NLO Lecture 2: Important Nonlinear Optical Processes

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1.2 How does Nonlinear Optics work?

P: Polarization (Dipole moment / unit volume)

p: dipole moment per atom or molecule
***N*: Number density**

$$\mathbf{P} = N\mathbf{p}$$

***q*: charge that is displaced**
l: displacement

$$\mathbf{p} = q \cdot \mathbf{l}$$

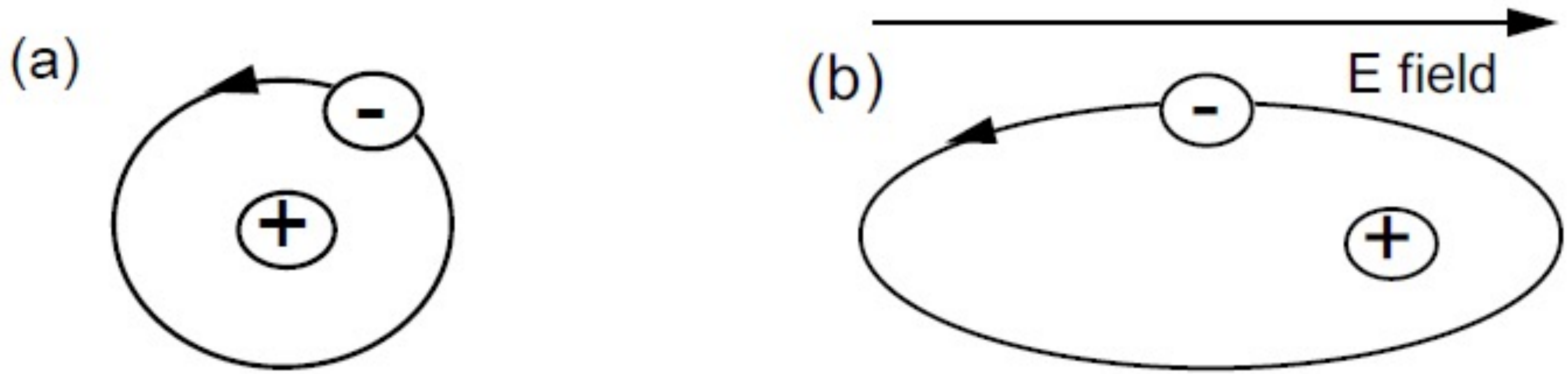


Figure 1.1: **A simple atom model** explaining the effect of in optical electric field on the induced polarization in an atom: (a) without field, (b) with field.

Perturbation Expansion

p: nonlinear dipole moment of atom or molecule

$$\mathbf{p} = q\mathbf{l} = q\left\{\alpha^{(1)}\left(\frac{E}{E_a}\right) + \alpha^{(2)}\left(\frac{E}{E_a}\right)^2 + \alpha^{(3)}\left(\frac{E}{E_a}\right)^3 + \cdots\right\}\frac{\mathbf{E}}{|\mathbf{E}|}. \qquad (1.1)$$

$\alpha^{(i)}$: typical excursion of electron cloud at the critical field is on the order of the Bohr radius

$$\alpha^{(i)} = d_a = 10^{-10}\mathrm{m}$$

E_a : critical field where perturbation theory breaks down: ionization field strength

$$E_a = \frac{e_0}{4\pi\epsilon_0 d_a^2} = 1.4 \cdot 10^{11}\frac{V}{m} = 1.4GV/cm, \qquad (1.2)$$

$$\epsilon_0 = 8.854 \cdot 10^{-12}\ \mathrm{F/m\ the\ vacuum\ dielectric\ constant}$$

Estimate for nonlinear susceptibilities

1 mol, i.e. the typical density is $N_A = 6 \cdot 10^{23}$ cm^{-3}

Nonlinear susceptibilities

$$P = \epsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right], \qquad (1.3)$$

order i	$\chi^{(i)}$	model value	typ. material value
1	$\chi^{(1)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a} \approx 7.5$	n=2.9	Quartz: n=1.45
2	$\chi^{(2)}$ … $= 5.$		
3	$\chi^{(3)}$ … $= 3.$		

Table 1.2: Linear and nonlinear optical susceptibilities from a simple atom model. We used n_0(KDP)= 2.3, $d_a = \alpha^{(i)} = 10^{-10}$ m, $e = e_0 = 1.6 \cdot 10^{-19}$ C, $\epsilon_0 = 8.854 \cdot 10^{-12}$ F/m, $E_a = \frac{e_0}{4\pi\epsilon_0 d_a^2} = 1.4 \cdot 10^{11}$ V/m, $N = 6 \cdot 10^{23} \cdot 10^6 \mathrm{m}^{-3}$.

Estimate for (nonlinear) susceptibilities

Refractive index:

$$n^2 = \left(1 + \chi^{(1)}\right) . \tag{1.4}$$

As table (1.2) shows, the model predicts

$$\chi^{(1)} = \frac{N e_0 d_a}{E_a \varepsilon_0} \tag{1.5}$$

refractive index $n = 2.9$

About right!

1.3 Important nonlinear optical processes

Let's assume:

that there are two waves with angular frequencies ω_1 and ω_2 and resulting wave numbers, then the second order term includes

$$E^2 = \left(\hat{E}_1 \cos\left(\omega_1 t - k_1 z + \varphi_1\right) + \hat{E}_2 \cos\left(\omega_2 t - k_2 z + \varphi_2\right)\right)^2 . \qquad (1.6)$$

or

$$\begin{aligned} E^2 &= \hat{E}_1^2 \cos^2\left(\omega_1 t - k_1 z + \varphi_1\right) + \hat{E}_2^2 \cos^2\left(\omega_2 t - k_2 z + \varphi_2\right) \\ &+ 2\hat{E}_1 \hat{E}_2 \cos\left(\omega_1 t - k_1 z + \varphi_1\right) \cos\left(\omega_2 t - k_2 z + \varphi_2\right) . \qquad (1.7) \end{aligned}$$

Using the addition theorem of the Cosine-function

$$\cos(\alpha) \cdot \cos(\beta) = \frac{1}{2}\left[\cos(\alpha+\beta) + \cos(\alpha-\beta)\right]$$

we find

$$\begin{aligned} E^2 &= \frac{1}{2}\left(\hat{E}_1^2 + \hat{E}_2^2\right) \\ &+ \frac{1}{2}\left(\hat{E}_1^2 \cos\left[2\left(\omega_1 t - k_1 z + \varphi_1\right)\right] + \hat{E}_2^2 \cos\left[2\left(\omega_2 t - k_2 z + \varphi_2\right)\right]\right) \\ &+ \hat{E}_1 \hat{E}_2 \cos\left((\omega_1 - \omega_2)t + (k_1 - k_2)z + \varphi_1 - \varphi_2\right) \qquad (1.8) \\ &+ \hat{E}_1 \hat{E}_2 \cos\left((\omega_1 + \omega_2)t + (k_1 + k_2)z + \varphi_1 + \varphi_2\right) . \end{aligned}$$

Important nonlinear processes

susceptibility	nonlinear process
$\chi^{(2)}(2\omega_1;\omega_1,\omega_1)$	frequency doubling
$\chi^{(2)}(\omega_3;\omega_1,\pm\omega_2)$	sum- and difference-frequency generation, 2-photon absorption, saturable absorption
$\chi^{(2)}(\omega_1;\omega_1,0)$	linear electro-optic effect, Pockels effect
$\chi^{(2)}(0;\omega_1,-\omega_1)$	optical rectification
$\chi^{(3)}(\omega_1;\omega_1,0,0)$	DC Kerr effect
$\chi^{(3)}(\omega_1;\omega_1,\omega_1,-\omega_1)$	self-phase modulation, self-focusing 2-photon absorption, saturable absorption
$\chi^{(3)}(2\omega_1;\omega_1,\omega_1,0)$	field-induced second-harmonic generation
$\chi^{(3)}(3\omega_1;\omega_1,\omega_1,\omega_1)$	frequency tripling
$\chi^{(3)}(\omega_2;\omega_2,\omega_1,-\omega_1)$	stimulated Raman scattering $(\omega_{vib}=\omega_2-\omega_1)$
$\chi^{(3)}(2\omega_1-\omega_2;\omega_1,\omega_1,-\omega_2)$	four-wave mixing, CARS $(\omega_{vib}=\omega_2-\omega_1)$

SFG process

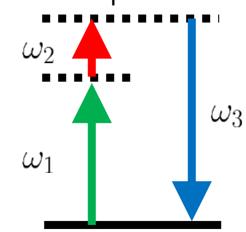
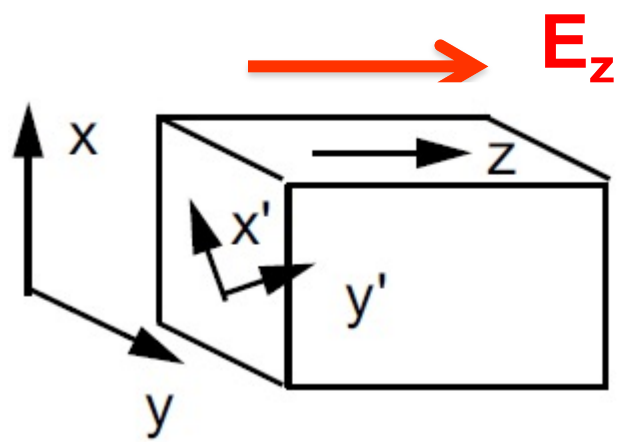


Table 1.3: Important nonlinear optical susceptibilities and corresponding nonlinear optical processes. The first argument in the susceptibility gives the frequency of the generated wave and the other arguments after the semicolon give the frequency components of the input waves.

1.3.1 Linear electro-optical or Pockels Effect

KDP: Potassium dihydrogen phosphat: KH_2PO_4



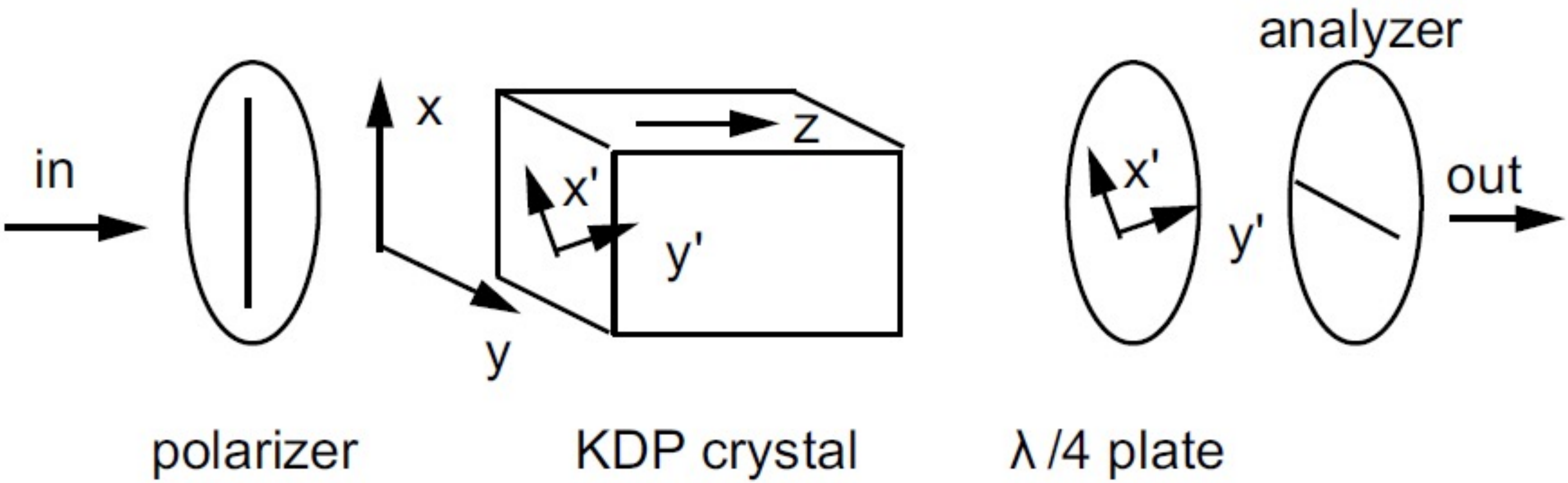
KDP crystal

Induced birefringence when electric field is applied in z-direction

$$\Delta\phi = k(n_{x'} - n_{y'})L$$

Electro-optic modulator

KDP: Potassium dihydrogen phosphat:



Induzed birefringence when electric field is applied in z-direction

$$\Delta\phi = k(n_{x'} - n_{y'})L$$

Electro-optic modulator

$$\epsilon = n^2 = 1 + \chi \quad \Rightarrow \quad n^2 - 1 = \chi = \chi^{(1)} + \chi^{(2)} E + \frac{3}{4}\chi^{(3)}|E|^2 + \cdots$$

$$\text{or} \qquad n^2 = (n_0 + \Delta n)^2 \approx n_0^2 + 2n_0\Delta n.$$

$$\Delta n = \frac{\chi^{(2)} E_z}{2n_0} = n_{x'} - n_{y'}$$

$$V_\pi = E_z \cdot L$$

$$V_\pi = \frac{\lambda n_0}{\chi^{(2)}}$$

$$T = \frac{1}{2}\left[1 + \sin\left(\pi \frac{V}{V_\pi}\right)\right]$$

Modulator transmission

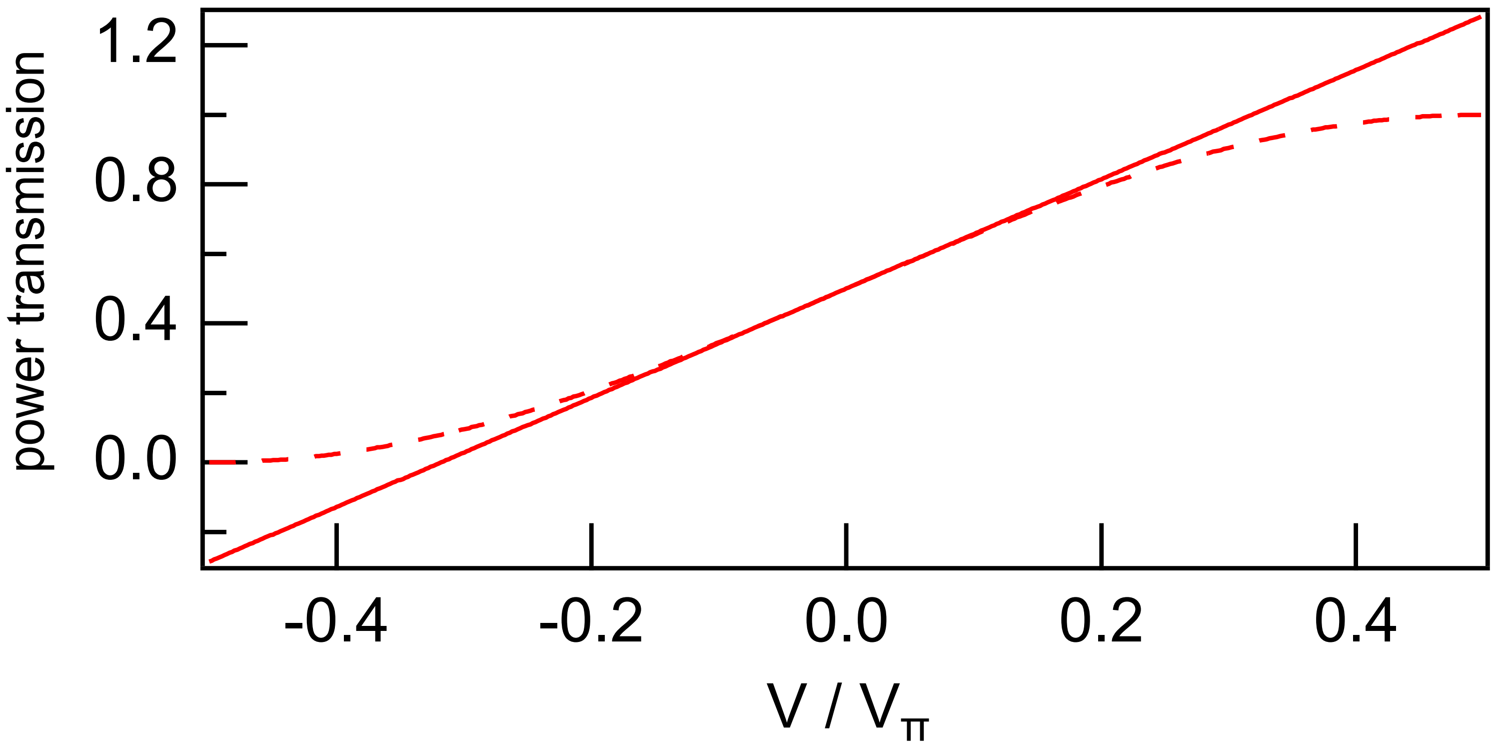


Figure 1.3: Transmission through an electro-optic modulator

Estimate for nonlinear susceptibilities

1 mol, i.e. the typical density is $N_A = 6 \cdot 10^{23}\ \mathrm{cm}^{-3}$

Nonlinear susceptibilities

$$P = \epsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ \cdots \right], \tag{1.3}$$

order i	$\chi^{(i)}$	model value	typ. material value
1	$\chi^{(1)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a} \approx 7.5$	n=2.9	Quartz: n=1.45
2	$\chi^{(2)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a^2} = 5.4 \cdot 10^{-11}\ \frac{\mathrm{m}}{\mathrm{V}}$	$V_\pi = \frac{\lambda n_0}{\chi^{(2)}} = 30$ kV	KDP: $V_\pi = 7.5$ kV
3	$\chi^{(3)}$ $= 3.$		

Table 1.2: Linear and nonlinear optical susceptibilities from a simple atom model. We used n_0(KDP)= 2.3, $d_a = \alpha^{(i)} = 10^{-10}$ m, $e = e_0 = 1.6 \cdot 10^{-19}$ C, $\epsilon_0 = 8.854 \cdot 10^{-12}$ F/m, $E_a = \frac{e_0}{4\pi\epsilon_0 d_a^2} = 1.4 \cdot 10^{11}$ V/m, $N = 6 \cdot 10^{23} \cdot 10^6 \mathrm{m}^{-3}$.

1.3.2 Self-phase modulation

$I \approx \left|\hat{E}\right|^2/(2Z_0)$ according to

$$n = n_0(\omega) + n_{2I} I, \qquad (1.13)$$

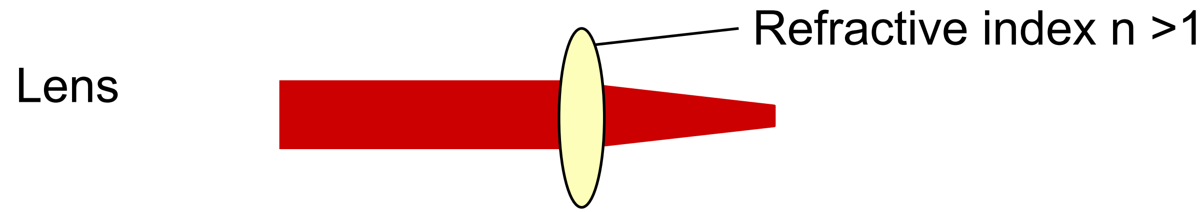
Z_0 is the impedance of the wave in the medium

$$\Delta n = n_{2I} I = \frac{3}{4}\frac{\chi^{(3)}|E|^2}{2n_0} = \frac{3}{4}\frac{\chi^{(3)} Z_0 I}{n_0}.$$

$$n_{2I} = \frac{3}{4}\frac{\chi^{(3)} Z_0}{n_0^2} = \frac{3}{4}\frac{\chi^{(3)}}{\varepsilon_0 c_0 n_0^2}$$

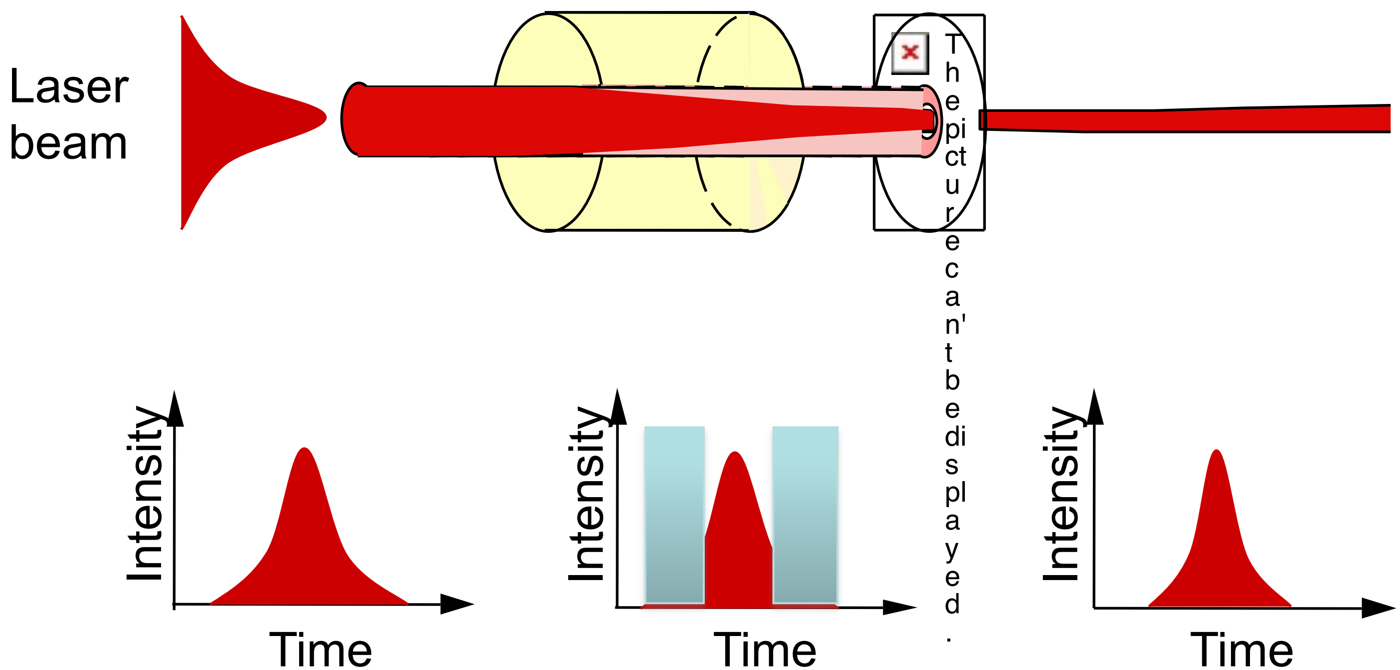
order i	$\chi^{(i)}$	model value	typ. material value
1	$\chi^{(1)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a} \approx 7.5$	n=2.9	Quartz: n=1.45
2	$\chi^{(2)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a^2} = 5.4 \cdot 10^{-11}\,\frac{\mathrm{m}}{\mathrm{V}}$	$V_\pi = \frac{\lambda n_0}{\chi^{(2)}} = 30$ kV	KDP: $V_\pi = 7.5$ kV
3	$\chi^{(3)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a^3} = 3.7 \cdot 10^{-22}\,\frac{\mathrm{m}^2}{\mathrm{V}^2}$	$n_2 = \frac{3\chi^{(3)}}{4n_0^2\epsilon_0 c_0} = 1.25 \cdot 10^{-20}\,\frac{\mathrm{m}^2}{\mathrm{W}}$	Quartz: $n_2 = 3.2 \cdot 10^{-20}\,\frac{\mathrm{m}^2}{\mathrm{W}}$

1.3.3 Self-focusing



Intensity-dependent refractive index: "Kerr Lens" **catastrophic self-focusing**

Kerr-Lens Mode locking

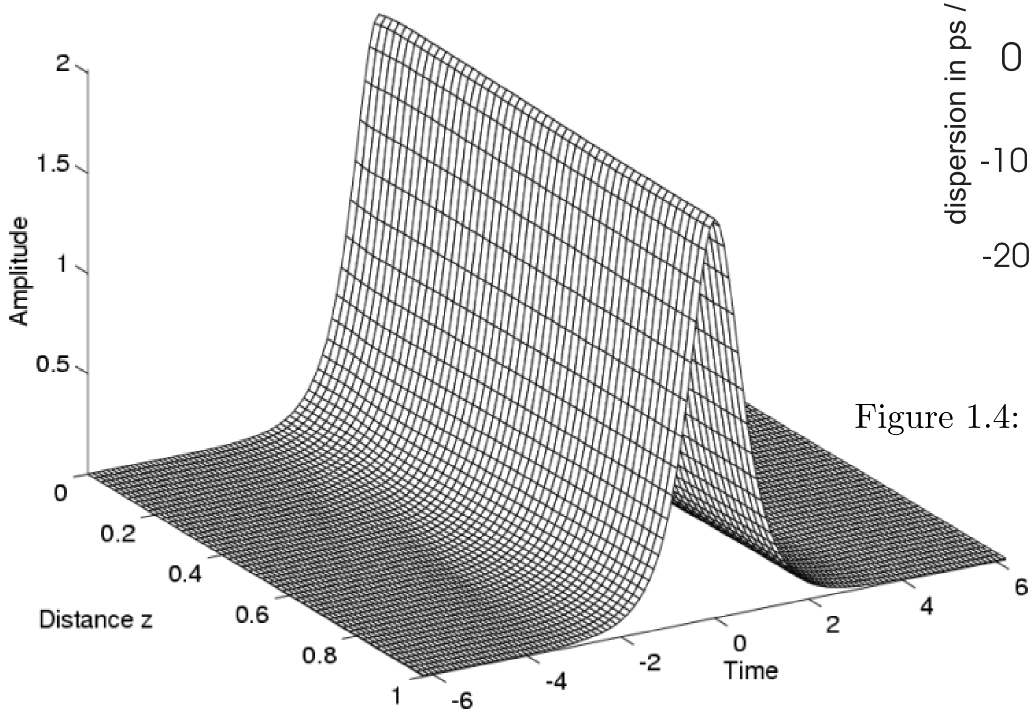


1.3.4 Optical Solitons

Nonlinear Schrödinger Equation (NLSE)

$$j\frac{\partial A(z,t)}{\partial z} = -D_2\frac{\partial^2 A}{\partial t^2} + \delta|A|^2 A \qquad D_2 = \frac{\beta_2}{2} \qquad \beta_2 \text{ is GVD.}$$

$$A_s(t) = A_0 \frac{1}{\cosh\left(\frac{t}{\tau}\right)},$$



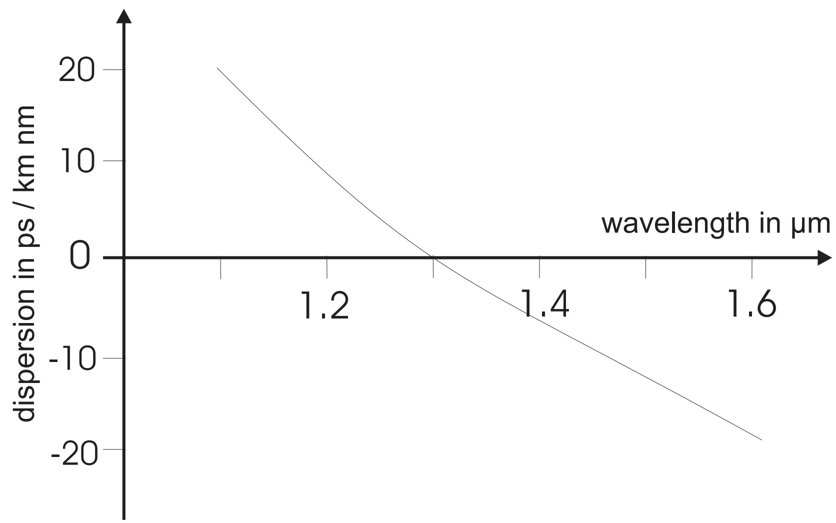


Figure 1.4: Disperson of standard optical fiber in practical units.

2 Nonlinear optical susceptibilities

A general electric field can be written as a superposition of waves with different frequencies (sum or integral)

$$\mathbf{E}(\mathbf{r},t) = \sum_{\omega_a>0} \sum_{i=1}^{3} \frac{1}{2} \left\{ \hat{E}_i(\omega_a) e^{j(\omega_a t - \mathbf{k}_a \mathbf{r})} + c.c. \right\} \mathbf{e}_i . \tag{2.1}$$

$$E_i(-\omega_a) \; = \; E_i(\omega_a)^*$$

$$\mathbf{P}(\mathbf{r},t) = \sum_n \mathbf{P}^{(n)}(\mathbf{r},t) \tag{2.2}$$

with

$$\mathbf{P}^{(n)}(\mathbf{r},t) = \sum_{\omega_b>0} \sum_{i=1}^{3} \frac{1}{2} \left\{ \hat{P}_i^{(n)}(\omega_b) e^{j(\omega_b t - \mathbf{k}_b \mathbf{r})} + c.c. \right\} \mathbf{e}_i . \tag{2.3}$$

Nonlinear optical susceptibilities

For the i-th component of the n-th order nonlinear polarization with frequenz ω_b we define the susceptibility tensor as

$$\hat{P}_i^{(n)}(\omega_b) = \frac{\varepsilon_0}{2^{m-1}} \sum_P \sum_{j...k} \chi^{(n)}_{ij...k}(\omega_b : \omega_1, ..., \omega_n) \hat{E}_j(\omega_1) \cdots \hat{E}_k(\omega_n), \quad (2.4)$$

$$\omega_b = \sum_{i=1}^{n} \omega_i \text{ and } \mathbf{k}_b = \sum_{i=1}^{n} \mathbf{k}_i. \quad (2.5)$$

where the sum over P is the summation over all possible permutations of frequencies $\omega_1, ..., \omega_n$, that lead to the same resulting frequency ω_b and m is the number of fields with a frequency different from zero. For visualization a few examples

$$\hat{P}_i^{(2)}(\omega_3) = \varepsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3 : \omega_1, \omega_2) \hat{E}_j(\omega_1) \hat{E}_k(\omega_2), \quad (2.6)$$

$$\omega_3 = \omega_1 + \omega_2 \text{ and } \mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2. \quad (2.7)$$

($\longrightarrow$ Sum Frequency Generation, SFG)

Nonlinear optical susceptibilities

$$\hat{P}_i^{(2)}(\omega_3) = \varepsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3 : \omega_1, -\omega_2)\hat{E}_j(\omega_1)\hat{E}^*_k(\omega_2), \tag{2.8}$$

$$\omega_3 = \omega_1 - \omega_2 \text{ und } \mathbf{k}_3 = \mathbf{k}_1 - \mathbf{k}_2. \tag{2.9}$$

($\longrightarrow$ Differenz Frequency Generation, DFG)

$$\hat{P}_i^{(3)}(\omega_4) = \frac{6\varepsilon_0}{4} \sum_{jkl} \chi^{(3)}_{ijkl}(\omega_4 : \omega_1, \omega_2, -\omega_3)\hat{E}_j(\omega_1)\hat{E}_k(\omega_2)\hat{E}^*_l(\omega_3), \tag{2.10}$$

$$\omega_4 = \omega_1 + \omega_2 - \omega_3 \text{ und } \mathbf{k}_4 = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3. \tag{2.11}$$

($\longrightarrow$ Four Wave Mixing, FWM)

Remember, the susceptibilities are symmetric with respect to a permutation of the input frequencies $\{\omega_i\}$, since it is arbitrary which frequency is considered to be ω_1,i.e.there is

$$\chi^{(n)}_{ijk}(\omega : \omega_1, \omega_2, ...) = \chi^{(n)}_{ikj}(\omega : \omega_2, \omega_1, ...). \tag{2.12}$$

2.2 Classical model for nonlinear optical susceptibility

$$F(x) = -\frac{\partial V(x)}{\partial x} = -m\omega_0^2 x\left(1 + \frac{x}{a} + \frac{x^2}{b^2}\right) \qquad (2.13)$$

$$= -m\omega_0^2 x - m\beta_2 x^2 - m\beta_3 x^3 \qquad (2.14)$$

$$\text{with } \beta_2 = \frac{\omega_0^2}{a} \text{ and } \beta_3 = \frac{\omega_0^2}{b^2}\,. \qquad (2.15)$$

$$m\frac{d^2x}{dt^2} = -2\frac{\omega_0}{Q}m\frac{dx}{dt} + F(x) - e_0 E(t)$$

$$\frac{d^2x}{dt^2} + 2\frac{\omega_0}{Q}\frac{dx}{dt} + \omega_0^2 x + \beta_2 x^2 + \beta_3 x^3 = -\frac{e_0}{m}E(t). \qquad (2.16)$$

Perturbation Solution:

$$|\beta_2 x + \beta_3 x^2| \ll \omega_0^2 \qquad x(t) = x_0(t) + \epsilon x_1(t) + \epsilon^2 x_2(t) + \ldots$$

Zero order solution

$$(0) \quad : \quad \left(\frac{d^2}{dt^2} + 2\frac{\omega_0}{Q}\frac{d}{dt} + \omega_0^2\right) x_0(t) = -\frac{e_0}{m}E(t) \tag{2.18}$$

$$(1) \quad : \quad \left(\frac{d^2}{dt^2} + 2\frac{\omega_0}{Q}\frac{d}{dt} + \omega_0^2\right) x_1(t) = -\beta_2 \left(x_0\right)^2 - \beta_3 \left(x_0\right)^3 \tag{2.19}$$

$$(2) \quad : \quad \left(\frac{d^2}{dt^2} + 2\frac{\omega_0}{Q}\frac{d}{dt} + \omega_0^2\right) x_2(t) = -2\beta_2 x_0 x_1 - 3\beta_3 x_0^2 x_1 \tag{2.20}$$

2.2.1 Linear Susceptibility

$$\begin{aligned} x_0(t) &= \frac{1}{2}\left(\hat{x}_0(\omega)e^{j\omega t} + c.c.\right) \\ P^{(1)}(t) &= \frac{1}{2}\left(\hat{P}^{(1)}(\omega)e^{j\omega t} + c.c.\right) = -Ne_0 \cdot x_0(t) \end{aligned}$$

in case of a time-varying field with amplitude $E(\omega)$ and frequency ω

$$E(t) = \frac{1}{2}\left(\hat{E}(\omega)e^{j\omega t} + c.c.\right) \qquad (2.21)$$

Eq. (2.18) with $x_0(t)$ or its Fourier transforms

$$(0) \quad : \quad \hat{x}_0(\omega) = \frac{-e_0}{m\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)}\hat{E}(\omega),$$

$$(1) \quad : \quad \hat{P}^{(1)}(\omega) = \frac{Ne_0^2}{m\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)}\hat{E}(\omega) = \varepsilon_0\chi^{(1)}\hat{E}(\omega).$$

Therefore, the linear susceptibility is

$\omega_P = \sqrt{\frac{Ne_0^2}{m\varepsilon_0}}$ is the plasma frequency

$$\chi^{(1)}(\omega) = \frac{Ne_0^2}{m\varepsilon_0\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)} = \frac{\omega_P^2}{\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)} \qquad (2.22)$$

Real and Imaginary Part of the Susceptibility

$$\chi^{(1)} = \chi^{(1)\prime} + j\chi^{(1)\prime\prime} \tag{2.23}$$

$$\chi^{(1)\prime} = \frac{\omega_P^2}{\omega_0^2} \frac{\left(1 - \frac{\omega^2}{\omega_0^2}\right)}{\left[\left(1 - \frac{\omega^2}{\omega_0^2}\right)^2 + \frac{4}{Q^2}\frac{\omega^2}{\omega_0^2}\right]} \tag{2.24}$$

$$\chi^{(1)\prime\prime} = -\frac{\omega_P^2}{\omega_0^2} \frac{\frac{2}{Q}\frac{\omega}{\omega_0}}{\left[\left(1 - \frac{\omega^2}{\omega_0^2}\right)^2 + \frac{4}{Q^2}\frac{\omega^2}{\omega_0^2}\right]} \tag{2.25}$$

Real and Imaginary Part of the Susceptibility

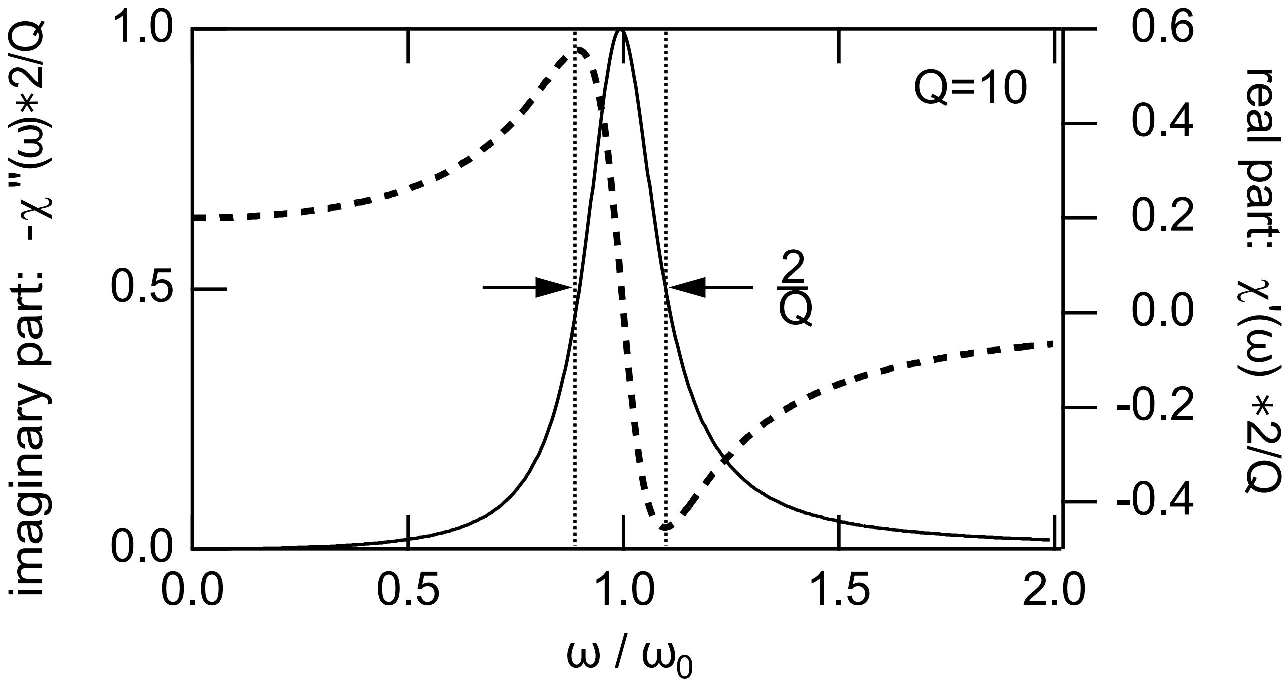


Figure 2.1: Susceptibility arising from the linear harmonic oscillator model for the electron cloud surrounding an atomic core.

Real and Imaginary Part of the Susceptibility

$$
\begin{aligned}
\chi^{(1)}(\omega) &= \frac{\omega_P^2}{\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)} && (2.26)\\
&= \frac{\omega_P^2}{2j\omega_0'}\left[\frac{1}{\left(\frac{1}{Q} + j\left(\omega - \omega_0'\right)\right)} - \frac{1}{\left(\frac{1}{Q} + j\left(\omega + \omega_0'\right)\right)}\right] && (2.27)\\
&\approx \frac{\omega_P^2}{2j\omega_0}\left[\frac{1}{\left(\frac{1}{Q} + j\left(\omega - \omega_0\right)\right)} - \frac{1}{\left(\frac{1}{Q} + j\left(\omega + \omega_0\right)\right)}\right] && (2.28)\\
&\approx \frac{\omega_P^2}{2j\omega_0}\frac{1}{\left(\frac{1}{Q} + j\left(\omega - \omega_0\right)\right)}, \text{für } \omega \quad \text{um } +\omega_0. && (2.29)
\end{aligned}
$$

where $\omega_0' = \omega_0\sqrt{1 - \frac{1}{Q^2}}$ is the exact resonance frequency of the damped harmonic oscillator.

2.2.2. Nonlinear Susceptibility

$$
\begin{aligned}
x_1(t) &= \hat{x}_1(0) + \frac{1}{2}\left(\hat{x}_1(\omega)e^{j\omega t} + c.c.\right) \\
&\quad + \frac{1}{2}\left(\hat{x}_1(2\omega)e^{j2\omega t} + c.c.\right) + \frac{1}{2}\left(\hat{x}_1(3\omega)e^{j3\omega t} + c.c.\right)
\end{aligned}
$$

With the susceptibility $\chi^{(1)}(\omega)$, which is up to the prefactor$-Ne_0/\varepsilon_0$ equal to the impulse repsonse of Eq.(2.18), we can find the first order amplitudes of all the different frequency components according to

$$
\begin{aligned}
\hat{x}_1(0) &= -\beta_2 \frac{1}{\omega_0^2}\left|\left(\frac{-e_0}{m\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)}\right)\right|^2 \left|\hat{E}(\omega)\right|^2 && (2.30) \\
&= -\beta_2 \frac{\chi^{(1)}(0)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-2} \left|\chi^{(1)}(\omega)\right|^2 \left|\hat{E}(\omega)\right|^2, && (2.31) \\
\hat{x}_1(2\omega) &= \frac{-\beta_2}{2}\frac{\chi^{(1)}(2\omega)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-2} \chi^{(1)}(\omega)^2 \hat{E}(\omega)^2, && (2.32) \\
\hat{x}_1(\omega) &= \frac{-3\beta_3}{4}\frac{\chi^{(1)}(\omega)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-3} \left|\chi^{(1)}(\omega)\right|^2 \left(\chi^{(1)}(\omega)\right) && (2.33) \\
&\quad \times \left|\hat{E}(\omega)\right|^2 \hat{E}(\omega), && (2.34) \\
\hat{x}_1(3\omega) &= \frac{-\beta_3}{4}\frac{\chi^{(1)}(3\omega)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-3} \chi^{(1)}(\omega)^3 \hat{E}(\omega)^3. && (2.35)
\end{aligned}
$$

Susceptibilities

$$\chi^{(2)}(0;\omega,-\omega) = -\frac{m\beta_2}{e_0}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-2}\chi^{(1)}(0)\left|\chi^{(1)}(\omega)\right|^2, \quad (2.36)$$

$$\chi^{(2)}(2\omega;\omega,\omega) = \frac{-m\beta_2}{2e_0}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-2}\chi^{(1)}(2\omega)\chi^{(1)}(\omega)^2, \quad (2.37)$$

$$\chi^{(3)}(\omega;\omega,-\omega,\omega) = \frac{-3m\beta_3}{4e_0}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-3}\left|\chi^{(1)}(\omega)\right|^2\left(\chi^{(1)}(\omega)\right)^2, \quad (2.38)$$

$$\chi^{(3)}(3\omega;\omega,\omega,\omega) = \frac{-m\beta_3}{4e_0}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-3}\chi^{(1)}(3\omega)\chi^{(1)}(\omega)^3. \quad (2.39)$$

2.3 Miller's δ-Coefficient

$$\delta_{ijk} = \frac{\chi^{(2)}_{ijk}(2\omega:\omega,\omega)}{\chi^{(1)}_{ii}(2\omega)\chi^{(1)}_{jj}(\omega)\chi^{(1)}_{kk}(\omega)} = \frac{\chi^{(2)}_{ijk}(2\omega:\omega,\omega)}{\left(n^2(2\omega)-1\right)\left(n^2(\omega)-1\right)^2}$$

$$= \frac{-m\beta_2}{2}\frac{\varepsilon_0^2}{N^2 e_0^3}.$$

Experimentally one finds, that these coefficients do not depend strongly on the material for inorganic materials. We assume that the deviation x (see Eq. (2.13)) is the lattice constant with $a \approx (N)^{-1/3}$, then we obtain with Eq. (2.15) for the Miller coefficient

$$|\delta_{ijk}| \approx \frac{m\omega_0^2}{2}\frac{\varepsilon_0^2}{N^{5/3}e_0^3}.$$

$\lambda_0 = 200$ nm, $\omega_0 = 3\pi\cdot$ fs^{-1} $\qquad a = 3 \cdot 10^{-10}$ m^{-1}

$$|\delta_{ijk}| \approx 3.7 \cdot 10^{-12}\frac{V}{m}$$