Lecture 17

Intensity-dependent refractive index

Optical Kerr effect.

Third-order nonlinear optics

NL polarization density, time domain

NL polarization. Scalar version; no dispersioion of $\chi^{(3)}$

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

Time-domain NL polarization density containing components at different frequencies

Consider $\chi^{(3)}$ to be constant for now

Time-domain, sum of all fields

Third harmonic generation

Third harmonic generation (THG)

Assume that the input field has only one frequency component $\boldsymbol{\omega}$

And assume a scalar version of the 3-wave interaction (the input field is along x-axis)

Fourier component

$$E(t) = \frac{1}{2}(E_1 e^{i\omega t} + c.c.)$$

E_1=$E_1(\omega)$

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

NL polarization (scalar version) in time domain:

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

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

$$= \frac{1}{8}\epsilon_0 \chi^{(3)} \left(E_1 e^{i\omega t} + E_1^* e^{-i\omega t}\right)\left(E_1 e^{i\omega t} + E_1^* e^{-i\omega t}\right)\left(E_1 e^{i\omega t} + E_1^* e^{-i\omega t}\right) = \tag{17.1}$$

$$= \frac{1}{8}\epsilon_0 \chi^{(3)} [\underbrace{E_1^3 e^{3i\omega t} + E_1^{*3} e^{-3i\omega t}}_{\text{Third harmonic}} + \underbrace{3E_1(E_1E_1^*)e^{i\omega t} + 3E_1^*(E_1E_1^*)e^{-i\omega t}}_{\text{Self phase modulation (SPM) terms}}] =$$

Third harmonic generation (THG)

pick only components with $\pm 3\omega$

$$P_{3\omega}(t) \quad \rightarrow \quad \frac{1}{8}\epsilon_0\chi^{(3)}[E_1^3 e^{3i\omega t} + c.c.] = \underbrace{\frac{1}{4}\chi^{(3)}E_1^3}_{\text{ampl.}}\ \frac{1}{2}(e^{3i\omega t} + c.c.)$$

As usual, we now look for a polarization density component at a specific frequency 3ω

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

Fourier component (amplitude)

Hence we get:

$$\boxed{P(3\omega) = \frac{1}{4}\epsilon_0\chi^{(3)}E_1^3(\omega)} \qquad (17.2)$$

- NL polarization at third harmonic 3ω

- will come back to this topic later.

Intensity- dependent refraction
or
self phase modulation (SPM)

Intensity- dependent refraction

Fourier component

$$E(t) = \frac{1}{2}E_1 e^{i\omega t} + c.c.$$

$$E_1 = E_1(\omega)$$

Assume that the input field has only one frequency component **ω**

And assume a scalar version of the 3-wave interaction (all fields are along x-axis)

NL polarization (scalar version) in time domain:

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

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

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

$$= \frac{1}{8}\epsilon_0 \chi^{(3)} \left(E_1 e^{i\omega t} + E_1^* e^{-i\omega t}\right)\left(E_1 e^{i\omega t} + E_1^* e^{-i\omega t}\right)\left(E_1 e^{i\omega t} + E_1^* e^{-i\omega t}\right) =$$

$$= \frac{1}{8}\epsilon_0 \chi^{(3)} [E_1^3 e^{3i\omega t} + c.c. \underbrace{+3E_1(E_1 E_1^*)e^{i\omega t} + c.c.}]$$

pick only components with ±ω → $\frac{3}{8}\epsilon_0 \chi^{(3)} (E_1 |E_1|^2 e^{i\omega t} + c.c.)$

Intensity- dependent refraction

Fourier component

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

What what is the meaning of the $P(\omega) = P^{NL}(\omega)$ term?

Racall relation between polarization density P and the electric field : $P = \epsilon_0 \chi E$ $\qquad \chi = n^2 - 1$

$P^{NL}(\omega)$ is a term that can be described by $P^{NL}(\omega) = \Delta P = \Delta(\epsilon_0\ \chi E_1) = \epsilon_0 \Delta\chi E_1$

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

on the other hand, $\chi = n_0^2 - 1;\quad \Delta\chi = \Delta(n_0^2 - 1) = 2n_0\Delta n$ $\qquad n_0$ - linear ref. index

→ $$\Delta n = \frac{3}{8n_0}\chi^{(3)}|E_1|^2$$ refractive index change induced by high optical intensity

Recall that the intensity $I = \frac{1}{2}c\epsilon_0 n_0|E_1|^2$

Hence $$\Delta n = \frac{3}{8n_0}\chi^{(3)}\frac{2I}{c\epsilon_0 n_0} = \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c}I \qquad (17.3a)$$

Intensity- dependent refraction

The usual way of defining the intensity-dependent refractive index is by means of the equation:

linear ref. index

intensity dependent ref. index [cm^2/W] or [m^2/W]

$$n = n_0 + \underbrace{n_2 I}_{\Delta n} \qquad (17.4)$$

Hence we find that n_2 is related to $\chi^{(3)}$ by

self:

$$\boxed{n_2 = \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c}} \qquad (17.5)$$

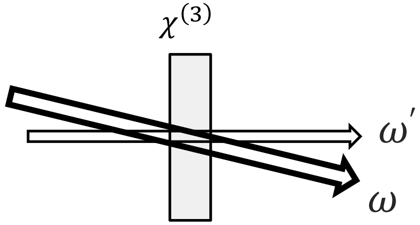
Cross phase modulation (XPM):
two waves with **parallel** polarization

Intensity- dependent refraction: two waves with || polarization

Co-polarized beams

Now we use two separate beams, where the presence of the strong beam of amplitude $E_2(\omega)$ leads to a modification of the refractive index experienced by a weak probe wave of amplitude $E_1(\omega')$. As before, we assume a scalar version of the 3-wave interaction (all fields are along x-axis)

$$E(t) = \frac{1}{2}E_1 e^{i\omega' t} + \frac{1}{2}E_2 e^{i\omega t} + c.c.$$



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

sum of all fields

NL polarization:

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

now there are two fields

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} (\frac{1}{2}E_1 e^{i\omega' t} + \frac{1}{2}E_2 e^{i\omega t} + c.c.)^3 =$$

$$= \frac{1}{8}\epsilon_0 \chi^{(3)} \left(E_1 e^{i\omega' t} + E_1^* e^{-i\omega' t} + E_2\, e^{i\omega t} + E_2^* e^{-i\omega t}\right)$$
$$\times (E_1 e^{i\omega' t} + E_1^* e^{-i\omega' t} + E_2\, e^{i\omega t} + E_2^* e^{-i\omega t})$$
$$\times (E_1 e^{i\omega' t} + E_1^* e^{-i\omega' t} + E_2\, e^{i\omega t} + E_2^* e^{-i\omega t})$$

total 64 terms

○

pick only components with $\pm\omega'$

Intensity- dependent refraction: two waves with || polarization

$$\rightarrow \quad \frac{1}{8}\epsilon_0\chi^{(3)}6\{\ (E_2E_2^*)\ E_1e^{i\omega' t}\ + c.c.\}\ = \frac{3}{4}\epsilon_0\chi^{(3)}(|E_2|^2E_1e^{i\omega' t}\ + c.c.) = \frac{3}{2}\epsilon_0\chi^{(3)}\frac{1}{2}(|E_2|^2E_1e^{i\omega' t}\ + c.c.)$$

Look for the nonlinear polarization component at the angular frequency $\pm\omega'$ in the form $P^{(3)}(t) = \frac{1}{2}P(\omega')e^{i\omega t} + c.c.$

the Fourier component

$$\rightarrow \quad \boxed{P(\omega') = \frac{3}{2}\epsilon_0\chi^{(3)}|E_2|^2E_1} \qquad (17.6)$$

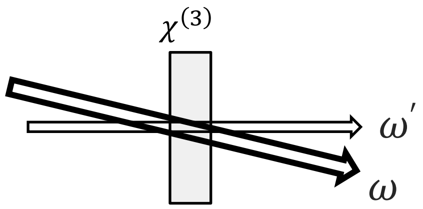
$$\boxed{\chi^{(3)} = \chi^{(3)}_{xxxx}}$$

Hence we find that n_2 in the $\boldsymbol{n = n_0 + n_2 I}$ representation, is given by

cross: $$\boxed{n_2 = \frac{3\chi^{(3)}}{2n_0^2\epsilon_0 c}} \qquad (17.7)$$

Hence, a strong wave affects the refractive index of a weak wave **twice as much** as it affects its own refractive index.

In fact, a strong wave can be of the <u>same frequency</u>, as soon it is distinguishable from the weak wave (say by angle)

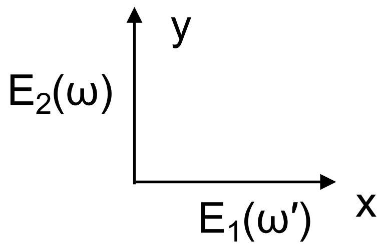


Cross phase modulation (XPM):
two waves with **perpendicular** polarization

Cross-nonlinear refraction: two waves with ⊥ polarization

This is the case commonly encountered in fibers.

Now we use two separate beams with diffferent frequencies and orthogonal polarizations: a weak probe wave of amplitude $E_1(\omega')$ alond x-axis and a strong beam of amplitude $E_2(\omega)$ along y-axis. Now, we us a tensor version of the 3-wave interaction.



Look at **cross-nonlinear refraction**: how a strong beam with the orthogonal polarization changes ref. index of the 'probe' beam

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

$$E_x = \frac{1}{2} E_1 e^{i\omega' t} + c.c.$$

$$E_y = \frac{1}{2} E_2 e^{i\omega t} + c.c.$$

only cross components

$$P_x^{(3)}(t) = \epsilon_0 \{\chi_{xxyy} E_x E_y^2 + \chi_{xyyx} E_x E_y^2 + \chi_{xyxy} E_x E_y^2\} =$$

$$= \epsilon_0 \{\chi_{xxyy} + \chi_{xyyx} + \chi_{xyxy}\} \left(\frac{1}{2} E_1 e^{i\omega' t} + c.c.\right)\left(\frac{1}{2} E_2 e^{i\omega t} + c.c.\right)\left(\frac{1}{2} E_2 e^{i\omega t} + c.c.\right)$$

pick only components with $\pm\omega'$

Cross-nonlinear refraction: two waves with ⊥ polarization

$$\rightarrow \quad \frac{1}{8}\epsilon_0\{\chi_{xxyy}+\chi_{xyyx}+\chi_{xyxy}\}\,2\,(E_2E_2^*)\,(E_1e^{i\omega' t}+c.c.\}=$$

$$=\frac{1}{4}\epsilon_0\{\chi_{xxyy}+\chi_{xyyx}+\chi_{xyxy}\}|E_2|^2(E_1e^{i\omega' t}+c.c.)$$

$$=\frac{1}{4}\epsilon_0\chi_{xxxx}|E_2|^2(e^{i\omega' t}+c.c.)$$

use: $\chi_{xxyy}+\chi_{xyyx}+\chi_{xyxy}=\chi_{xxxx}$

In the Kleinman limit all the cross-polarization terms are equal in isotropic media

Finally we get the Fourier component:

$$\rightarrow \quad \boxed{P(\omega')=\frac{1}{2}\epsilon_0\chi^{(3)}|E_2|^2E_1} \qquad (17.8)$$

$$\boxed{\chi^{(3)}=\chi_{xxxx}^{(3)}}$$

And the main result:

$$\boxed{n_2=\frac{\chi^{(3)}}{2n_0^2\epsilon_0 c}} \qquad (17.9)$$

This is how a strong beam with the orthogonal polarization changes ref. index of the ‘probe’ beam

Self- and cross- nonlinear refraction: summary

$$\boxed{n = n_0 + n_2 I}$$

	NL ref. index n_2	Relative NL ref. index
Self phase modulation (**SPM**)	$n_2 = \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c}$	1
Cross phase modulation (**XPM**): two waves with || polarization	$n_2 = \frac{3\chi^{(3)}}{2n_0^2\epsilon_0 c}$	2
Cross phase modulation (**XPM**): two waves with ⊥ polarization	$n_2 = \frac{\chi^{(3)}}{2n_0^2\epsilon_0 c}$	$\frac{2}{3}$

Cross-nonlinear refraction: summary

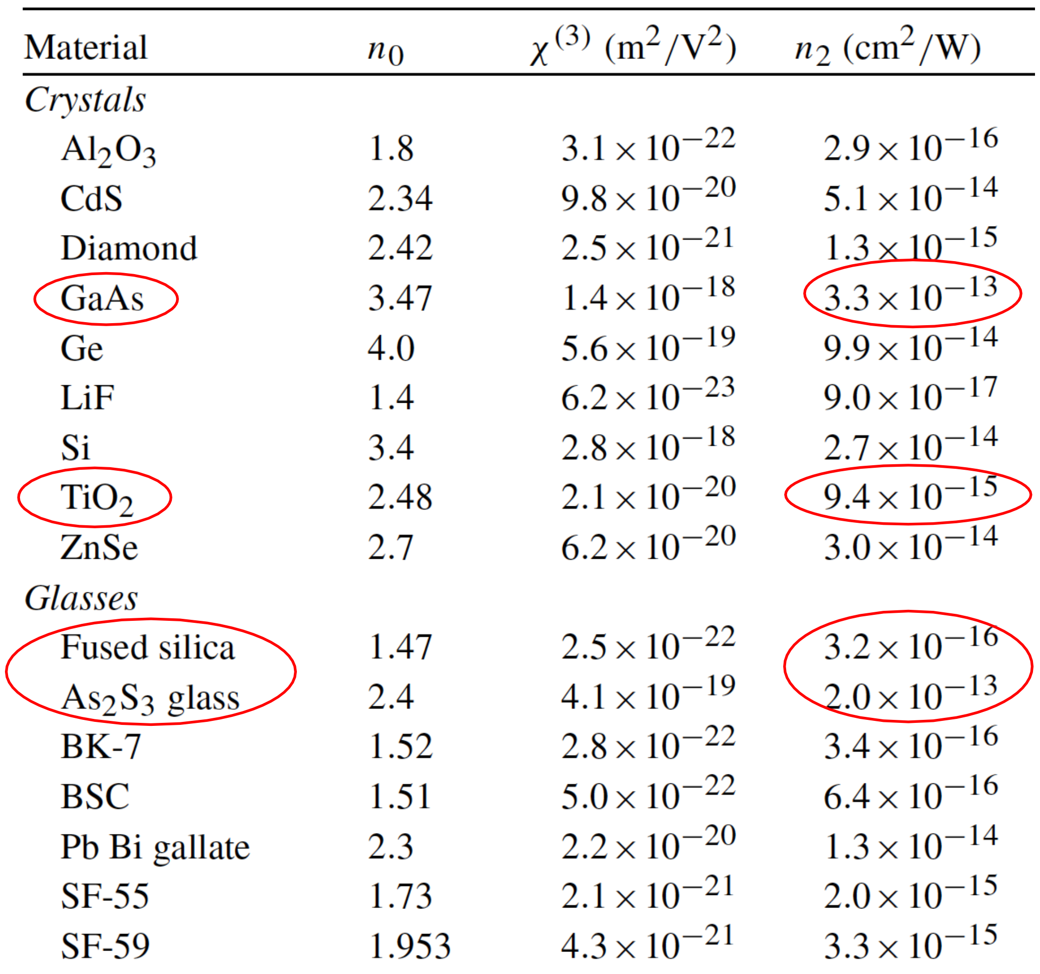
Hence, when two intensities are comparable, for the probe beam we have:

$$\Delta n = n_2(\underset{\nwarrow\,\text{probe beam}}{I_1} + 2\underset{\nwarrow\,\text{pump beam, co-polarized}}{I_2}) \tag{17.10a}$$

$$\Delta n = n_2(\underset{\nwarrow\,\text{probe beam}}{I_1} + \frac{2}{3}\underset{\nwarrow\,\text{pump beam, orthogonally polarized}}{I_2}) \tag{17.10b}$$

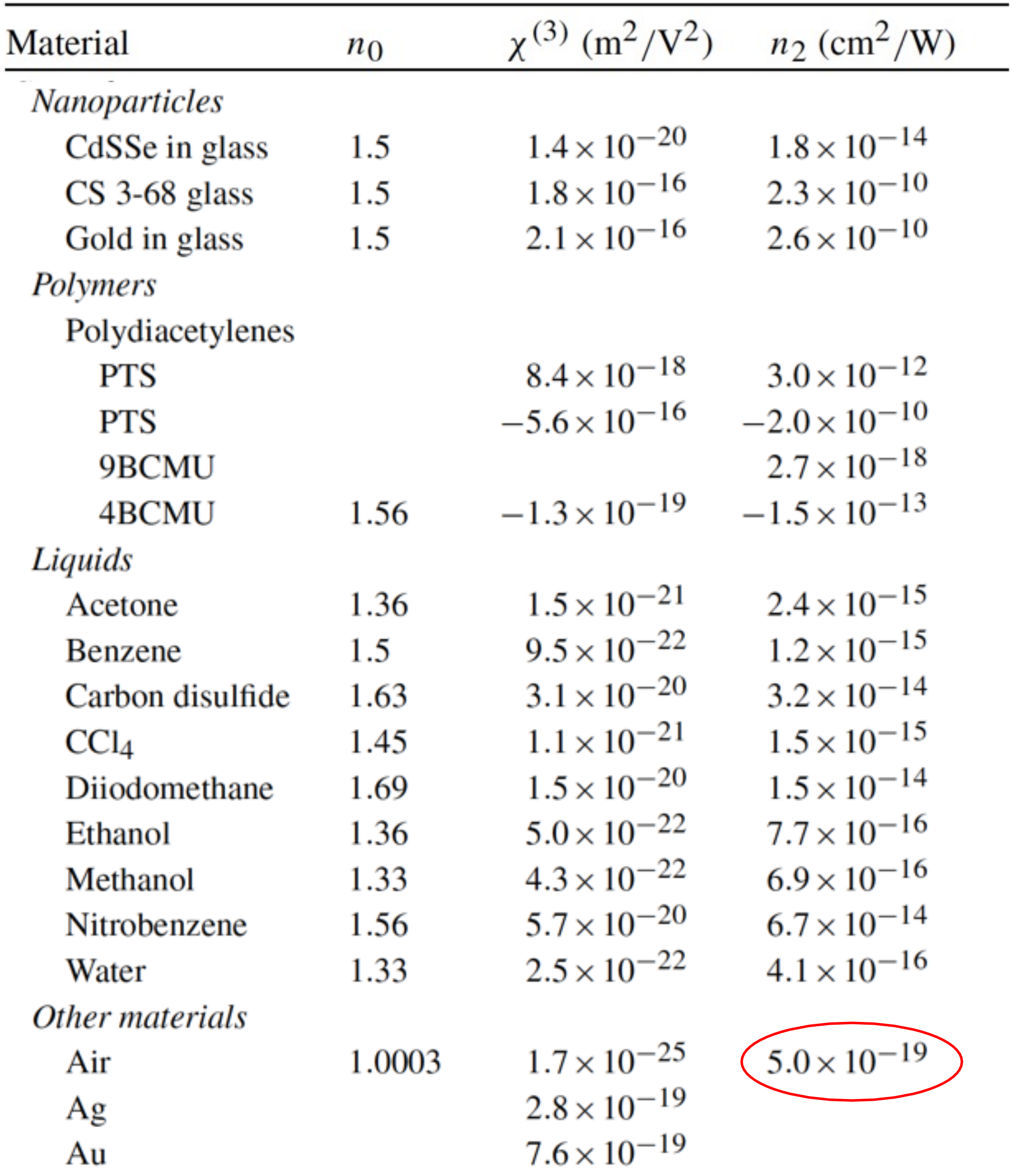
where $n_2 = \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c}$ - **self phase** modulation ceff.

Third-order nonlinear optical coefficients of various materials



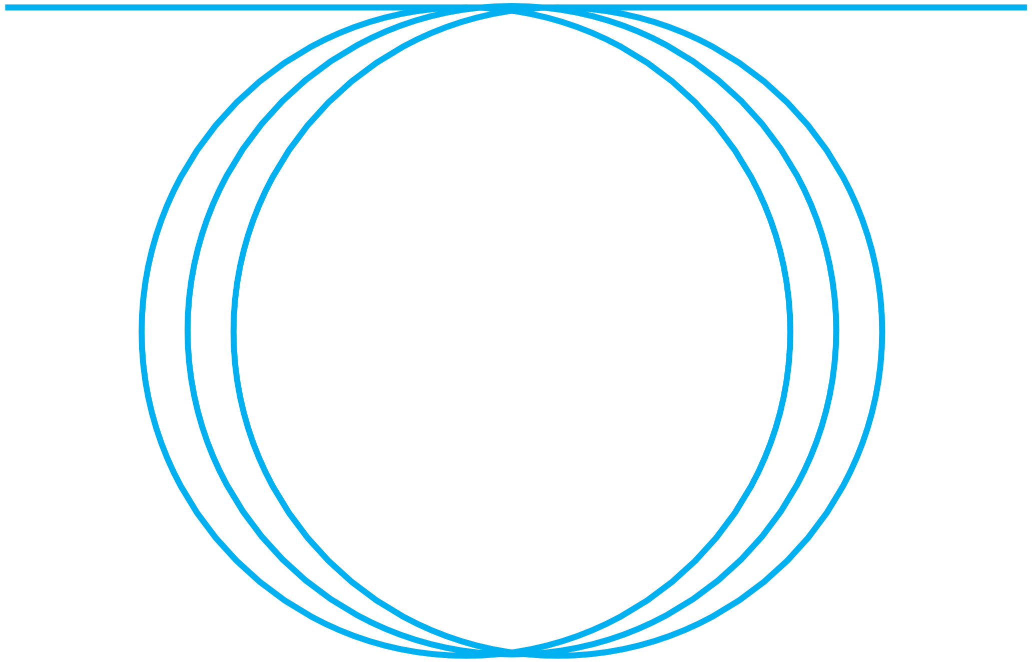
Material	n_0	$\chi^{(3)}$ (m^2/V^2)	n_2 (cm^2/W)
Crystals			
Al_2O_3	1.8	3.1×10^{-22}	2.9×10^{-16}
CdS	2.34	9.8×10^{-20}	5.1×10^{-14}
Diamond	2.42	2.5×10^{-21}	1.3×10^{-15}
GaAs	3.47	1.4×10^{-18}	3.3×10^{-13}
Ge	4.0	5.6×10^{-19}	9.9×10^{-14}
LiF	1.4	6.2×10^{-23}	9.0×10^{-17}
Si	3.4	2.8×10^{-18}	2.7×10^{-14}
TiO_2	2.48	2.1×10^{-20}	9.4×10^{-15}
ZnSe	2.7	6.2×10^{-20}	3.0×10^{-14}
Glasses			
Fused silica	1.47	2.5×10^{-22}	3.2×10^{-16}
As_2S_3 glass	2.4	4.1×10^{-19}	2.0×10^{-13}
BK-7	1.52	2.8×10^{-22}	3.4×10^{-16}
BSC	1.51	5.0×10^{-22}	6.4×10^{-16}
Pb Bi gallate	2.3	2.2×10^{-20}	1.3×10^{-14}
SF-55	1.73	2.1×10^{-21}	2.0×10^{-15}
SF-59	1.953	4.3×10^{-21}	3.3×10^{-15}

for SI units **m^2/W** : to get **m^2/W** - divide this by 10^4



Material	n_0	$\chi^{(3)}$ (m^2/V^2)	n_2 (cm^2/W)
Nanoparticles			
CdSSe in glass	1.5	1.4×10^{-20}	1.8×10^{-14}
CS 3-68 glass	1.5	1.8×10^{-16}	2.3×10^{-10}
Gold in glass	1.5	2.1×10^{-16}	2.6×10^{-10}
Polymers			
Polydiacetylenes			
PTS		8.4×10^{-18}	3.0×10^{-12}
PTS		-5.6×10^{-16}	-2.0×10^{-10}
9BCMU			2.7×10^{-18}
4BCMU	1.56	-1.3×10^{-19}	-1.5×10^{-13}
Liquids			
Acetone	1.36	1.5×10^{-21}	2.4×10^{-15}
Benzene	1.5	9.5×10^{-22}	1.2×10^{-15}
Carbon disulfide	1.63	3.1×10^{-20}	3.2×10^{-14}
CCl_4	1.45	1.1×10^{-21}	1.5×10^{-15}
Diiodomethane	1.69	1.5×10^{-20}	1.5×10^{-14}
Ethanol	1.36	5.0×10^{-22}	7.7×10^{-16}
Methanol	1.33	4.3×10^{-22}	6.9×10^{-16}
Nitrobenzene	1.56	5.7×10^{-20}	6.7×10^{-14}
Water	1.33	2.5×10^{-22}	4.1×10^{-16}
Other materials			
Air	1.0003	1.7×10^{-25}	5.0×10^{-19}
Ag		2.8×10^{-19}	
Au		7.6×10^{-19}	

Third-order nonlinear optical coefficients of various materials



What is the intensity needed to get π phase shift for L=1m fiber at λ=1 μm?

Optical fiber: Fused silica (SiO_2)

$n_2=3.2\times10^{-16}$ cm²/W

$$\Delta\varphi=\Delta(kL)=\frac{2\pi}{\lambda}L\Delta n=\pi$$

$$\Delta n=\frac{1}{2}\frac{\lambda}{L}=5\ 10^{-7}$$

$$\Delta n=In_2$$

$I=5\text{x}10^{-7}/\ 3.2\times10^{-16}$=1.56 GW/cm²

for (10μm)² core, this corresponds to the power of 1.56 kW

- can be achieved only with pulsed lasers

Optical fiber: As_2S_3 glass

$n_2=2.0\times10^{-13}$ cm²/W

for (20μm)² core

- need the power of 10 W only

DC Kerr effect

Electro-optic effect with DC field (DC Kerr effect)

Assume that we have an optical field $Re\{E_1 e^{i\omega t}\}$ in the presence of a DC field E_{DC} such that $|E_1|<<|E_{DC}|$

Fourier component

$$E(t) = \frac{1}{2} E_1 e^{i\omega t} + c.c.$$

NL polarization (scalar version) in time domain:

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

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

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} (\frac{1}{2} E_1 e^{i\omega t} + \frac{1}{2} E_1^* e^{-i\omega t} + E_{DC})^3 =$$

$$= \frac{1}{2} \epsilon_0 \chi^{(3)}\, 3\, E_{DC}^2 \left(E_1 e^{i\omega t} + c.c\right) + \cdots$$

components with $\pm\omega$

components with $0, \pm 2\omega, \pm 3\omega$

frequency set:

$$\omega_p + \omega_q + \omega_r$$

p, q, r = -ω, ω, 0

ω+ω+ω=3ω	1	-ω-ω-ω=-3ω	1
ω+ω+0=2ω	3	-ω-ω+0=-2ω	3
ω+0+0=ω	3	-ω+0+0=-ω	3
ω-ω+ω=ω	3	-ω+ω-ω=-ω	3
ω-ω+0=0	3	ω-ω+0=0	3
0+0+0=0	1		

total 27

Electro-optic effect with DC field (DC Kerr effect)

pick only components with $\pm\omega$

$$P^{(3)}(t) = \frac{3}{2}\epsilon_0\chi^{(3)}\ E_{DC}^2\left(E_1 e^{i\omega t} + c.c\right)$$

$$\rightarrow \qquad P^{NL}(\omega) = 3\epsilon_0\chi^{(3)}E_{DC}^2 E_1(\omega)$$

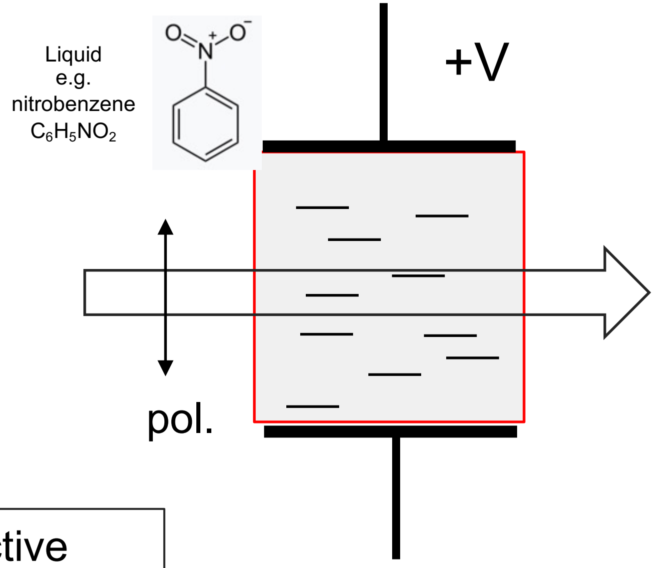
on the other hand $\quad P^{NL}(\omega) = \Delta P = \Delta(\epsilon_0\ \chi E_1) = \epsilon_0\Delta\chi E_1$

$$\rightarrow \qquad \Delta\chi = 3\chi^{(3)}E_{DC}^2$$

$$\Delta\chi = \Delta(n_0^2 - 1) = 2n_0\Delta n$$

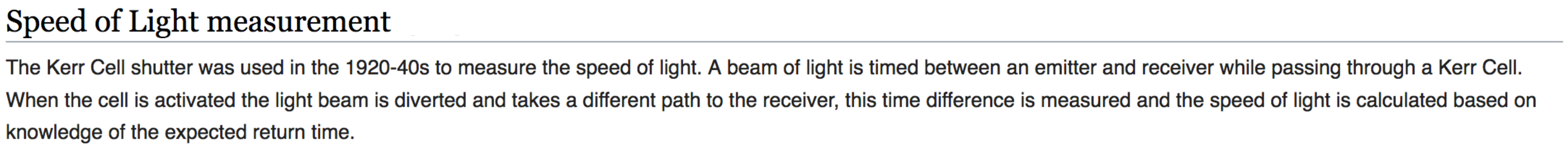
Here $\chi^{(3)} = \chi^{(3)}_{xxxx}$ but not the same as optical $\chi^{(3)}$

$$\boxed{\Delta n = \frac{3}{2n_0}\chi^{(3)}E_{DC}^2} \qquad (17.11)$$



Kerr effect – the quadratic electro-optic effect – a change in the refractive index of a material in response to an applied electric field.

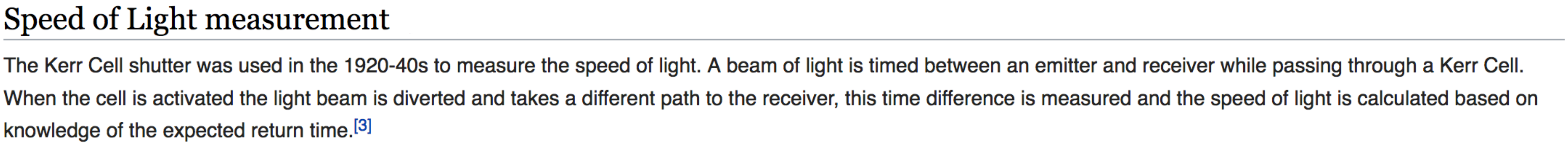
- not the same as linear EO effect in crystals - it is quadratic !



Speed of Light measurement

The Kerr Cell shutter was used in the 1920-40s to measure the speed of light. A beam of light is timed between an emitter and receiver while passing through a Kerr Cell. When the cell is activated the light beam is diverted and takes a different path to the receiver, this time difference is measured and the speed of light is calculated based on knowledge of the expected return time.

Electro-optic effect with DC field (DC Kerr effect)



Speed of Light measurement

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