\epsilon_0 \hbar^3} \frac{1}{\gamma_{21}} \frac{\mu_{14}\mu_{12}\mu_{23}\mu_{34}}{(\omega_{31}-2\omega)(\omega_{21}-\omega)}$$

added damping term

At 2-photon resonance 1-3

$$Im\{\chi^{(3)}\} = -\frac{N}{\epsilon_0 \hbar^3} \frac{1}{\gamma_{21}} \frac{\mu_{14}\mu_{12}\mu_{23}\mu_{34}}{(\omega_{31}-2\omega)(\omega_{21}-\omega)} \qquad (19.6)$$

$$Re\{\chi^{(3)}\} = 0$$



2-photon transitions can take place where 1-photon transitions may be forbidden

Two-photon absorption

Recall slowly varying envelope approximation (SVEA) equation

$$\frac{\partial E(z)}{\partial z} = -\frac{i\omega c}{2n}\mu_0 P = -\frac{i\omega}{2\varepsilon_0 cn} P \qquad (2.3) \qquad \text{(from L2)}$$

Recall NL polarization for the $\omega = \omega + \omega - \omega$ process

$$P(\omega) = \frac{3}{4}\epsilon_0 \chi^{(3)} |E_1(\omega)|^2 E_1(\omega) \qquad (17.3) \qquad \text{(from L17)}$$

Hence we have at 1-2 2-photon resonance:

$$\frac{\partial E}{\partial z} = -\frac{i\omega}{2\varepsilon_0 cn} P = -\frac{i\omega}{2\varepsilon_0 cn}\frac{3}{4}\epsilon_0 \chi^{(3)} |E|^2 E$$

$$= -\frac{i3\omega}{8nc}\{-i|Im(\chi^{(3)}|\}|E|^2 E = -\frac{3\omega}{8nc}|Im(\chi^{(3)}||E|^2 E$$

$$= -\frac{3\omega}{8nc}\frac{2I}{c\epsilon_0 n}|Im(\chi^{(3)}|E = -\frac{3\omega}{4\epsilon_0 n^2 c^2} I|Im(\chi^{(3)}|E$$

$$I = \frac{1}{2} c\epsilon_0 n|E|^2$$

$$\frac{\partial E}{\partial z} = -\alpha_2^E IE \qquad \alpha_2^E = \frac{3\omega}{4\epsilon_0 n^2 c^2}|Im(\chi^{(3)}|$$

$$\frac{\partial I}{\partial z} = -\alpha_2 I^2 \qquad \alpha_2 = \frac{3\omega}{2\epsilon_0 n^2 c^2}|Im(\chi^{(3)}| \qquad (19.7)$$

Two-photon absorption (another way to derive)

How does the power change if $Im\{\chi^{(3)}\} \neq 0$?

Power ~ $|E(\omega)|^2 = E(\omega)E^*(\omega)$

$$\frac{d}{dz}|E(\omega)|^2 = \frac{d}{dz}E(\omega)E^*(\omega) = E^*\frac{dE}{dz} + E\frac{dE^*}{dz} = E^*\left(-i\frac{3\omega}{8nc}(-i\,|Im\{\chi^{(3)}\}|)\,|E|^2E\right) + E\left(-i\frac{3\omega}{8nc}(-i\,|Im\{\chi^{(3)}\}|)|E|^2E^*\right) =$$

$Im\{\chi^{(3)}\} = -|Im\{\chi^{(3)}\}|$

$$= -\frac{3\omega}{4nc}|E|^4\,|Im\{\chi^{(3)}\}|$$

Recall that the intensity $I = \frac{1}{2}c\epsilon_0 n|E|^2$ → $|E|^2 = \frac{2I}{c\epsilon_0 n}$

$$\frac{2}{c\epsilon_0 n}\frac{dI}{dz} = -\frac{3\omega}{4nc}\left(\frac{2I}{c\epsilon_0 n}\right)^2|Im\{\chi^{(3)}\}|$$

$$\boxed{\frac{dI}{dz} = -\frac{3\omega}{2n^2\epsilon_0 c^2}\,|Im\{\chi^{(3)}\}|\,I^2 = -\alpha_2 I^2}$$

power is absorbed if $\chi^{(3)}$ has imaginary portion

Two-photon absorption (2PA) coefficint

$$\boxed{\alpha_2 = \frac{3\omega}{2\epsilon_0 n^2 c^2}\,|Im\{\chi^{(3)}\}|} \quad (19.8)$$

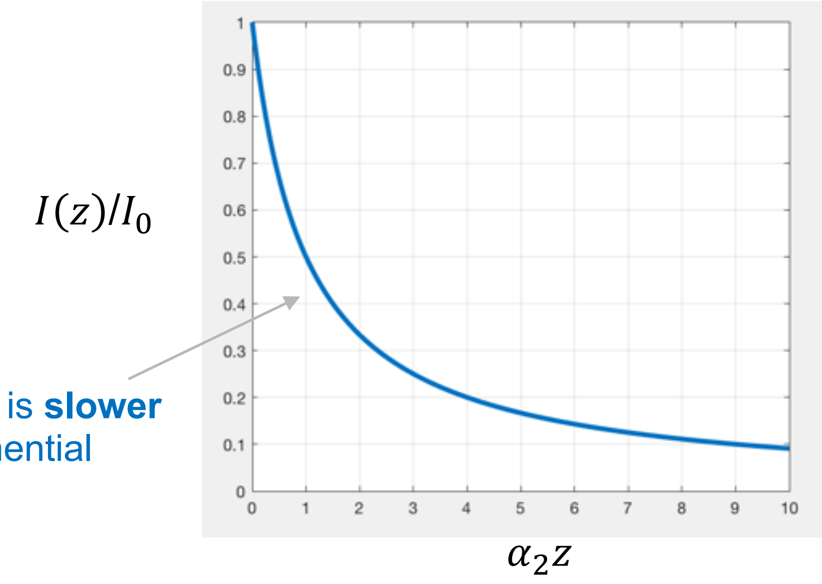
same as (19.7) !

2PA units: cm/W or m/W

Two-photon absorption

It is easy to integrate $\frac{dI}{dz} = -\alpha_2 I^2$

to get : $$I(z) = \frac{I_0}{1+I_0\alpha_2 z} \qquad (19.9)$$

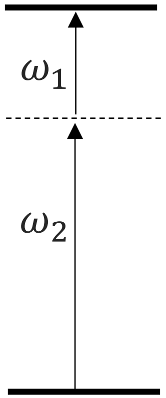


the decay rate is **slower** than exponential

Non-degenerate 2PA

$$\frac{dI_1}{dz} \sim -\alpha_2^{non-degen.} I_1\, I_2$$

$$\frac{dI_2}{dz} = \frac{\omega_2}{\omega_1}\frac{dI_1}{dz}$$



$\omega_1 = \omega_1 + \omega_2 - \omega_2$

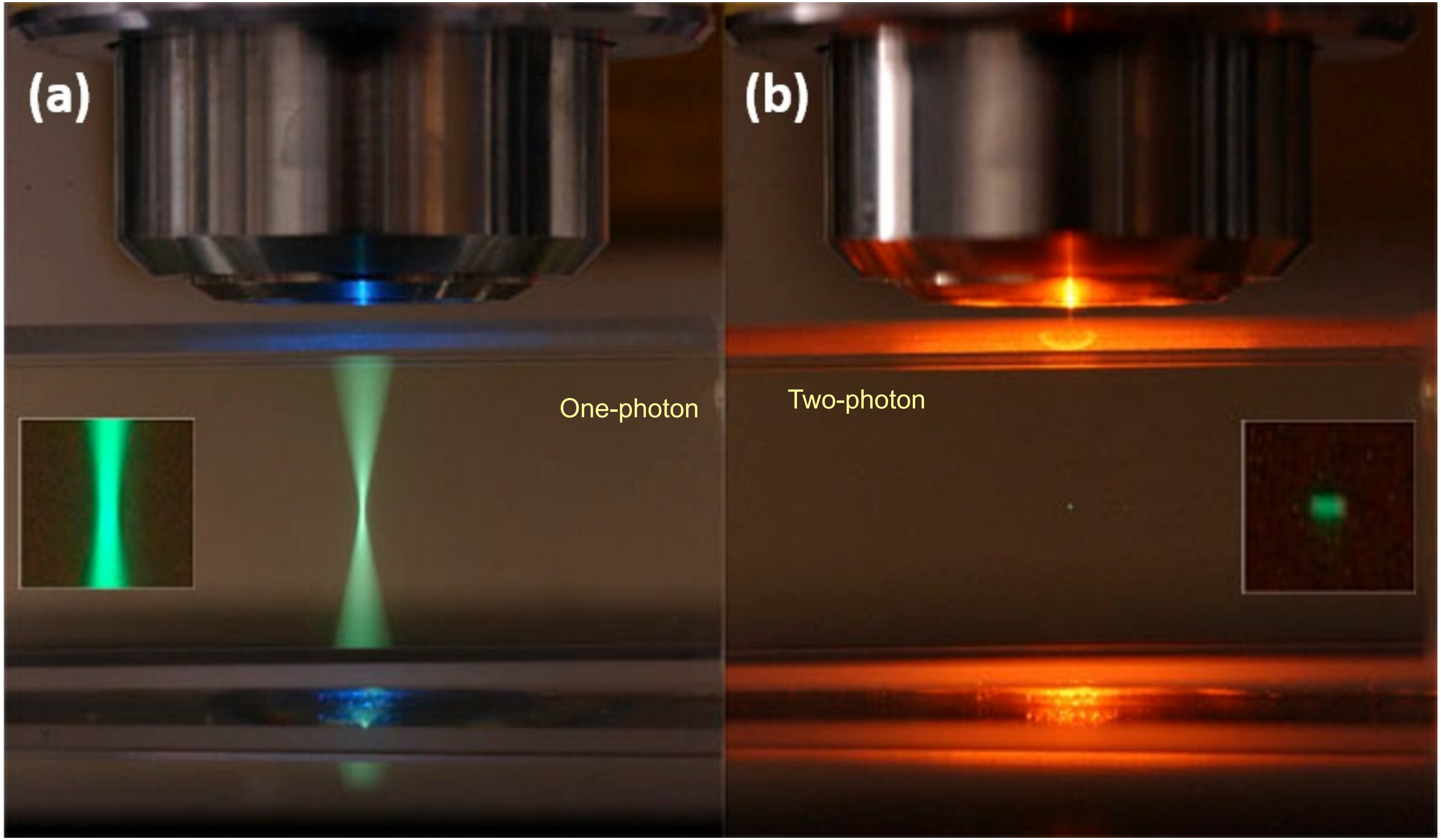
2-photon absorption (2PA) is a $\chi^{(3)}$ process: $\omega = \omega + \omega - \omega$

3-photon absorption (3PA) is a $\chi^{(5)}$ process: $\omega = \omega + \omega - \omega + \omega - \omega$

4-photon absorption (4PA) is a $\chi^{(7)}$ process: $\omega = \omega + \omega - \omega + \omega - \omega + \omega - \omega$

....

Two-photon absorption



Is the energy conserved in the self-phase modulation (SPM) process?

Is the energy conserved in the SPM process?

$Now\ \chi^{(3)} is\ real$ (far from resonances)

$Im\{\chi^{(3)}\} = 0$

Slowly varying envelope approximation (SVEA) equation

$$\frac{\partial E(z)}{\partial z} = -\frac{i\omega c}{2n}\mu_0 P = -\frac{i\omega}{2\varepsilon_0 cn} P$$

$$P(\omega) = \frac{3}{4}\epsilon_0\chi^{(3)}|E_1(\omega)|^2 E_1(\omega)$$

$$\frac{dE(z,\omega)}{dz} = -i\frac{3\omega}{8nc}\chi^{(3)}|E(\omega)|^2 E(\omega) \qquad (19.10)$$

Does the power change as a result of SPM?

Power ~ $|E(\omega)|^2 = E(\omega)E^*(\omega)$

$$\frac{d}{dz}|E(\omega)|^2 = \frac{d}{dz}E(\omega)E^*(\omega) = E^*\frac{dE}{dz} + E\frac{dE^*}{dz} = E^*\left(-i\frac{3\omega}{8nc}\chi^{(3)}|E|^2 E\right) + E\left(+i\frac{3\omega}{8nc}\chi^{(3)}|E|^2 E^*\right) = 0$$

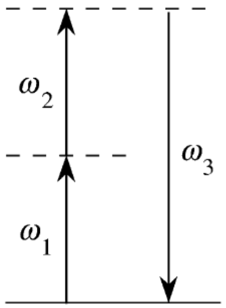
$$\chi^{(3)} = Re\{\chi^{(3)}\}$$

No power change in SPM if $\chi^{(3)}$ is real

Parametric vs non-parametric process

3-photon process

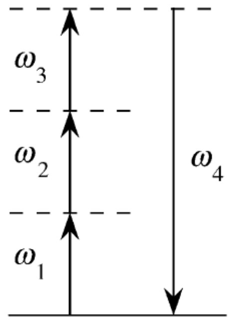
$\omega_3 = \omega_1 + \omega_2$



In a parametric process, population can be removed from the ground state only for those brief intervals of time when it resides in a virtual level. According to the uncertainty principle, population can reside in a virtual level for a time interval of the order of $\sim \frac{\hbar}{\Delta E}$, where ΔE is the energy difference between the virtual level and the nearest real level.

4-photon process

$\omega_4 = \omega_1 + \omega_2 + \omega_3$

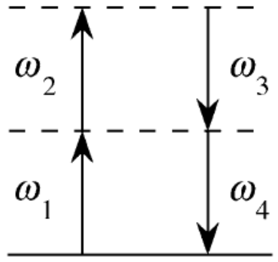


Photon energy is always conserved in a parametric process

4-photon process

$\omega_3 = \omega_1 + \omega_2 - \omega_3$

self-phase modulation
when all ω's are the same



Parametric processes due to four-wave mixing (FWM)

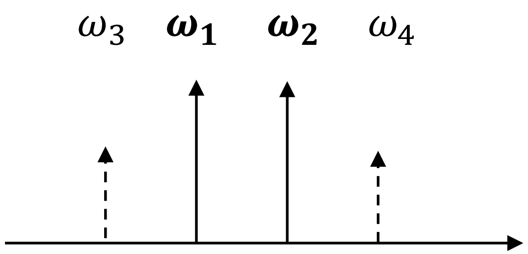
Four-wave mixing (FWM), two pump waves

We have two strong waves $\boldsymbol{\omega_1}$ $\boldsymbol{\omega_2}$

that can create two new waves ω_3 ω_4

in a 4-wave process $\boldsymbol{\omega_1} + \boldsymbol{\omega_2} = \omega_3 + \omega_4$

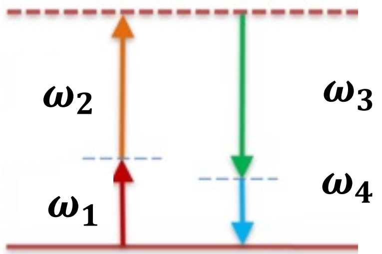
Phase matching is essential here; will talk about it in the next lecture. Assume Δk=0 for now



total field $\mathcal{E}(t) = \frac{1}{2}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + E_3 e^{i\omega_3 t} + E_4 e^{i\omega_4 t} + c.c.)$

total NL polariz. $\mathcal{P}_{NL}(t) = \frac{1}{8}\epsilon_0 \chi^{(3)}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + E_3 e^{i\omega_3 t} + E_4 e^{i\omega_4 t} + c.c.)^3$

8^3 =512 terms



Now let us pick only components with $\pm\omega_3$ and $\pm\omega_4$ - due to interaction of all 4 waves

$$\mathcal{P}_{NL}(t)\big|_{\omega_3} = \frac{1}{8}\epsilon_0 \chi^{(3)}(6E_1 E_2 E_4^* e^{i\omega_3 t} + c.c.) = \frac{3}{4}\epsilon_0 \chi^{(3)}(E_1 E_2 E_4^* e^{i\omega_3 t} + c.c.)$$

$$\mathcal{P}_{NL}(t)\big|_{\omega_4} = \frac{1}{8}\epsilon_0 \chi^{(3)}(6E_1 E_2 E_3^* e^{i\omega_3 t} + c.c.) = \frac{3}{4}\epsilon_0 \chi^{(3)}(E_1 E_2 E_3^* e^{i\omega_3 t} + c.c.)$$

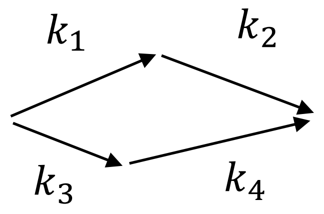
Four-wave mixing (FWM), two pump waves

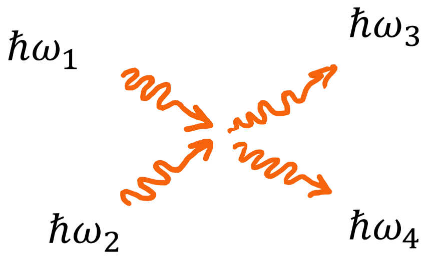
Fourier components for P_{NL}

$$P(\omega_3) = \frac{3}{2}\epsilon_0\chi^{(3)}(E_1E_2E_4^*)$$

$$\rightarrow \qquad\qquad\qquad\qquad\qquad\qquad (19.11)$$

$$P(\omega_4) = \frac{3}{2}\epsilon_0\chi^{(3)}(E_1E_2E_3^*)$$





interaction of 4 photons

Now plug them into SVEA eaquation: $\dfrac{\partial E(\omega_i)}{\partial z} = -\dfrac{i\omega_i}{2nc\epsilon_0}P_{NL}(\omega_i)$

to get:

$$\frac{dE(\omega_3)}{dz} = -\frac{i\omega_3}{2nc\epsilon_0}\frac{3}{2}\epsilon_0\chi^{(3)}(E_1E_2E_4^*)e^{-\Delta kz} = -i\frac{3}{4}\frac{\omega_3\chi^{(3)}}{nc}E_1E_2E_4^*e^{-\Delta kz}$$

$$\frac{dE(\omega_4)}{dz} = -\frac{i\omega_4}{2nc\epsilon_0}\frac{3}{2}\epsilon_0\chi^{(3)}(E_1E_2E_3^*)e^{-\Delta kz} = -i\frac{3}{4}\frac{\omega_4\chi^{(3)}}{nc}E_1E_2E_3^*e^{-\Delta kz}$$

(19.12)

$\Delta k = k_1 + k_2 - k_3 - k_4 + \ldots$

.. plus NL phase modulation terms

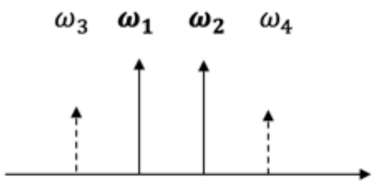
Assume no pump depletion E_1, E_2 - const
and Δk=0 (an additional phase mismatch comes from NL phase modulation)

Four-wave mixing (FWM), two pump waves

$$\frac{dE_3}{dz} = -\, i\frac{3}{4}\frac{\omega_3\chi^{(3)}}{nc}\, E_1 E_2 E_4^*$$

(19.13)

$$\frac{E_4}{dz} = -\, i\frac{3}{4}\frac{\omega_4\chi^{(3)}}{nc}\, E_1 E_2 E_3^*$$



$E_1 = E(\omega_1)$

......

.....

Introduce a new field variables (as in Lecture 5): $A_i = \sqrt{\frac{n_i}{\omega_i}} E_i$

Then it simplifies to:

$\frac{d}{dz}$

$$\frac{d\,A_3}{dz} = -\, igA_1A_2A_4^*$$

(19.10)

*

$$\frac{d\,A_4}{dz} = -\, igA_1A_2A_3^*$$

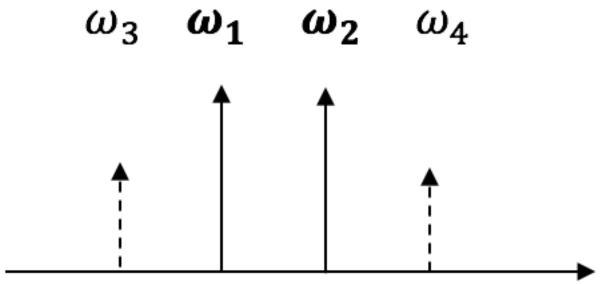
where $\mathrm{g} = \frac{3\chi^{(3)}}{4c}\sqrt{\frac{\omega_1\omega_2\omega_3\omega_4}{n_1n_2n_3n_4}}$

→ $\frac{d^2A_3}{dz^2} - \Gamma^2 A_3 = 0$ and $\frac{d^2A_4}{dz^2} - \Gamma^2 A_4 = 0$ where $\Gamma = |\,A_1||\,A_2|g$

Four-wave mixing (FWM), two pump waves

Similarity to the 3-wave optical parametric amplification (Lecture 12)

The general solutions to this equation are: $e^{\pm\Gamma z}$ or the same as a combination of $cosh(\Gamma z)$ & $sinh(\Gamma z)$



Generation of new frequency components via four-wave mixing.

Look for solutions in the form:

$$A_3 = a_3 cosh(\Gamma z) + b_3 \sinh(\Gamma z)$$

$$A_4 = a_4 cosh(\Gamma z) + b_4 \sinh(\Gamma z)$$

Initial conditions: $A_3 = A_{30}$, $A_4 = 0$

Solution:

$$A_3 = A_{30}\, cosh(\Gamma z)$$

$$A_4 = -iA_{30}^{*} \sinh(\Gamma z) \qquad (19.14)$$

The 4-photon **OPA** uses four-wave mixing to provide optical gain of the 'seed' at ω_3. Simultneously, the wave at ω_4 grows from zero. For each photon created at ω_3 a photon at ω_4 is created

The corresponding photon-flux densities are:

$$\Phi_3 = \Phi_{30} cosh^2(\Gamma z)$$

$$\Phi_4 = \Phi_{30} sinh^2(\Gamma z) \qquad (19.14)$$

In terms of intensities:

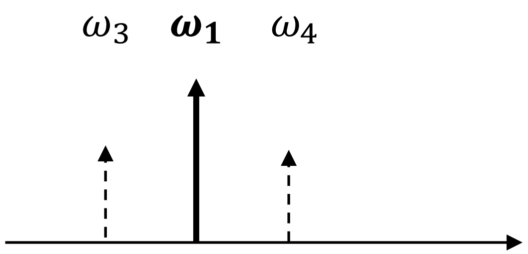
$$I_3 = I_{30}\, cosh^2(\Gamma z)$$

$$I_4 = \frac{\omega_4}{\omega_3} I_{30}\, sinh^2(\Gamma z) \qquad (19.16)$$

These expressions are symmetric under the interchange of ω_3 and ω_4

Four-wave mixing (FWM), one pump wave

(a degenerate four-wave mixing)



One strong wave $\boldsymbol{\omega_1}$ creates two new waves ω_3 ω_4

in a 4-wave process $\boldsymbol{\omega_1} + \boldsymbol{\omega_1} = \omega_3 + \omega_4$

total field $\mathcal{E}(t) = \frac{1}{2}\left(E_1 e^{i\omega_1 t} + E_3 e^{i\omega_3 t} + E_4 e^{i\omega_4 t} + c.c.\right)$

total NL polariz. $\mathcal{P}_{NL}(t) = \frac{1}{8}\epsilon_0 \chi^{(3)}\left(E_1 e^{i\omega_1 t} + E_3 e^{i\omega_3 t} + E_4 e^{i\omega_4 t} + c.c.\right)^3$

Now let us pick only components with $\pm\omega_3$ and $\pm\omega_4$ - due to interaction of all 3 waves

$$\mathcal{P}_{NL}(t)\big|_{\omega_3} = \frac{1}{8}\epsilon_0 \chi^{(3)}\left(3E_1 E_1 E_4^* e^{i\omega_3 t} + c.c.\right) = \frac{3}{8}\epsilon_0 \chi^{(3)}\left(E_1^2 E_4^* e^{i\omega_3 t} + c.c.\right)$$

$$\mathcal{P}_{NL}(t)\big|_{\omega_4} = \frac{1}{8}\epsilon_0 \chi^{(3)}\left(3E_1 E_1 E_3^* e^{i\omega_4 t} + c.c.\right) = \frac{3}{8}\epsilon_0 \chi^{(3)}\left(E_1^2 E_3^* e^{i\omega_4 t} + c.c.\right)$$

Four-wave mixing (FWM), one pump wave

Phase matched, no pump depletion:

note that this coeff is 2 times less than in the non-degenerate case

$$\frac{d}{dz} \qquad \frac{d\,A_3}{dz} = -\,igA_1A_2A_4^*$$

$$* \qquad \frac{d\,A_4}{dz} = -\,igA_1A_2A_3^* \qquad (19.14)$$

where $g = \frac{3\chi^{(3)}}{8c}\sqrt{\frac{\omega_1\omega_2\omega_3\omega_4}{n_1n_2n_3n_4}} \approx \frac{3\chi^{(3)}\omega^2}{8cn^2}$

→ $$\frac{d^2A_3}{dz^2} - \Gamma^2 A_3 = 0$$ and $$\frac{d^2A_4}{dz^2} - \Gamma^2 A_4 = 0$$

where $\Gamma = |\,A_1|^2 g$

Exponential gain coefficient $\mathbf{\Gamma}$ is proportional to I_1 and $\chi^{(3)}$

The corresponding photon-flux densities are:

$$\Phi_3 = \Phi_{30} cosh^2(\Gamma z)$$

$$\Phi_4 = \Phi_{30} sinh^2(\Gamma z) \qquad (19.17)$$

In terms of intensities:

$$I_3 = I_{30}\, cosh^2(\Gamma z)$$

$$I_4 = \frac{\omega_4}{\omega_3} I_{30}\, sinh^2(\Gamma z) \qquad (19.18)$$