Nonlinear Optics (WiSe 2018/19)

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Office hour: Tuesday, 9-10 am

Lectures: Fr 8:30-10:00 and 10:15-11:00, SemRm 4, Jungiusstr. 9
Recitations: Fr 11:15-12:00, SemRm 4, Jungiusstr. 9
Start: 19.10.2018

Teaching Assistant:
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Email: tobias.kroh@cfel.de
Office hour: Thursday, 9:30-11 am

Course Secretary: Christine Berber
office 03.095, phone 040-8998-6351, Email: christine.berber@cfel.de

Prerequisites: A basic course in Electrodynamics

Required Text: Class notes will be distributed in class.

Requirements: 9 Problem Sets, Term Paper, and Term paper presentation
Collaboration on problem sets is encouraged.

Grade breakdown: Problem sets (30%), Participation (30%), Term paper (40%)

Recommended Text:
Nonlinear Optics, R. W. Boyd, Academic Press, Third Edition (2008)

Additional References:
The Principles of Nonlinear Optics, Y. R. Shen, J. Wiley & Sons NY (1984).
The Elements of Nonlinear Optics, P. N. Butcher & D. Cotter, Cambridge Studies in Modern Optics 9 (1990).
Nonlinear Fiber Optics, G. P. Agrawal, Academic Press (1998).
Solitons: an introduction, P. G. Drazin & R. S. Johnson, Cambridge Texts In Applied Mathematics, NY (1989).
Fundamentals of Attosecond Optics, Z. Chang, CRC Press (2016).
Attosecond and Strong-Field Physics, C. D. Lin, A.-T. Le, C. Jin, and H. Wei, Cambridge University Press (2018).
Extreme Nonlinear Optics, M. Wegener, Springer (2005).

Syllabus

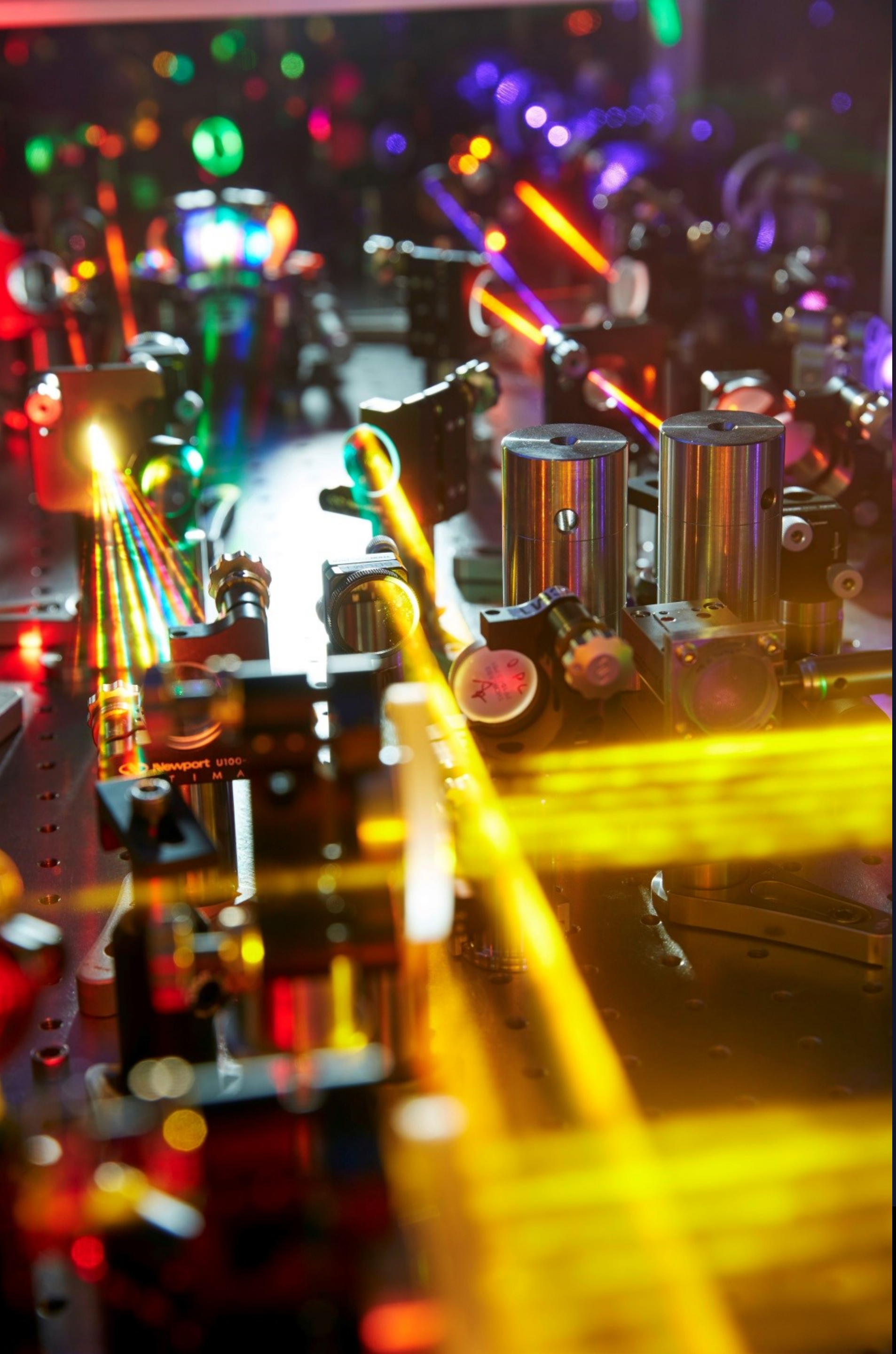
1	Franz Kärtner 19/10/2018	**Introduction to Nonlinear Optics**
2		**Important Nonlinear Optical Processes Overview**
3	Oliver Mücke 26/10/2018	**Nonlinear Optical Susceptibilities** *Problem Set 1 Out*
4		**Susceptibility Tensors**
5	Franz Kärtner 2/11/2018	**Nonlinear Wave Equation** *Problem Set 1 Due, Problem Set 2 Out*
6		**Second-Harmonic Generation**
7	Oliver Mücke 9/11/2018	**Frequency Doubling of Pulses, Quasi-Phase Matching** *Problem Set 2 Due, Problem Set 3 Out*
8		**Optical Parametric Oscillation/Amplification, Difference Frequency Generation**
9	Franz Kärtner 16/11/2018	**Electro-Optic Effect and Modulators** *Problem Set 3 Due, Problem Set 4 Out*
10		**Acousto-Optic Modulators and Bragg Cells**
11	Franz Kärtner 23/11/2018	**Third-Order Nonlinear Effects** *Problem Set 4 Due, Problem Set 5 Out*
12		**Self-Phase Modulation and Self-Focusing**

Syllabus

13	Franz Kärtner 30/11/2018	**Raman and (Stimulated) Brillouin Scattering** *Problem Set 5 Due, Problem Set 6 Out; Distr. Term Paper Proposals*
14		**Optical Solitons**
15	Oliver Mücke 7/12/2018	**Ultrashort-Pulse Optical Parametric Amplification** *Problem Set 6 Due, Problem Set 7 Out*
16		**Ultrashort-Pulse Optical Parametric Chirped-Pulse Amplification**
17	Oliver Mücke 14/12/2018	**High-Energy Few-Cycle Parametric Sources I** *Problem Set 7 Due, Problem Set 8 Out*
18		**High-Energy Few-Cycle Parametric Sources II: NOPA, OPCPA, passive CEP stabilization in OPA**
19	Oliver Mücke 21/12/2018	**Nonlinear Optics with Two-Level Systems** *Problem Set 8 Due, Term Paper Proposal Due*
20		**Carrier-Wave Rabi Flopping**

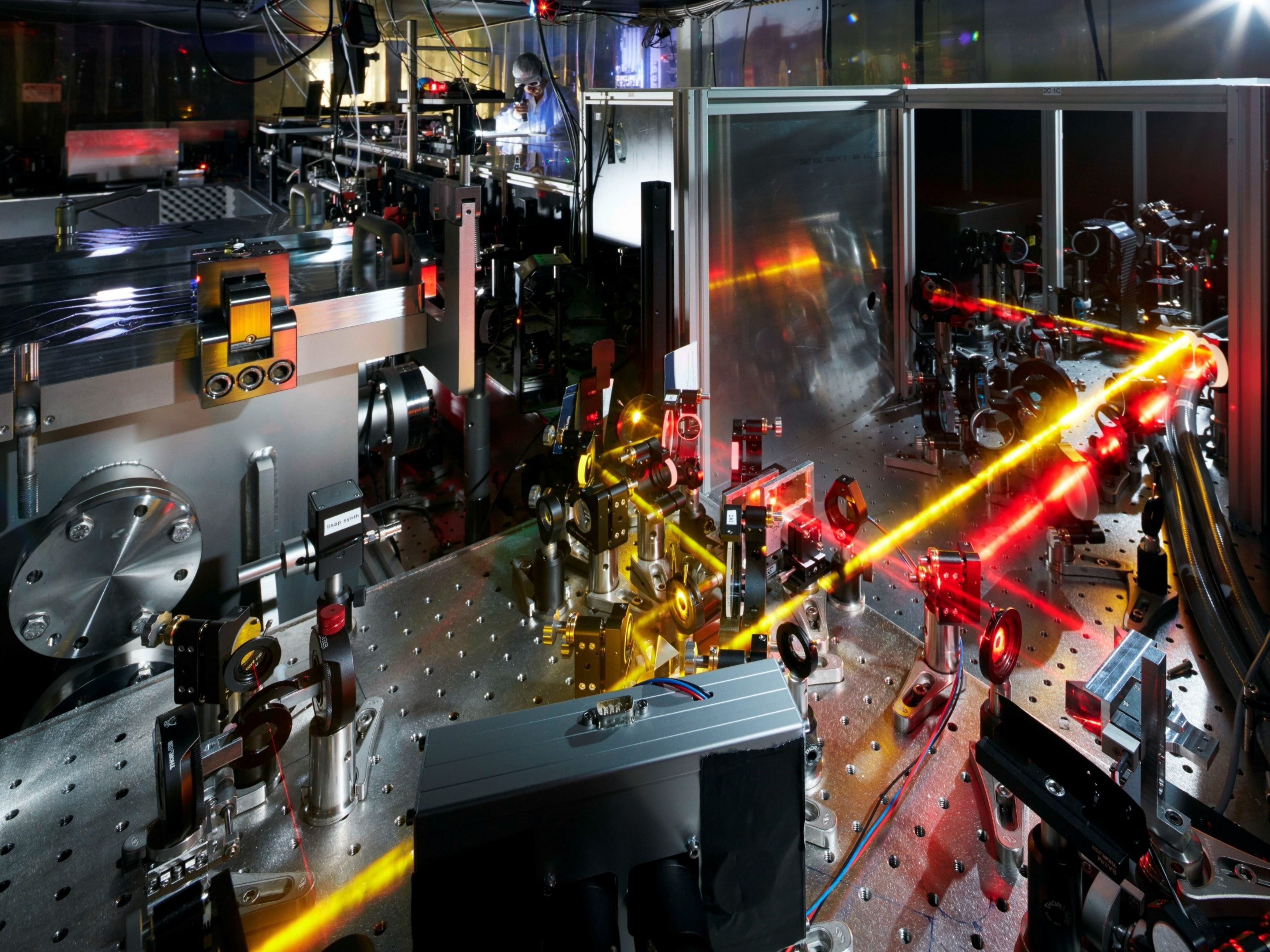
Syllabus

21	Franz Kärtner 11/1/2019	**Ultrafast Terahertz (THz) Sources** *Problem Set 9 Out*
22		**Applications of Ultrafast Terahertz (THz) Sources**
23	Oliver Mücke 18/1/2019	**High-Harmonic Generation** *Problem Set 9 Due*
24		**Attosecond Science**
25	Oliver Mücke 25/1/2019	**Strong-Field Physics in Solids I**
26		**Strong-Field Physics in Solids II**
27	--	**Term Paper Presentation**
28		**Term Paper Presentation**



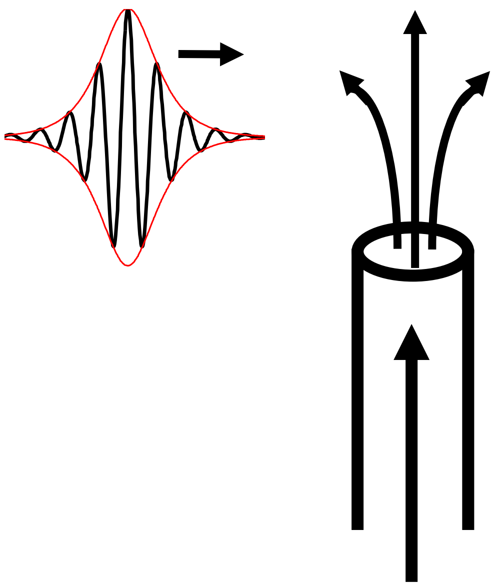
C. Manzoni *et al.* LPR **9**, 129 (2015)

O. D. Mücke *et al.*, IEEE J. Sel. Top. Quantum Electron. **21**, 8700712 (2015)


tisap synth

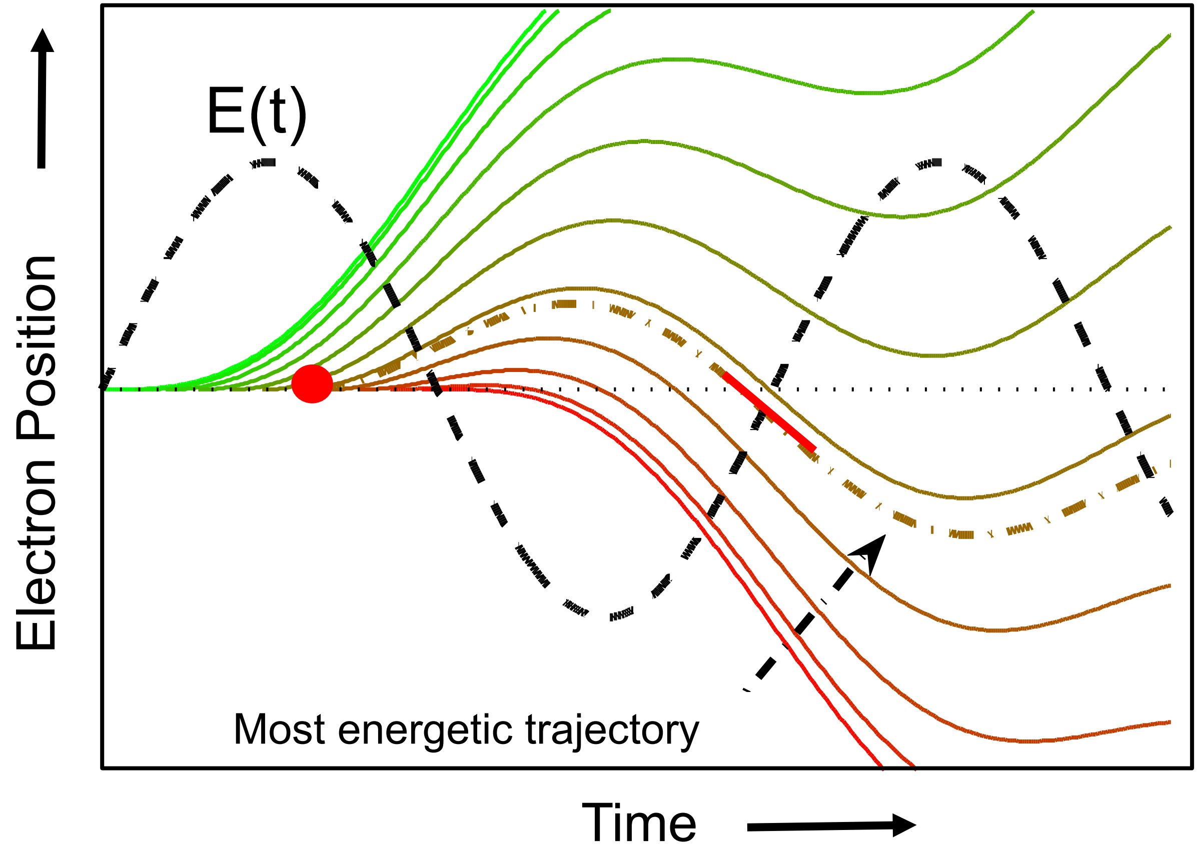
High-order harmonic generation (HHG)

high-energy pulse



gas nozzle

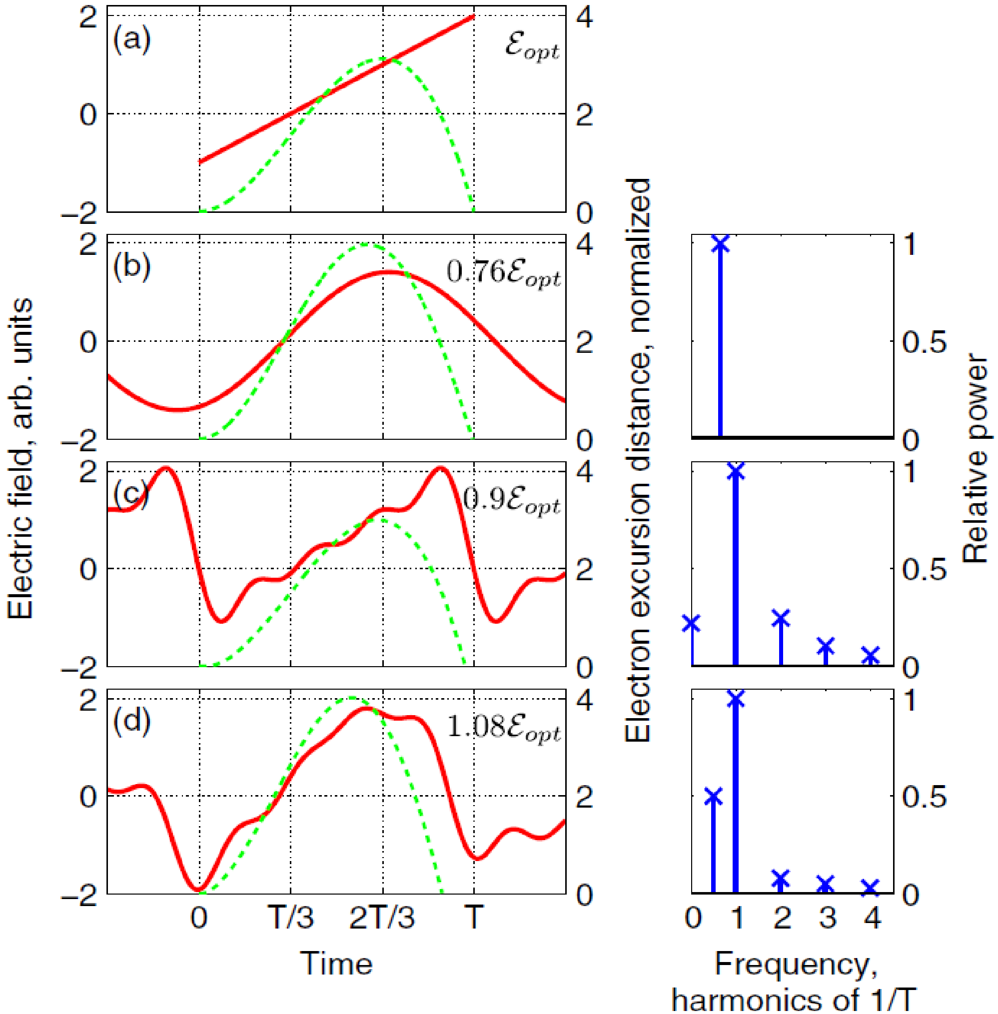
tunnel ionization + propagation + recombination

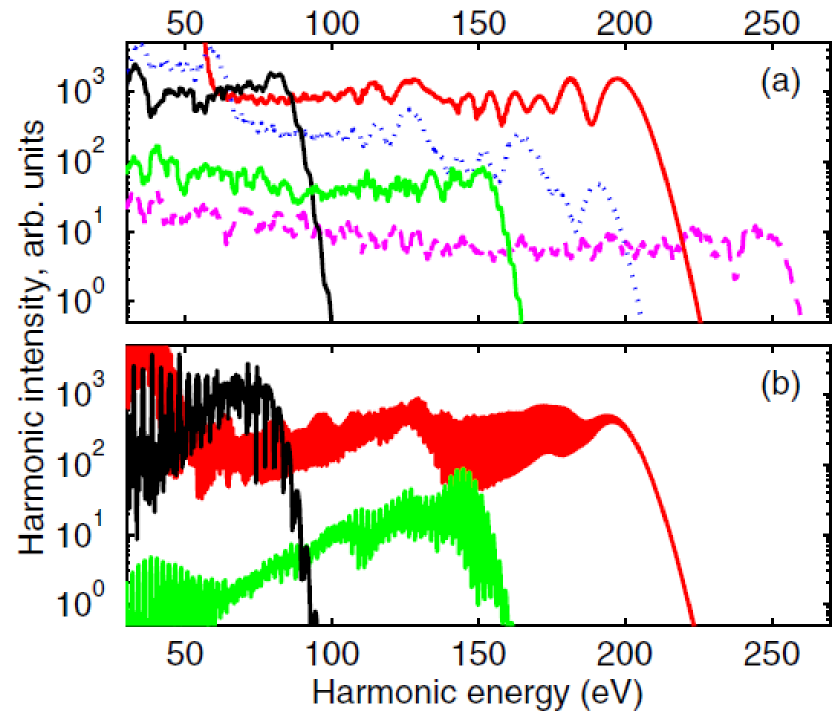


$\hbar\omega_{\mathrm{cutoff}} = I_{\mathrm{p}} + 3.17U_{\mathrm{p}}$ $U_{\mathrm{p}} \propto \lambda^2 I$

M. Y. Kuchiev, JETP Lett. **45**, 404 (1987)
P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993)
K. J. Schafer *et al.*, Phys. Rev. Lett. **70**, 1599 (1993)

Maximizing the recollision energy within an optical period





800nm + 400nm + 267nm + 200nm + 1600nm

ω + 2ω + 3ω + 4ω + 0.5 ω

"perfect waveform" for HHG

maximize	cut-off energy
sinusoidal	~ $3.17U_P$
synthesized	~ $9U_P$

L. E. Chipperfield *et al.*, Phys. Rev. Lett. **102**, 063003 (2009)
C. Jin *et al.*, Nature Commun. **5**:4003 (2014)
S. Haessler *et al.*, Phys. Rev. X **4**, 021028 (2014)

1.1 Why Nonlinear Optics?

- Many devices used in ultrashort pulse laser physics and optical communications are based on nonlinear optical phenomena
- The capacity of optical communication systems are limited by fiber nonlinearities
- The limits to ultrafast laser physics are determined by optical nonlinearities
- Microwave measurement techniques are based on nonlinear optical techniques (electro-optic (EO) sampling)
- Frequency conversion to UV, EUV and the mid-IR and THz region
- Strong-field physics in gases, liquids and solids
- High-order harmonic generation (HHG)
- Laser materials processing
- Nonlinear optical microscopy
- Biomedical applications and laser surgery

Nobel Prize in Physics 2018

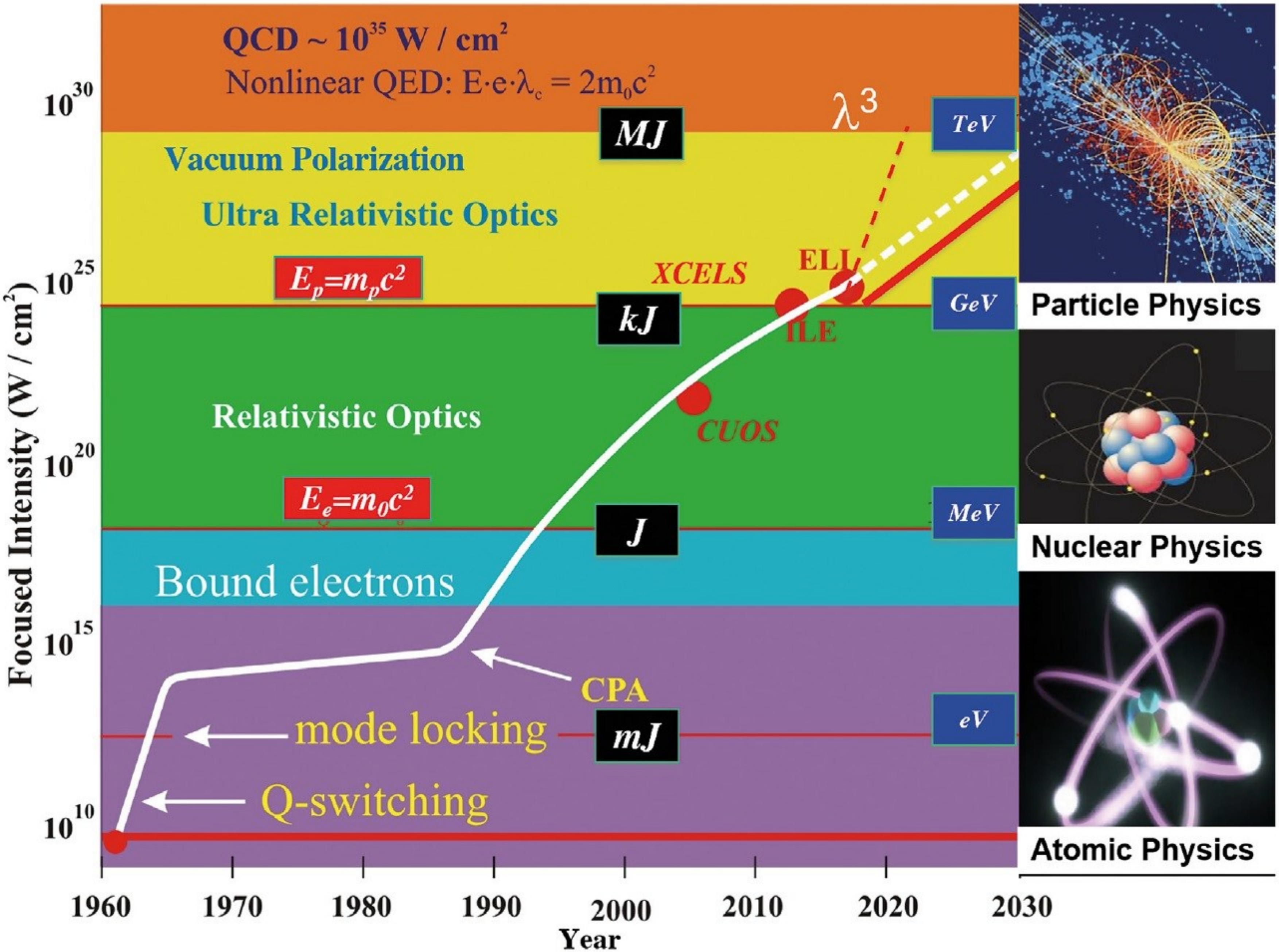
"for the optical tweezers and their application to biological systems"

"for their method of generating high-intensity, ultra-short optical pulses“
chirped-pulse amplification (CPA)

Typical optical nonlinearities are weak

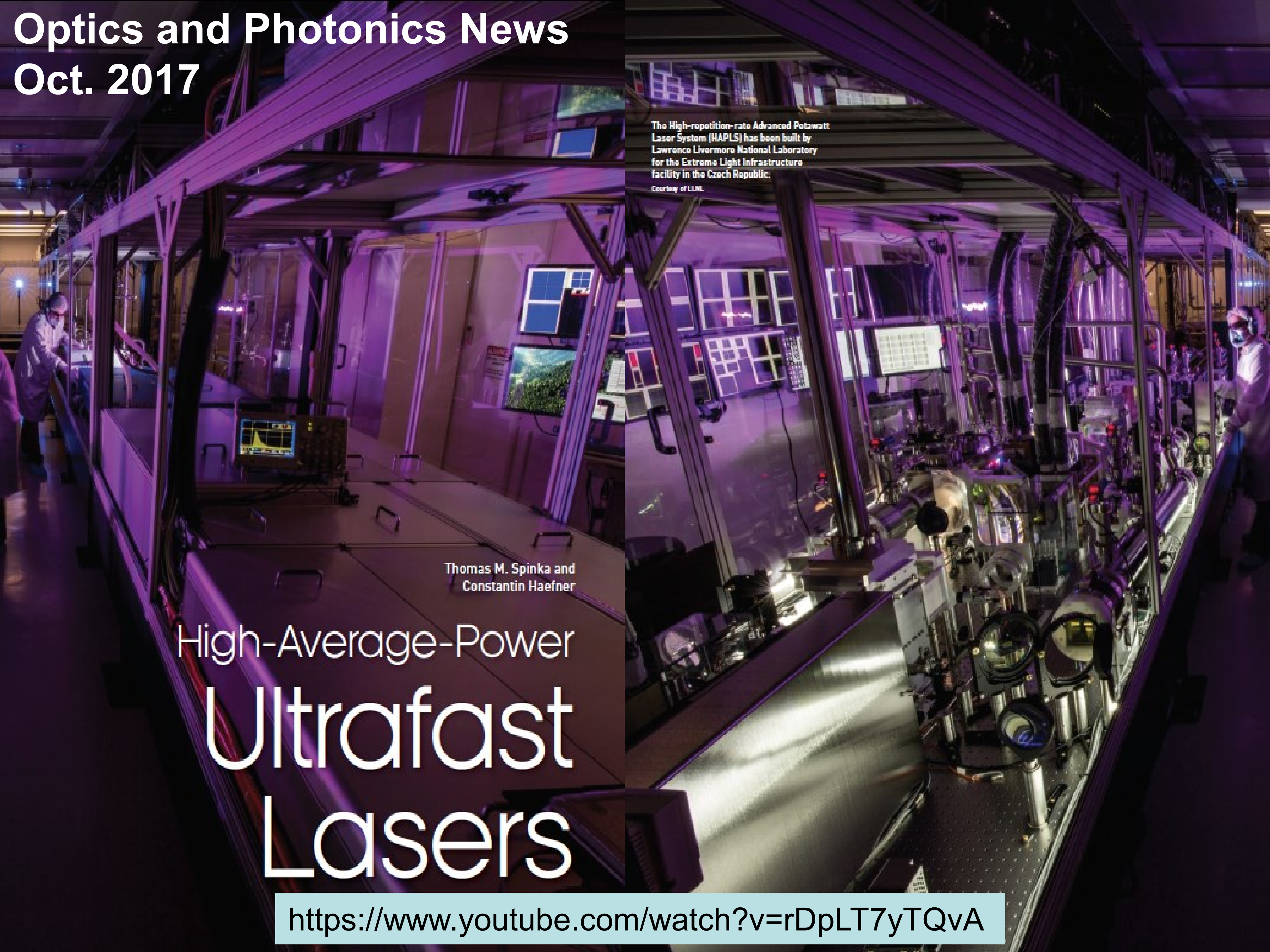
	typ. Ti:sa CPA	L3-HAPLS @ ELI	SULF 10 PW
pulse energy E_p	5 mJ	$\geq$30 J	130 J
repetition rate f_r	3 kHz	10 Hz	
pulse duration τ_p	30 fs	$\leq$30 fs	24 fs
peak power P	166 GW	$\geq$1 PW	5.4 PW
peak intensity I	3×10^{15} W/cm^2	3.5×10^{19} W/cm^2	2×10^{22} W/cm^2
peak field E	1.6 GV/cm	162 GV/cm	3.9 TV/cm

Table 1.1: Ti:sapphire laser source parameters for a typical Ti:sapphire chirped-pulse amplifier (CPA) commonly used in attoscience, the High-Repetition-Rate Advanced Petawatt Laser System (HAPLS) [16] designed/built by Lawrence Livermore National Laboratory (LLNL) for the Extreme Light Infrastructure (ELI), and the Shanghai Superintense Ultrafast Laser Facility (SULF) [17, 18, 19], which will be scaled up to the 100-PW Station of Extreme Light (SEL) until 2023 [17]. $P = E_p/\tau_p$, $I = P/A$. For the typical Ti:sa CPA, $A = \pi r^2$ with $r = 40\,\mu$m. For L3-HAPLS, focusing is assumed to reach a laser strength parameter $a_0 = 4$, with $a_0 = 0.85 \times 10^{-9}\,\lambda[\mu\text{m}]\,(I[\text{W/cm}^2])^{1/2}$. $E = \sqrt{2Z_0 I}$ with $Z_0 = 377\,\Omega$.



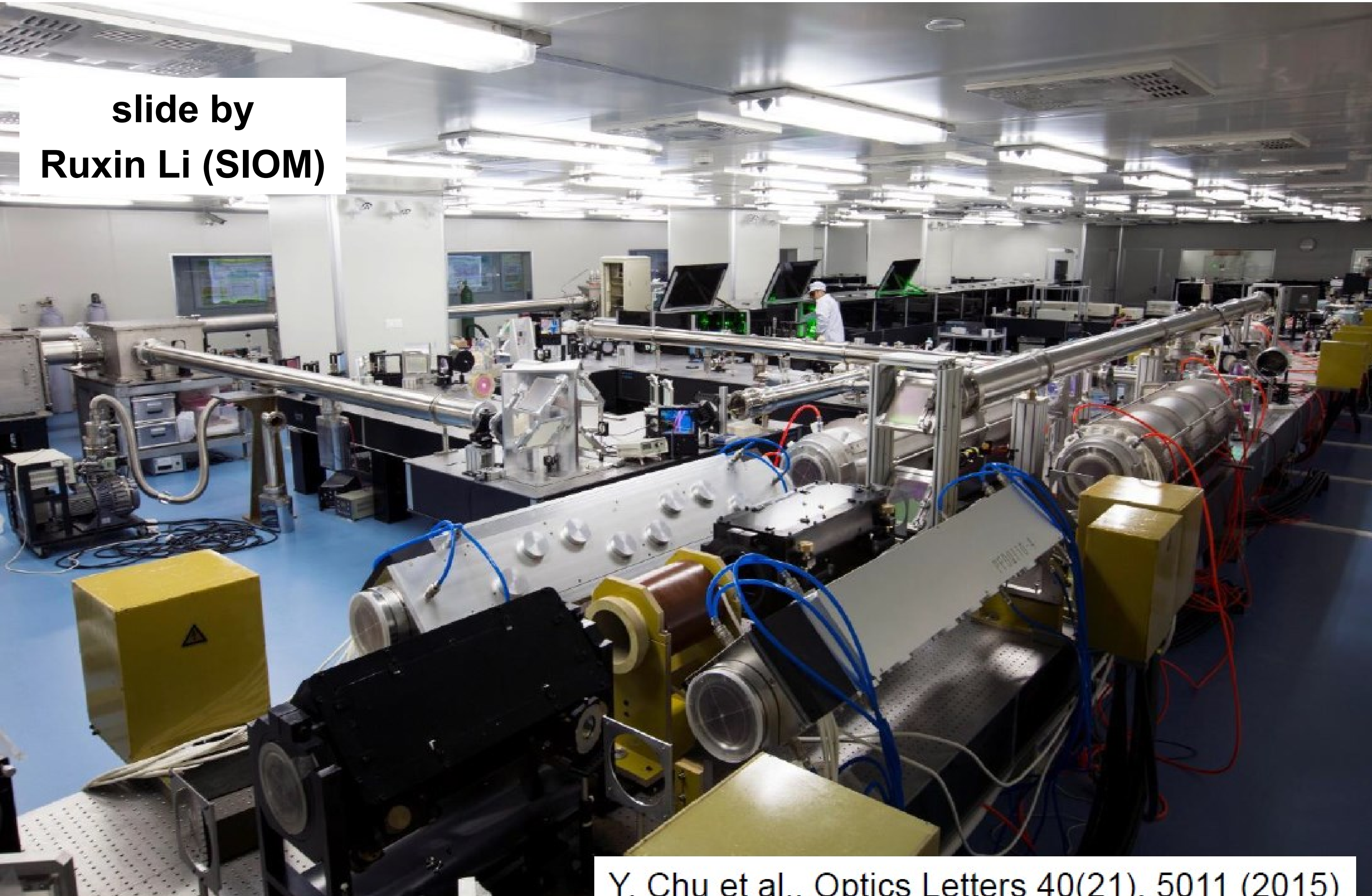
G. Mourou, J. A. Wheeler, and T. Tajima, Europhys. News **46**, 31 (2015)

Optics and Photonics News
Oct. 2017



https://www.youtube.com/watch?v=rDpLT7yTQvA

SULF The 5PW CPA amplifier (2014)



slide by Ruxin Li (SIOM)

Y. Chu et al., Optics Letters 40(21), 5011 (2015)

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

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

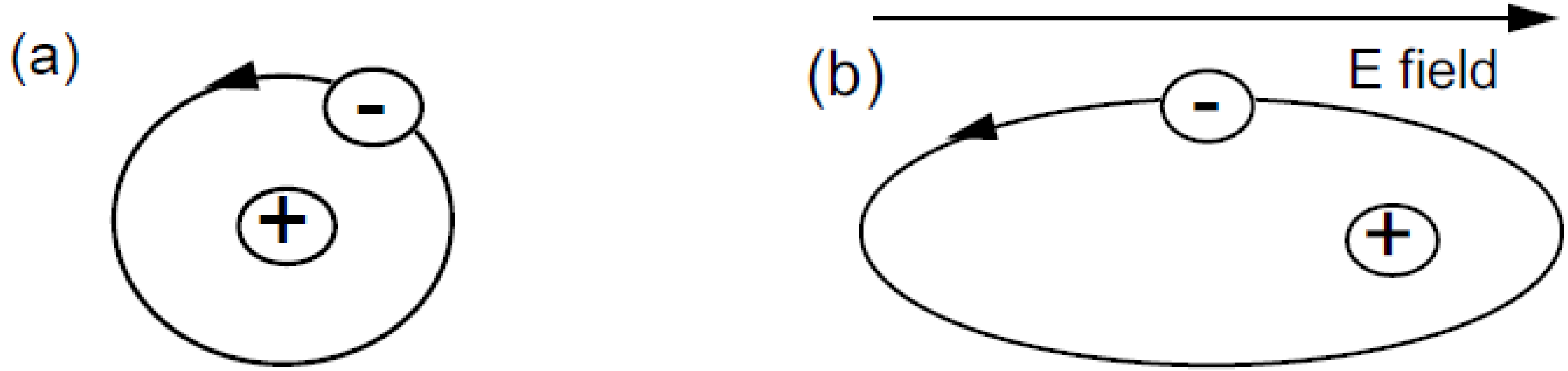


Figure 1.1: **A simple atom model** explaining the effect of in optical electric field on the induced polarization in an atom: (a) without field, (b) with field.

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 $N_A = 6 \cdot 10^{23}\ \text{cm}^{-3}$

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	$\chi^{(2)}$ [illegible] $= 5.$[illegible]	[illegible]	[illegible]
3	$\chi^{(3)}$ [illegible] $= 3.$[illegible]	[illegible]	[illegible]

Table 1.2: Linear and nonlinear optical susceptibilities from a simple atom model. We used $n_0(\text{KDP}) = 2.3$, $d_a = \alpha^{(i)} = 10^{-10}$ 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, $N = 6 \cdot 10^{23} \cdot 10^6 \text{m}^{-3}$.

Estimate for (nonlinear) susceptibilities

refractive index:

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

As table (1.2) shows, the model predicts

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

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

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

or

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

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

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

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 ($\omega_{vib}=\omega_2-\omega_1$)

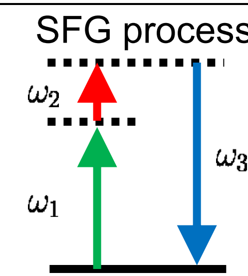
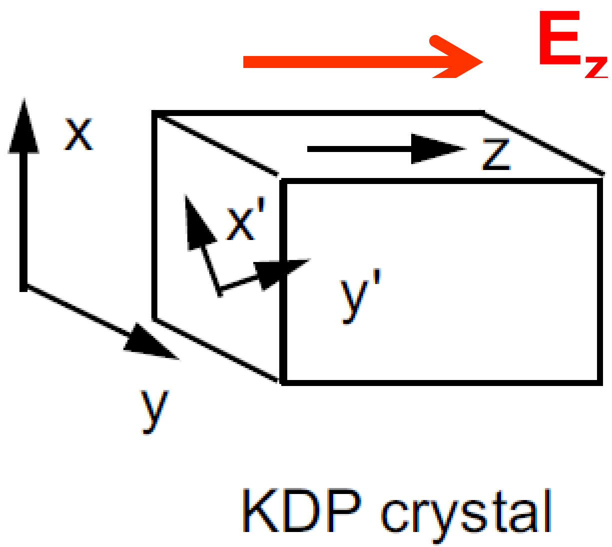


Table 1.3: Important nonlinear optical susceptibilities and corresponding nonlinear optical processes. The first argument in the susceptibility gives the frequency of the generated wave and the other arguments after the semicolon give the frequency components of the input waves.

1.3.1 Linear electro-optical or Pockels effect

KDP: potassium dihydrogen phosphate: KH_2PO_4

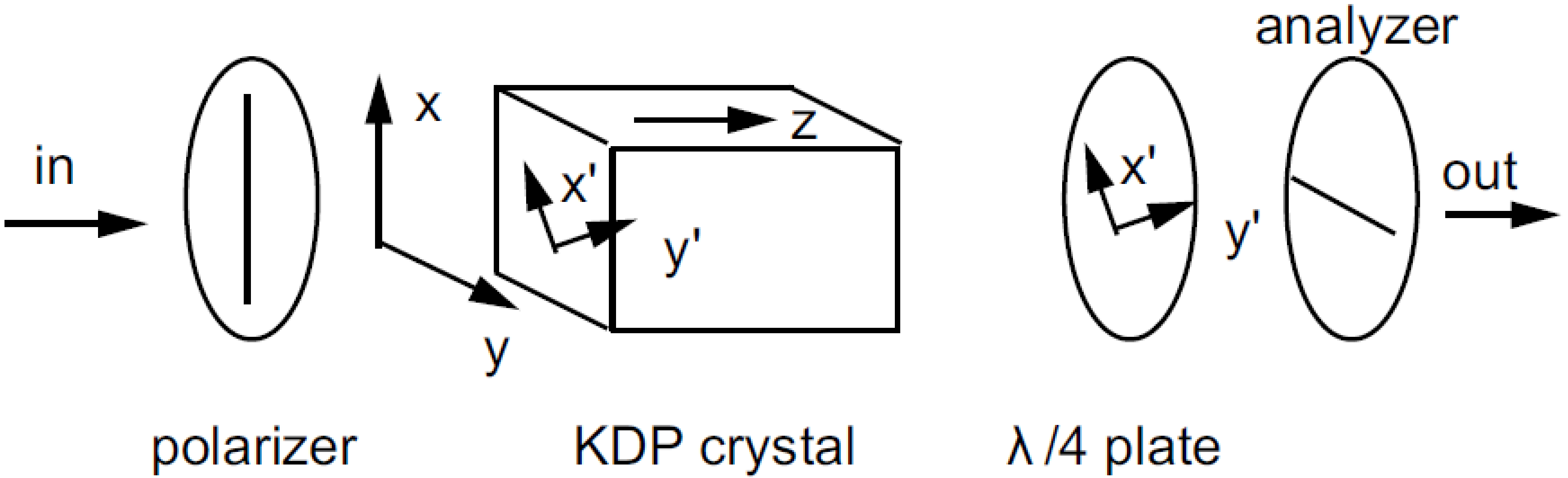


Induced birefringence when electric field is applied in z-direction

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

Electro-optic modulator (EOM)

KDP: potassium dihydrogen phosphate:



Induced birefringence when electric field is applied in z-direction

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

Electro-optic modulator (EOM)

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

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

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

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

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

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

Modulator transmission

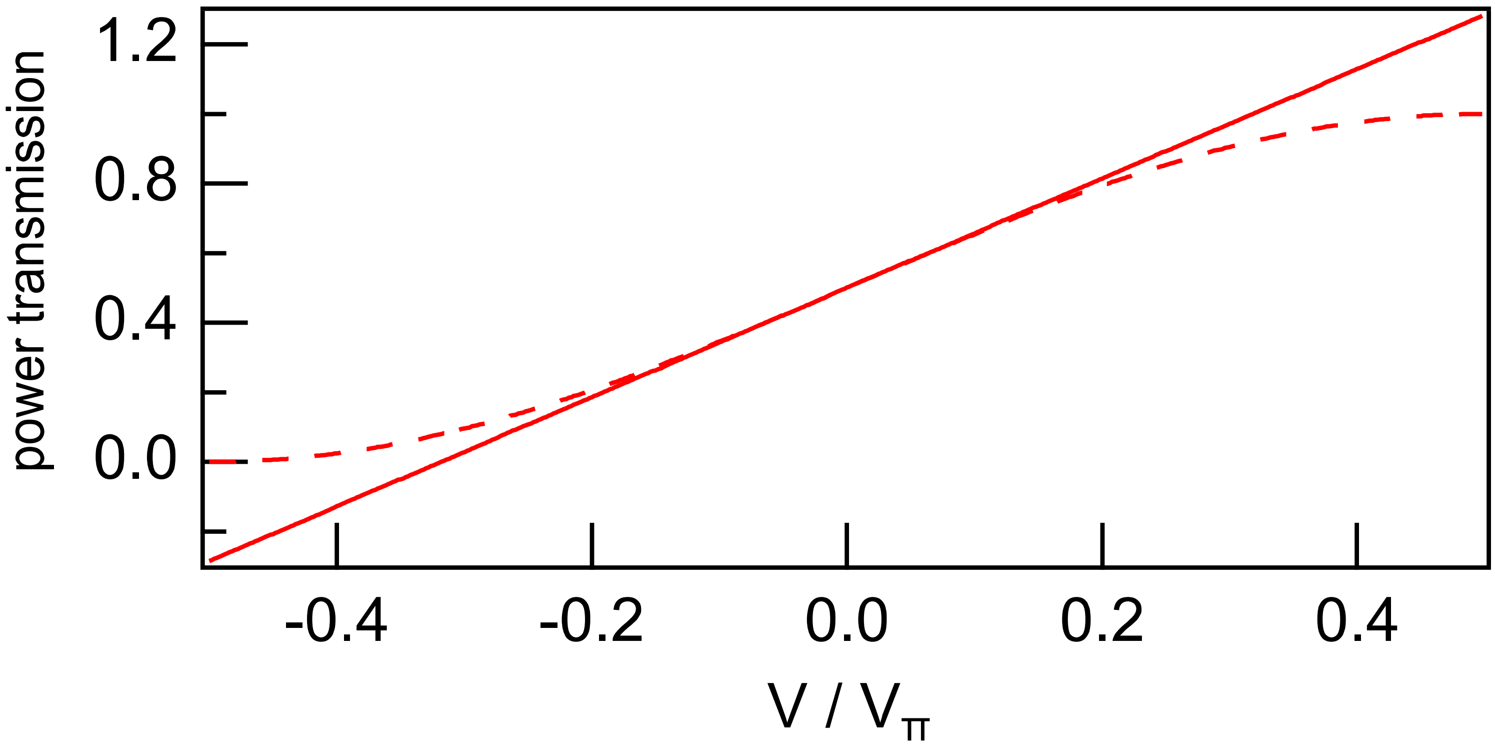


Figure 1.3: Transmission through an electro-optic modulator

Estimate for nonlinear susceptibilities

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

Nonlinear susceptibilities

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

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

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, $N = 6 \cdot 10^{23} \cdot 10^6 \text{m}^{-3}$.

1.3.2 Self-phase modulation

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

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

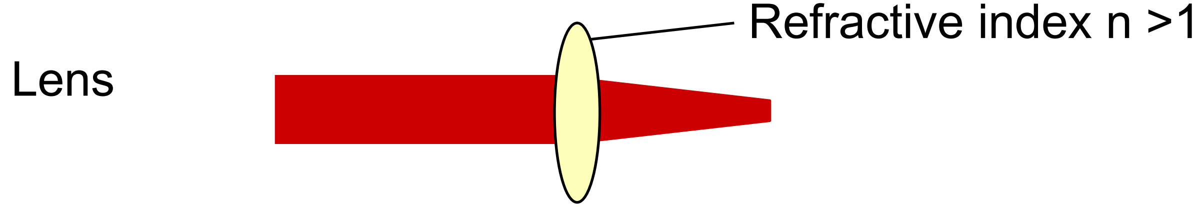
Z_0 is the impedance of the wave in the medium

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

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

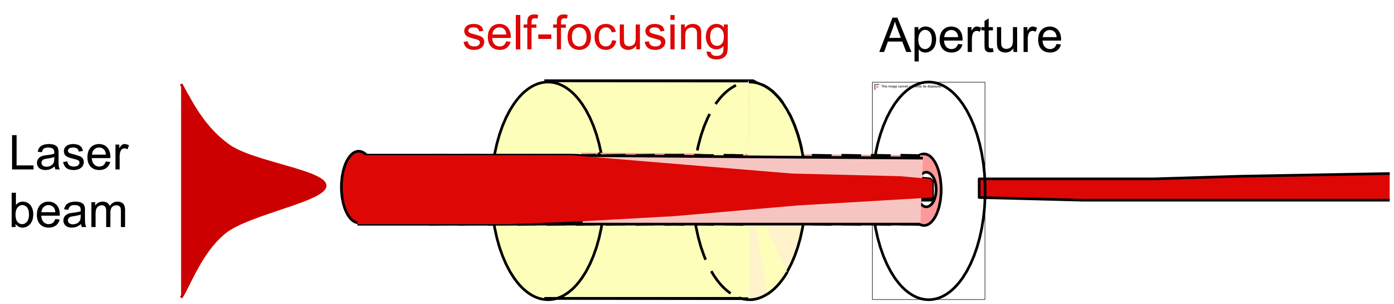
order i	$\chi^{(i)}$	model value	typ. material value
1	$\chi^{(1)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a} \approx 7.5$	n=2.9	Quartz: n=1.45
2	$\chi^{(2)} = \frac{Ne\alpha^{(1)}}{\epsilon_0 E_a^2} = 5.4 \cdot 10^{-11} \frac{\text{m}}{\text{V}}$	$V_\pi = \frac{\lambda n_0}{\chi^{(2)}} = 30$ kV	KDP: $V_\pi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

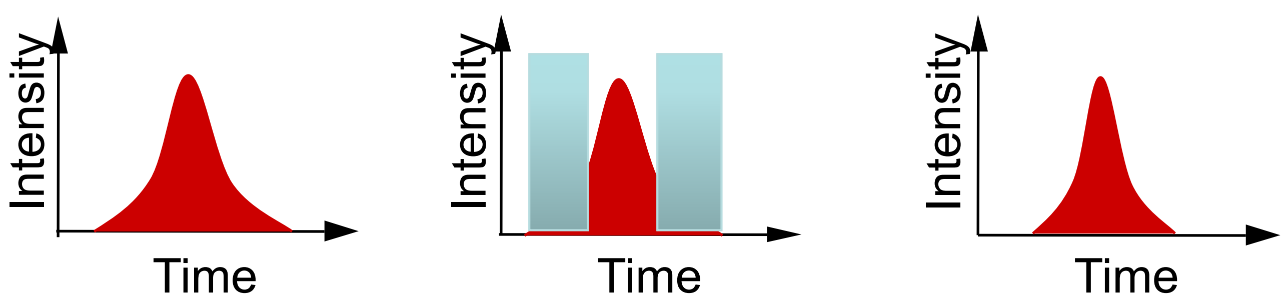
1.3.3 Self-focusing



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

Kerr-Lens Mode locking



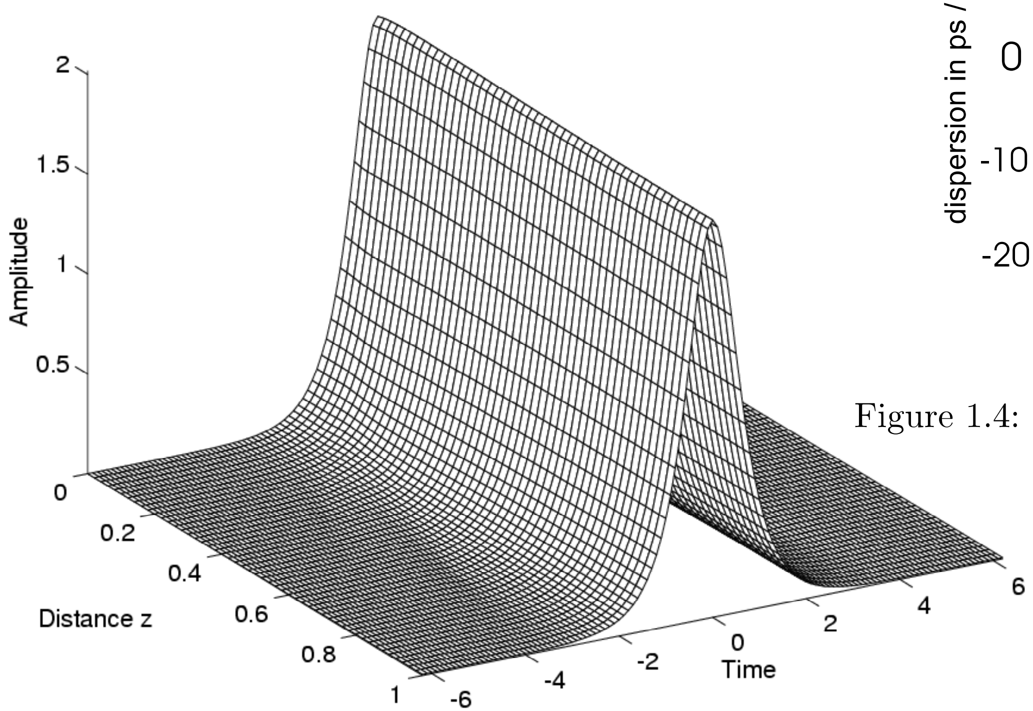


1.3.4 Optical solitons

Nonlinear Schrödinger Equation (NLSE)

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

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



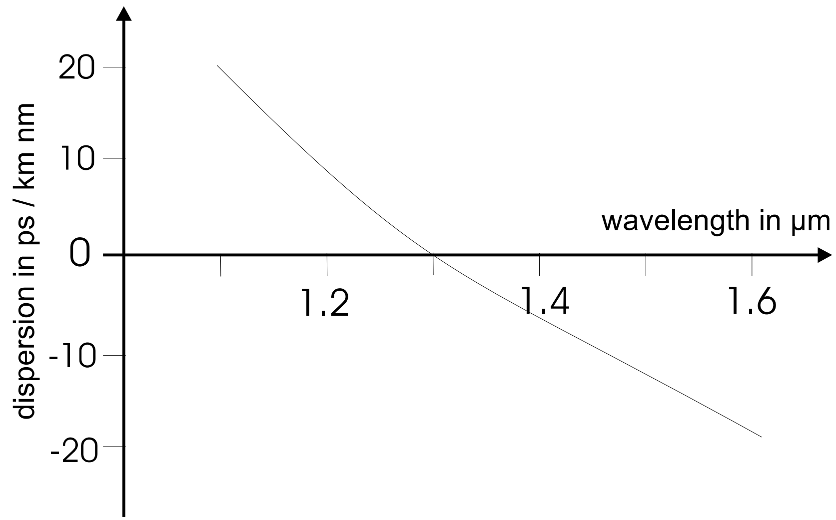


Figure 1.4: Disperson of standard optical fiber in practical units.

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

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

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

($\longrightarrow$ Differenz Frequency Generation, DFG)

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

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

($\longrightarrow$ 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

$$\chi_{ijk}^{(n)}(\omega : \omega_1, \omega_2, ...) = \chi_{ikj}^{(n)}(\omega : \omega_2, \omega_1, ...). \quad (2.12)$$

2.2 Classical model for nonlinear optical susceptibility

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

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

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

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

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

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

$$x_0(t) = \frac{1}{2}\left(\hat{x}_0(\omega)e^{j\omega t} + c.c.\right)$$

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

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

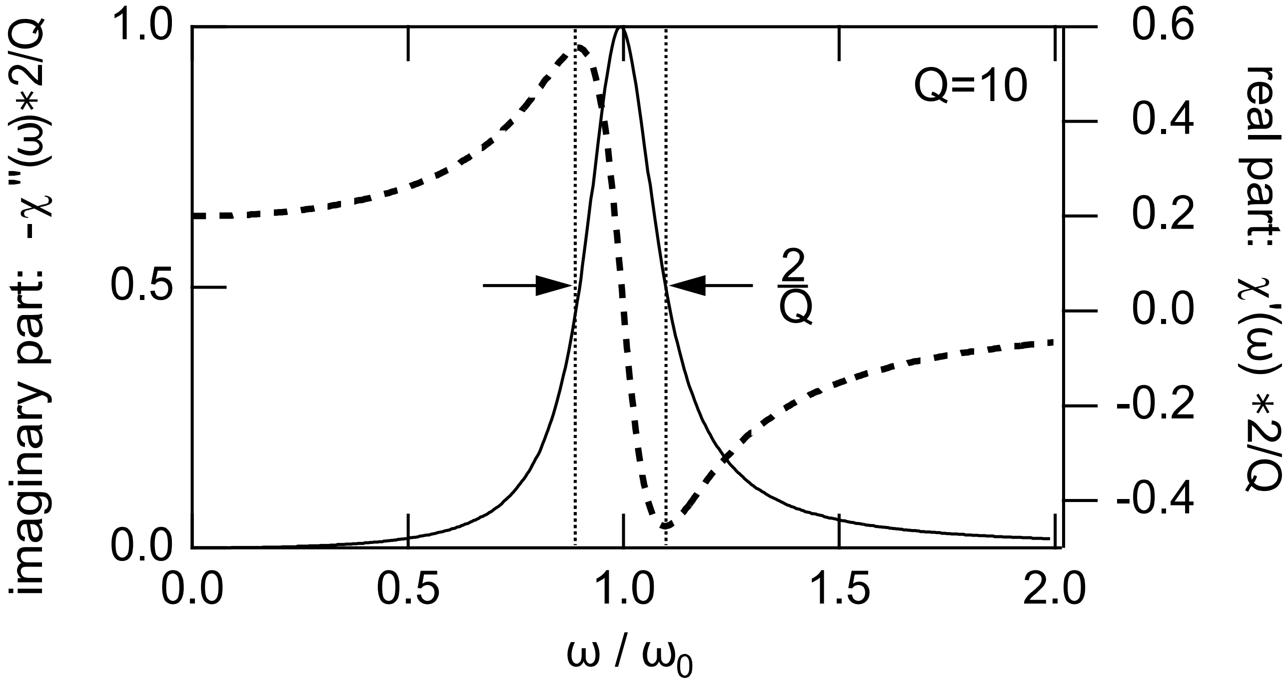


Figure 2.1: Susceptibility arising from the linear harmonic oscillator model for the electron cloud surrounding an atomic core.

Real and imaginary part of the susceptibility

$$\chi^{(1)}(\omega) = \frac{\omega_P^2}{\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)} \tag{2.26}$$

$$= \frac{\omega_P^2}{2j\omega_0'}\left[\frac{1}{\left(\frac{1}{Q} + j\left(\omega - \omega_0'\right)\right)} - \frac{1}{\left(\frac{1}{Q} + j\left(\omega + \omega_0'\right)\right)}\right] \tag{2.27}$$

$$\approx \frac{\omega_P^2}{2j\omega_0}\left[\frac{1}{\left(\frac{1}{Q} + j\left(\omega - \omega_0\right)\right)} - \frac{1}{\left(\frac{1}{Q} + j\left(\omega + \omega_0\right)\right)}\right] \tag{2.28}$$

$$\approx \frac{\omega_P^2}{2j\omega_0}\frac{1}{\left(\frac{1}{Q} + j\left(\omega - \omega_0\right)\right)}, \text{für } \omega \quad \text{um } +\omega_0. \tag{2.29}$$

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

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

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

$$\hat{x}_1(0) = -\beta_2 \frac{1}{\omega_0^2}\left|\left(\frac{-e_0}{m\left(\omega_0^2 - \omega^2 + j\frac{2}{Q}\omega_0\omega\right)}\right)\right|^2 \left|\hat{E}(\omega)\right|^2 \tag{2.30}$$

$$= -\beta_2 \frac{\chi^{(1)}(0)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-2} \left|\chi^{(1)}(\omega)\right|^2 \left|\hat{E}(\omega)\right|^2, \tag{2.31}$$

$$\hat{x}_1(2\omega) = \frac{-\beta_2}{2}\frac{\chi^{(1)}(2\omega)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-2} \chi^{(1)}(\omega)^2 \hat{E}(\omega)^2, \tag{2.32}$$

$$\hat{x}_1(\omega) = \frac{-3\beta_3}{4}\frac{\chi^{(1)}(\omega)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-3} \left|\chi^{(1)}(\omega)\right|^2 \left(\chi^{(1)}(\omega)\right) \tag{2.33}$$

$$\times \left|\hat{E}(\omega)\right|^2 \hat{E}(\omega), \tag{2.34}$$

$$\hat{x}_1(3\omega) = \frac{-\beta_3}{4}\frac{\chi^{(1)}(3\omega)}{\omega_P^2}\left(-\frac{Ne_0}{\varepsilon_0}\right)^{-3} \chi^{(1)}(\omega)^3 \hat{E}(\omega)^3. \tag{2.35}$$

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

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

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

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