



Nonlinear Optics

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Lecture 1

Our research: Photonic Integration – Technologies and Applications







DSPOC: Digital Signal Processing in Optical Communications (Workshop & Lecture by Prof. Sebastian Randel)

This course will provide practical knowledge about the design and implementation of digital-signalprocessing (DSP) algorithms in optical communication systems. Topics will be introduced in lectures and applied in practical exercises in the computer lab using MATLAB. The course will cover the following topics:

- Modulation formats, entropy, spectral efficiency, pulse shaping
- Noise sources and statistics
- Performance evaluation: Bit-error ratio, Q-Factor, OSNR-Penalty, Mutual Information, Monte Carlo Simulation
- Modeling systems with intensity modulation and direct detection
- Digital coherent receivers (carrier recovery, timing recovery, chromatic dispersion compensation, adaptive equalization)
- Impact and compensation of chromatic dispersion and polarization-mode dispersion
- System impact of fiber nonlinearities

 Lecture (1 SWS):
 Tuesday
 11:30-13:00

 Workshop (2 SWS):
 Thursday
 15:45-17:15

Building 20.40, Neuer Hörsaal (NH) Building 20.21, SCC-PC-Pool L



27.06.2018

More information: www.ipq.kit.edu/Lectures.php



The need for high-speed optical communications







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Our Research: Teratronics and Photonics





Capacity challenges in communication networks





Source: Urs Hölzle (Google employee No. 8, today: VP Technical Infrastructure), OFC 2017



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Connection of Data Centers through Campus-Area Networks



U. Hölzle, OFC 2017 Plenary Talk



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Connection of Data Centers through Campus-Area Networks



U. Hölzle, OFC 2017 Plenary Talk



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Connection of Data Centers through Campus-Area Networks



- \Rightarrow Further scaling requires increase of data rate per fiber
- \Rightarrow Massively parallel wavelength division multiplexing (WDM)

U. Hölzle, OFC 2017 Plenary Talk



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Karlsruhe Institute of Tech

Massively parallel wavelength-division multiplexing (WDM) using optical frequency combs





Massively parallel wavelength-division multiplexing (WDM) using optical frequency combs





Nonlinear Optics



Contents

- Linear and nonlinear optics: Maxwell's equations and nonlinear optics, nonlinear wave equation, survey of nonlinear optical processes...
- The nonlinear optical susceptibility: Definition of the nonlinear optical susceptibility tensor, influence of spatial symmetry
- Second-order nonlinear effects: Linear electro-optic effect (Pockels effect), electrooptic modulators, difference-frequency generation and parametric amplification, phase matching
- Acousto-optics: Acousto-optic modulators, interaction of photons and phonons, Brillouin and Raman scattering
- Third-order nonlinear effects: Signal propagation in third-order nonlinear media, the nonlinear Schrödinger equation (NLSE); nonlinear signal processing, optical solitons and modulation instability

Further reading:

- R. W. Boyd. *Nonlinear Optics*. Academic Press, San Diego, 2003.
- G. P. Agrawal. *Nonlinear Fiber Optics*. Academic Press, 2013.
- G. I. Stegeman and R. A. Stegeman. *Nonlinear Optics: Phenomena, Materials, and Devices*, Wiley, 2012
- B. E. A. Saleh and M. C. Teich. *Fundamentals of Photonics*. John Wiley and Sons, 2007.
- Y. R. Shen. *Nonlinear Optics*. John Wiley and Sons, New York, 1984.



14 27.06.2018 Christian Koos

Wiley & Sons, Hoboken, NJ. (2012).

modification of optical properties by the presence of light"

Introduction to nonlinear optics

What is nonlinear optics?

Typical nonlinear-optical phenomena:

- Generation of new frequency components ٠ generation, e.g., third-harmonic generation (THG) or second-harmonic generation (SHG)
- Power-dependent transmission, e.g., ٠ nonlinear absorption or absorption bleaching
- Intensity-dependent interference ٠
- Intensity-dependent beam profiles, e.g., due ٠ to self-focussing









Boyd, "Nonlinear Optics", Academic Press 2003



Generating high optical power: Pulsed laser sources



Important for observation of nonlinear optical phenomena: High intensities

- · Exploit pulsed lasers with high peak power
- Use strongly focused light or optical waveguides with small cross sections

Nd: Glass Petawatt Laser, Lawerence Livermore Nat. Lab

- Peak power: ~ 2 PW
- Pulse stretching and recompression (factor 25000) to avoid damage of laser optics





Pulsed laser sources: Solid-state devices



Important for observation of nonlinear optical phenomena: High intensities

- Exploit pulsed lasers with high peak power
- Use strongly focused light or optical waveguides with small cross sections





Picosecond laser (Lumentum)

- Pulse duration: 10 ps
- Wavelength: 1064 nm
- Repetition frequency: up to 8 MHz
- Average Power: up to 50 W
- Peak power: up to 20 MW

Ti: Sapphire laser oscillator (Spectra Physics)

- Pulse duration: < 100 fs
- Wavelength: ~ 800 nm
- Repetition frequency: 80 MHz
- Average Power: > 1.1 W
- Peak power: > 170 kW

[1] https://www.lumentum.com/en/products/laser-ultrafast-industrial-picoblade

[2] http://www.spectra-physics.com/products/ultrafast-lasers-for-scientific-research/



Pulsed laser sources: Solid-state devices



Important for observation of nonlinear optical phenomena: High intensities

- Use strongly focused light or optical waveguides with small cross sections
- Exploit pulsed lasers with high peak power



Ti: Sapphire Laser (Spectra Physics)

- Pulse duration: < 100 fs
- Wavelength: ~ 800 nm
- Repetition frequency: 80 MHz
- Average power: > 4 W
- Peak power > 500 kW



Ergo-XG (Time-Bandwidth Products; now: Lumentum)

- Pulse duration: < 2 ps
- Wavelength: ~ 1550 nm
- Repetition frequency: 10 GHz
- Average power: > 50 mW
- Peak power > 2.5 W



Pulsed fiber lasers and mode-locked laser diodes





Mode-locked laser diode (III-V-labs)

- Pulse duration: ~ 1 ps
- Wavelength: 1550 nm
- Repetition frequency: up to 500 GHz
- Average Power: ~ 20 mW
- Peak power ~ 200 mW

Fiber laser (Keopsys)

- Pulse duration: 0.5 ns to 200 ns
- Wavelength: ~ 1550 nm
- Repetition frequency: 10 Hz to 1 MHz
- Average power: up to 1.2 W
- Peak power: up to 15 kW



[1] http://www.bpress.cn/im/tag/Keopsys/



Pulsed fiber lasers and mode-locked laser diodes





Fiber laser (Calmar)

- Pulse duration: 0.8 5 ps
- Wavelength: ~ 1550 nm
- Repetition frequency: 40 GHz
- Average Power: > 20 mW
- Peak power < 1 W

Mode-locked laser diode (III-V-labs)

- Pulse duration: ~ 1 ps
- Wavelength: 1550 nm
- Repetition frequency: up to 500 GHz
- Average Power: ~ 20 mW
- Peak power ~ 200 mW





Applications of nonlinear optics



Special features of nonlinear-optical processes:

- Ultra-short response times (fs!) => Ultra-fast signal processing!
- Broadband => Light generation / amplification at wavelength ranges that cannot be accessed by other gain media

Example: Green laser pointer, based on second-harmonic generation of 1064 nm light



-> Second-harmonic generation (SHG) in nonlinear KTP crystal: 532 nm



Applications of nonlinear optics: Supercontinuum generation in highly nonlinear fibers



High index-contrast "small-core" fiber: High optical intensities in the waveguide core



Leong et al. Journ. Lightw. Technol., Vol. 24, No. 1 (2006)

- Solid core + low-index air cladding
- Tight confinement of light within a small core area
- ⇒ Strong nonlinear effects; allows for supercontinuum generation!



http://www.bath.ac.uk/physics/groups/cppm/nonlinear_pcf.php



Applications of nonlinear optics:

Frequency comb generation for high-speed data transmission



Mode-locked laser + spectral broadening in highly nonlinear fiber (HNLF) Problem: Spectral dips for SPM-induced phase shifts of $\Delta \Phi \ge 1.5 \pi$



 \Rightarrow 325 carriers with 12.5 GHz spacing, linewidth < 10 kHz

Hillerkuss *et al.*, J. Opt. Commun. Netw. 4, 715–723 (2012)

Applications of nonlinear optics: Kerr frequency comb generation for high-speed data transmission

Recent Achievements: Temporal dissipative integrated soliton frequency comb

- 179 Carriers from chip-scale comb sources,
- 40 GBd, 16 QAM, PolMUX \Rightarrow **50.2 Tbit/s**



Chip-scale comb source

Si₃N₄

input

and Quantum Electronics

cw pump

laser

Karlsruhe Institute of Techno

100µm

output

Applications of nonlinear optics: Kerr frequency comb generation for ultrafast optical ranging



- Unique combination of large free spectral range and large bandwidth enables ultrafast and precise optical distance measurements
- Demonstration of optical ranging at 100 MHz sampling rate, while keeping nm-precision in mm-range



Trocha, P. et al., Science 359, 887-891 (2018)







Ultra-fast all-optical switching: Pump-probe measurement of response times





Applications of nonlinear optics: All-optical switching in nanophotonic silicon waveguides





SOI strip waveguides:

- Nonlinear response impaired by free carriers
- Free-carrier lifetime: 1.2 ± 0.1 ns

 $= \pi \begin{bmatrix} -16.0 \text{ dBm} \\ -16.0 \text{ dBm} \\ 0 \\ -\pi \end{bmatrix} \begin{bmatrix} -16.0 \text{ dBm} \\ 0 \\ 0.7 \text{ dBm on chip} \\ -2 \\ 0 \\ 2 \\ t \text{ [ps] 4} \end{bmatrix} \end{bmatrix} 4$

π

Silicon-organic hybrid (SOH) slot waveguides:

- No impairment by free carriers
- Suitable for ultra-fast all-optical signal processing

Koos *et al.*, Nature Photonics **3**, 216-218 (2009) Vallaitis *et al.*, Opt. Expr. **17**, 17357–17368 (2009)



Applications of nonlinear optics: Lithium Niobate (LiNbO₃) electro-optic modulator





z-cut Lithium Niobate (LiNbO₃)

- Push-pull operation with RF-signals of opposite polarity
- Good overlap of RF-field and optical field → Low voltage; good electrooptic efficiency



E. L. Wooten et al., IEEE Journal of Selected Topics in Quantum Electronics 6 (1), Jan/Feb 2000, pp. 69 ff.



x-cut Lithium Niobate

- Push-pull operation a single RF signal.
- Needs approx. 20% higher voltage compared to device on z-cut substrate



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Applications of nonlinear optics: Silicon-organic hybrid (SOH) electro-optic modulators

Silicon-organic hybrid (SOH) integration: Combine silicon-on-insulator (SOI) waveguides with

functional organic cladding materials



SOH: 252 Gbit/s $U_{
m pp} pprox 1.0 ~
m V$ $W_{\rm bit}$ = 22 fJ / bit

> Conventional: 112 Gbit/s $U_{
> m pp} pprox 5.0 ~
> m V$ $W_{\rm bit}$ > 1000 fJ / bit

Koos et al., J. Lightw. Technol. 24, 256-268 (2016) Koeber et al., Light: Science & Applications 4, e255, doi:10.1038/lsa.2015.28, (2015) Wolf et al., OFC 2017, paper Th5C.1. (postdeadline paper)

Applications of nonlinear optics: Silicon-organic hybrid (SOH) electro-optic modulators



Concept: Combine nanophotonic silicon waveguides with electro-optic organic cladding materials

- High-speed modulation: 3 dB bandwidth > 100 GHz (Allsilicon devices: 30 GHz)
- Highly efficient: U_πL < 1 Vmm (All-silicon devices: U_πL = 10 ... 40 V mm)
- Lowest energy consumption of a Mach-Zehnder modulator (MZM) in any material system:
 - < 2 fJ/bit (All-silicon MZM devices: 200 fJ/bit)
- No amplitude-phase coupling: Enables higherorder modulation formats (16 QAM)





Lauermann *et al.*, Opt. Express **22**, 29927–29936 (2014) Koeber *et al.*, Light: Science & Applications **4**, e255, doi:10.1038/lsa.2015.28, (2015) Lauermann *et al.*, J. Lightw. Technol. **33**, ,1210-1216 (2015) Hartmann *et al.*, ECOC 2015, Post-deadline paper PDP1.4 (2015)

Silicon-Organic Hybrid (SOH) Integration





Applications of nonlinear optics: Direct-write 3D laser lithography







Polymerization by one-photon absorption

Polymerization by two-photon absorption:

- Increased resolution
- Suppression of beam side-lobes



Figures adapted from Nanoscribe GmbH

32 27.06.2018 Christian Koos





Lindenmann et al., Opt. Express 20, 17667-17677 (2012)

Photonic wire bonding and photonic multi-chip integration



Multi-chip integration: Assemble photonic systems from discrete chips that combine strengths of different material platforms



Lindenmann *et al.*, Opt. Express **20**, 17667-17677 (2012) Lindenmann *et al.*, Journal of Lightw. Technology **33**, 755-760 (2015) Billah *et al.*, Cleo 2015, Paper STu2F.2

Additive 3D nanofabrication of photonic devices





Linear and Nonlinear Optics


Basic assumptions: No free charges, no currents
Nonmagnetic material
$$\nabla \cdot \mathbf{D}(\mathbf{r}, t) = 0 \qquad \mathbf{B}(\mathbf{r}, t) = \mu_0 \mathbf{H}(\mathbf{r}, t)$$
$$\nabla \times \mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{B}(\mathbf{r}, t)}{\partial t} \qquad \mathbf{D}(\mathbf{r}, t) = \epsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t)$$
$$\nabla \cdot \mathbf{B}(\mathbf{r}, t) = 0$$
$$\nabla \times \mathbf{H}(\mathbf{r}, t) = \frac{\partial \mathbf{D}(\mathbf{r}, t)}{\partial t}$$

Linear and nonlinear dielectric polarization:

- Linear media: Linear relationship between polarization P and electric field E
- Nonlinear media: Nonlinear relationship between P and E

Nonlinear Optics:

The field of nonlinear optics (often abbreviated as NLO) comprises the branch of optics that describes the behavior of light in nonlinear media, in which the dielectric polarization **P** responds nonlinearly to the electric field **E** of the light.





$$\begin{array}{ll} \textbf{General case:} \quad \mathbf{P}_{\mathsf{L}}(\mathbf{r},t) = \epsilon_{0} \int_{-\infty}^{\infty} \iiint_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r},\mathbf{r}',t,t') \mathbf{E}(\mathbf{r}',t') \, \mathrm{d}\mathbf{r}' \mathrm{d}t' \\ & \exists \mathbf{x} \exists \text{-matrix} \end{array} \\ \begin{array}{ll} \textbf{Time-invariant} \\ \textbf{media} \end{array} \quad \mathbf{P}_{\mathsf{L}}(\mathbf{r},t) = \epsilon_{0} \int_{-\infty}^{\infty} \iiint_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r},\mathbf{r}',\tau) \mathbf{E}(\mathbf{r}',t-\tau) \, \mathrm{d}\mathbf{r}' \mathrm{d}\tau \\ \\ \textbf{Media that are} \\ (additionally) \\ \textbf{local in space} \end{array} \quad \mathbf{P}_{\mathsf{L}}(\mathbf{r},t) = \epsilon_{0} \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r},\tau) \mathbf{E}(\mathbf{r},t-\tau) \, \mathrm{d}\tau \\ \\ \textbf{Isotropic media:} \qquad \mathbf{P}_{\mathsf{L}}(\mathbf{r},t) = \epsilon_{0} \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r},\tau) \mathbf{E}(\mathbf{r},t-\tau) \, \mathrm{d}\tau \\ \\ \textbf{Homogeneous} \\ \textbf{media:} \end{aligned} \quad \mathbf{P}_{\mathsf{L}}(\mathbf{r},t) = \epsilon_{0} \int_{-\infty}^{\infty} \chi^{(1)}(\tau) \mathbf{E}(\mathbf{r},t-\tau) \, \mathrm{d}\tau \\ \end{array}$$

Note: In these relations, χ represents the time-domain (susceptibility) influence function of the medium, and the units of χ depend on the relationship that is to be used. For time-invariant media that are local in space, the Fourier transform of $\chi(\mathbf{r},\tau)$ with respect to τ leads to the frequency-dependent electric susceptibility.



Lecture 2



Basic assumptions: No free charges, no currents
Nonmagnetic material
$$\nabla \cdot \mathbf{D}(\mathbf{r}, t) = 0 \qquad \mathbf{B}(\mathbf{r}, t) = \mu_0 \mathbf{H}(\mathbf{r}, t)$$
$$\nabla \times \mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{B}(\mathbf{r}, t)}{\partial t} \qquad \mathbf{D}(\mathbf{r}, t) = \epsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t)$$
$$\nabla \cdot \mathbf{B}(\mathbf{r}, t) = 0$$
$$\nabla \times \mathbf{H}(\mathbf{r}, t) = \frac{\partial \mathbf{D}(\mathbf{r}, t)}{\partial t}$$

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Frequency-domain representation of Maxwell's equations

Fourier transformation:

$$\Psi(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \widetilde{\Psi}(\omega) e^{j\omega t} d\omega \qquad \bullet \qquad \tilde{\Psi}(\omega) = \int_{-\infty}^{+\infty} \Psi(t) e^{-j\omega t} dt$$

Maxwell's equations for a linear time-invariant medium that is local in space:

$$\nabla \cdot \widetilde{\mathbf{D}}(\mathbf{r},\omega) = 0 \qquad \qquad \widetilde{\mathbf{B}}(\mathbf{r},\omega) = \mu_0 \widetilde{\mathbf{H}}(\mathbf{r},\omega), \\ \nabla \times \widetilde{\mathbf{E}}(\mathbf{r},\omega) = -j\omega \widetilde{\mathbf{B}}(\mathbf{r},\omega) \qquad \qquad \widetilde{\mathbf{D}}(\mathbf{r},\omega) = \epsilon_0 \widetilde{\mathbf{E}}(\mathbf{r},\omega) + \widetilde{\mathbf{P}}(\mathbf{r},\omega) \\ \nabla \cdot \widetilde{\mathbf{B}}(\mathbf{r},\omega) = 0 \\ \nabla \times \widetilde{\mathbf{H}}(\mathbf{r},\omega) = j\omega \widetilde{\mathbf{D}}(\mathbf{r},\omega), \qquad \qquad \widetilde{\mathbf{P}}_{\mathsf{L}}(\mathbf{r},\omega) = \epsilon_0 \widetilde{\chi}^{(1)}(\mathbf{r},\omega) \widetilde{\mathbf{E}}(\mathbf{r},\omega)$$

Note:

 $\tilde{\chi}^{(1)}(\mathbf{r},\omega)$ represents the Fourier transform of the time-domain influence function $\chi^{(1)}(\mathbf{r},\tau)$ and is a complex quantity. It is also referred to as the electric susceptibility and often denoted as $\underline{\chi}^{(1)}(\mathbf{r},\omega)$ without the tilde.



Maxwell's equations for complex time-domain amplitudes

Alternatively: Representation of harmonically oscillating field quantities by complex amplitudes of analytic time-domain signals rather than by Fourier transforms: $E(\mathbf{r}, t) = \text{Re} \{ \underline{\mathbf{E}}(\mathbf{r}, \omega_0) \exp(j\omega_0 t) \}$

Complex time-domain amplitude (a number, not a function of ω_0 !)

In linear optics, Maxwell's equations for complex time-domain amplitude are identical to those for Fourier transforms:

$$\nabla \cdot \underline{\mathbf{D}}(\mathbf{r}, \omega_{0}) = 0,$$

$$\nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega_{0}) = -\mathbf{j}\,\omega_{0}\underline{\mathbf{B}}(\mathbf{r}, \omega_{0}),$$

$$\nabla \cdot \underline{\mathbf{B}}(\mathbf{r}, \omega_{0}) = 0,$$

$$\nabla \times \underline{\mathbf{H}}(\mathbf{r}, \omega_{0}) = \mathbf{j}\,\omega_{0}\underline{\mathbf{D}}(\mathbf{r}, \omega_{0}).$$

$$\underline{\mathbf{B}}(\mathbf{r}, \omega_{0}) = \mu_{0}\underline{\mathbf{H}}(\mathbf{r}, \omega_{0}),$$

$$\underline{\mathbf{D}}(\mathbf{r}, \omega_{0}) = \epsilon_{0}\underline{\mathbf{E}}(\mathbf{r}, \omega_{0}) + \underline{\mathbf{P}}(\mathbf{r}, \omega_{0})$$

$$\underline{\mathbf{P}}_{\underline{\mathbf{L}}}(\mathbf{r}, \omega_{0}) = \epsilon_{0}\underline{\chi}^{(1)}(\mathbf{r}, \omega_{0}) \underline{\mathbf{E}}(\mathbf{r}, \omega_{0})$$
where $\underline{\chi}^{(1)}(\mathbf{r}, \omega_{0}) = \widetilde{\chi}^{(1)}(\mathbf{r}, \omega_{0})$

Note:

- In nonlinear optics, it is important to discriminate between complex timedomain amplitudes of monochromatic waves, and Fourier transforms.
- Nonlinear optics mostly uses timedomain products of complex field amplitudes rather than Fourier transforms to avoid convolutions of signal spectra in the frequency domain.

Fourier transform of time-domain influence function χ (t).



Constitutive relations:

$$\underline{\mathbf{D}}(\mathbf{r},\omega) = \epsilon_0 \underline{\mathbf{E}}(\mathbf{r},\omega) + \underline{\mathbf{P}}(\mathbf{r},\omega)$$

= $\epsilon_0 \left(1 + \underline{\chi}^{(1)}(\mathbf{r},\omega)\right) \underline{\mathbf{E}}(\mathbf{r},\omega)$
= $\epsilon_0 \underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega) \underline{\mathbf{E}}(\mathbf{r},\omega)$
= $\epsilon_0 \underline{n}^2(\mathbf{r},\omega) \underline{\mathbf{E}}(\mathbf{r},\omega)$

Complex dielectric constant and refractive index :

$$\underline{\epsilon}_{\mathsf{f}}(\mathbf{r},\omega) = 1 + \underline{\chi}(\mathbf{r},\omega) = \underline{n}^2(\mathbf{r},\omega)$$

Convention: Positive values of $n_i (\epsilon_{ri})$ are assigned to lossy media, negative values correspond to optical gain!

$$\underline{n} = n \text{of } n_i,$$

$$\epsilon_r = n^2 - n_i^2,$$

$$n^2 = \frac{1}{2} \epsilon_r \left(1 + \sqrt{1 + \epsilon_{ri}^2 / \epsilon_r^2} \right),$$

$$n \approx \sqrt{\epsilon_r} \qquad \text{(for } |\epsilon_{ri}| \ll \epsilon_r),$$

$$n \approx \sqrt{|\epsilon_{ri}|/2} \qquad \text{(for } |\epsilon_{ri}| \gg \epsilon_r)$$

$$\underline{\epsilon}_{r} = \epsilon_{r} \textcircled{o}_{j} \epsilon_{ri},$$

$$\epsilon_{ri} = 2nn_{i},$$

$$n_{i} = \epsilon_{ri}/(2n),$$

$$n_{i} \approx \epsilon_{ri}/(2\sqrt{\epsilon_{r}}),$$

$$n_{i} \approx \operatorname{sgn}(\epsilon_{ri})\sqrt{|\epsilon_{ri}|/2}.$$







Linearität 1. $\nabla(\alpha \Phi + \beta \Psi) = \alpha \nabla \Phi + \beta \nabla \Psi$ 2. $\nabla \cdot (\alpha F + \beta G) = \alpha \nabla \cdot F + \beta \nabla \cdot G$ 3. $\nabla \times (\alpha F + \beta G) = \alpha \nabla \times F + \beta \nabla \times G$	grad($\alpha \Phi + \beta \Psi$) = α grad $\Phi + \beta$ grad Ψ div($\alpha F + \beta G$) = α div $F + \beta$ div G rot($\alpha F + \beta G$) = α rot $F + \beta$ rot G
Operation auf Produkten 4. $\nabla(\Phi\Psi) = \Phi \nabla\Psi + \Psi \nabla\Phi$ 5. $\nabla(\mathbf{F} \cdot \mathbf{G}) = (\mathbf{F} \cdot \nabla)\mathbf{G} + (\mathbf{G} \cdot \nabla)\mathbf{F} + \mathbf{F} \times (\nabla \times \mathbf{G}) + \mathbf{G} \times (\nabla \times \mathbf{F})$ 6. $\nabla \cdot (\Phi \mathbf{F}) = \Phi \nabla \cdot \mathbf{F} + (\nabla \Phi) \cdot \mathbf{F}$ 7. $\nabla \cdot (\mathbf{F} \times \mathbf{G}) = \mathbf{G} \cdot \nabla \times \mathbf{F} - \mathbf{F} \cdot \nabla \times \mathbf{G}$ 8. $\nabla \times (\Phi \mathbf{F}) = \Phi \nabla \times \mathbf{F} + (\nabla \Phi) \times \mathbf{F}$ 9. $\nabla \times (\mathbf{F} \times \mathbf{G}) = (\mathbf{G} \cdot \nabla)\mathbf{F} - (\mathbf{F} \cdot \nabla)\mathbf{G} + \mathbf{F} + \mathbf{F}(\nabla \cdot \mathbf{G}) - \mathbf{G}(\nabla \cdot \mathbf{F})$	$grad(\Phi\Psi) = \Phi grad \Psi + \Psi grad \Phi$ $grad(\mathbf{F} \cdot \mathbf{G}) = (\mathbf{F} \cdot grad)\mathbf{G} +$ $+ (\mathbf{G} \cdot grad)\mathbf{F} + \mathbf{F} \times rot \mathbf{G} + \mathbf{G} \times rot \mathbf{F}$ $div(\Phi \mathbf{F}) = \Phi div \mathbf{F} + \mathbf{F} \cdot grad \Phi$ $div(\mathbf{F} \times \mathbf{G}) = \mathbf{G} \cdot rot \mathbf{F} - \mathbf{F} \cdot rot \mathbf{G}$ $rot(\Phi \mathbf{F}) = \Phi rot \mathbf{F} + (grad \Phi) \times \mathbf{F}$ $rot(\mathbf{F} \times \mathbf{G}) = (\mathbf{G} \cdot grad)\mathbf{F} -$ $- (\mathbf{F} \cdot grad)\mathbf{G} + \mathbf{F} div \mathbf{G} - \mathbf{G} div \mathbf{F}$
Zweifache Anwendung von ∇ 10. $\nabla \cdot (\nabla \times \mathbf{F}) = 0$ 11. $\nabla \times (\nabla \Phi) = 0$ 12. $\nabla \times (\nabla \times \mathbf{F}) = \nabla (\nabla \cdot \mathbf{F}) - \nabla^2 \mathbf{F}$	div rot $\mathbf{F} = 0$ rot grad $\mathbf{\Phi} = 0$ rot rot $\mathbf{F} =$ grad div $\mathbf{F} - \Delta \mathbf{F}$

Rade / Westergren, Mathematische Formeln, Springer



Wave equation and plane waves



Wave equation in homogeneous media:

$$\nabla^{2}\underline{\mathbf{E}}(\mathbf{r},\omega) + k_{0}^{2}\underline{\epsilon}_{\mathbf{r}}(\omega)\underline{\mathbf{E}}(\mathbf{r},\omega) = 0$$

$$\nabla^{2}\underline{\mathbf{H}}(\mathbf{r},\omega) + k_{0}^{2}\underline{\epsilon}_{\mathbf{r}}(\omega)\underline{\mathbf{H}}(\mathbf{r},\omega) = 0 \quad \text{where} \quad k_{0} = \frac{\omega}{c}$$
Solution for homogeneous media: Plane waves

$$\mathbf{E}(\mathbf{r},t) = \operatorname{Re}\left\{\underline{\mathbf{E}}(\mathbf{r},\omega) e^{j\omega t}\right\} = \operatorname{Re}\left\{\underline{\mathbf{E}}_{0} e^{j(\omega t - \underline{\mathbf{k}}\mathbf{r})}\right\}$$

$$\mathbf{H}(\mathbf{r},t) = \operatorname{Re}\left\{\underline{\mathbf{H}}(\mathbf{r},\omega) e^{j\omega t}\right\} = \operatorname{Re}\left\{\underline{\mathbf{H}}_{0} e^{j(\omega t - \underline{\mathbf{k}}\mathbf{r})}\right\} \quad \text{where} \quad \underline{\mathbf{k}}^{2} = k_{0}^{2}\underline{\epsilon}_{\mathbf{r}}(\omega)$$

Properties of plane waves:

• **k**, \mathbf{E}_0 , and \mathbf{H}_0 are mutually connected and form an orthogonal right-handed system:

$$\frac{\mathbf{k} \cdot \underline{\mathbf{E}}_0 = \mathbf{0}}{\mathbf{k} \cdot \underline{\mathbf{H}}_0 = \mathbf{0}} \qquad \underline{\mathbf{H}}_0 = \frac{1}{\omega \mu_0} \underline{\mathbf{k}} \times \underline{\mathbf{E}}_0 \qquad \underline{\mathbf{E}}_0 = -\frac{1}{\omega \epsilon_0 \epsilon_r} \underline{\mathbf{k}} \times \underline{\mathbf{H}}_0$$

• The attenuation of a plane wave is linked to the imaginary part n_i of the complex refractive index. For a plane wave propagating in positive z-direction, the power decays as $e^{-\alpha z}$, where the attenuation constant α is given by

$$\alpha = 2k_0 n_i$$

Note: A positive value of n_i corresponds to a positive attenuation coefficient α and therefore to optical loss.





Seneral form:

$$\nabla^{2}\underline{\mathbf{E}}(\mathbf{r},\omega) + \nabla \left(\frac{\nabla \underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega)}{\underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega)} \cdot \underline{\mathbf{E}}(\mathbf{r},\omega) \right) + k_{0}^{2}\underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega)\underline{\mathbf{E}}(\mathbf{r},\omega) = 0$$

$$\nabla^{2}\underline{\mathbf{H}}(\mathbf{r},\omega) + \frac{\nabla \underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega)}{\underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega)} \times (\nabla \times \underline{\mathbf{H}}(\mathbf{r},\omega)) + k_{0}^{2}\underline{\epsilon}_{\mathbf{r}}(\mathbf{r},\omega)\underline{\mathbf{H}}(\mathbf{r},\omega) = 0$$
where $k_{0} = \frac{\omega}{c}$

Homogeneous (and weakly inhomogeneous) media: ϵ can be assumed constant (approximately constant within distances of the order of a wavelength

$$\nabla^{2}\underline{\mathbf{E}}(\mathbf{r},\omega) + k_{0}^{2}\underline{\epsilon}_{\mathsf{r}}(\mathbf{r},\omega)\underline{\mathbf{E}}(\mathbf{r},\omega) = 0$$
$$\nabla^{2}\mathbf{H}(\mathbf{r},\omega) + k_{0}^{2}\underline{\epsilon}_{\mathsf{r}}(\mathbf{r},\omega)\mathbf{H}(\mathbf{r},\omega) = 0$$

Solution for homogeneous media: Plane waves

$$\begin{split} \mathbf{E}(\mathbf{r},t) &= \operatorname{Re}\left\{\underline{\mathbf{E}}(\mathbf{r},\omega)\,\mathrm{e}^{\,\mathrm{j}\,\omega t}\right\} = \operatorname{Re}\left\{\underline{\mathbf{E}}_{0}\,\mathrm{e}^{\,\mathrm{j}(\omega t - \underline{\mathbf{k}}\mathbf{r})}\right\}\\ \mathbf{H}(\mathbf{r},t) &= \operatorname{Re}\left\{\underline{\mathbf{H}}(\mathbf{r},\omega)\,\mathrm{e}^{\,\mathrm{j}\,\omega t}\right\} = \operatorname{Re}\left\{\underline{\mathbf{H}}_{0}\,\mathrm{e}^{\,\mathrm{j}(\omega t - \underline{\mathbf{k}}\mathbf{r})}\right\}\\ \text{where } \underline{\mathbf{k}}^{2} &= k_{0}^{2}\underline{\epsilon}_{\mathbf{r}}(\omega) \end{split}$$





 k, E₀, and H₀ are mutually connected and form an orthogonal righthanded system:

$$\mathbf{\underline{H}}_{0} = \frac{\mathbf{\underline{H}}_{0}}{\omega \mu_{0}} \mathbf{\underline{\underline{H}}}_{0} = \mathbf{\underline{H}}_{0} = \frac{\mathbf{\underline{H}}_{0}}{\omega \mu_{0}} \mathbf{\underline{\underline{H}}}_{0} \times \mathbf{\underline{\underline{H}}}_{0}$$
$$\mathbf{\underline{\underline{H}}}_{0} = -\frac{1}{\omega \epsilon_{0} \epsilon_{r}} \mathbf{\underline{\underline{H}}}_{0} \times \mathbf{\underline{\underline{H}}}_{0}$$

• The attenuation of a plane wave is linked to the imaginary part n_i of the complex refractive index. For a plane wave propagating in positive z-direction, the power decays as $e^{-\alpha z}$, where the attenuation constant α is given by

$$\alpha = 2k_0n_i$$

Note: A positive value of n_i corresponds to a positive attenuation coefficient α and therefore to optical loss.





In nonlinear optics, complex amplitudes of monochromatic electromagnetic waves are usually considered rather than Fourier spectra:

$$\psi(t) = \operatorname{Re}\left\{\underline{\psi}(\omega_0) \exp\left(j \,\omega_0 t\right)\right\}$$

- \Rightarrow If only the real part has a physical meaning, what is the role of the imaginary part?
- **Recall**: For a real time-domain signal $\psi(t) \in \mathbb{R}$, the Fourier spectrum $\widetilde{\psi}(\omega)$ has Hermitian symmetry.

$$\widetilde{\psi}(\omega) = \widetilde{\psi}^{\star}(-\omega).$$

Removing the redundant left part of the spectrum corresponds to adding an imaginary part to the time-domain signal that corresponds to the Hilbert transform of the real part,

Hilbert transform

$$\underline{\psi}(t) = \psi(t) + j\left(\psi(t) \star \frac{1}{\pi t}\right),$$

 $\psi(t)$ is referred to as the analytic representation of a real-valued time-domain function $\psi(t)$.



Recall: The electric susceptibility is the Fourier transform of a causal influence function in time domain.

$$P(t) = \epsilon_0 \int_{-\infty}^{\infty} \chi(\tau) \mathbf{E}(t-\tau) \, \mathrm{d}\tau \quad \bullet \qquad \underline{P}(\omega) = \epsilon_0 \underline{\chi}(\omega) \underline{\mathbf{E}}(\omega)$$

$$\chi(\tau) = 0 \quad \text{for} \quad \tau < 0 \qquad \qquad \widetilde{\chi}(\omega) = \chi(\omega) + \mathrm{j} \chi_i(\omega)$$

As a consequence, the real and the imaginary part of the complex susceptibility are connected by the Hilbert transform,

"Cauchy principal value", i.e., the
$$\chi(\omega) = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi_i(\omega_0)}{\omega_0 - \omega} d\omega_0$$

integration boundaries must approach the singularity
"symmetrically" from both sides. $\chi_i(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi(\omega_0)}{\omega_0 - \omega} d\omega_0$

Making further use of the fact that $\chi(t)$ is real, the Kramers-Kronig relations can be derived,

$$\chi(\omega) = -\frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\omega_0 \chi_i(\omega_0)}{\omega_0^2 - \omega^2} d\omega_0$$
$$\chi_i(\omega) = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\omega \chi(\omega_0)}{\omega_0^2 - \omega^2} d\omega_0$$



Kramers-Kronig relations - discussion



- The refractive index of a medium can be calculated from its absorption spectrum and vice versa. Absorption and dispersion are intimately related by fundamental principles.
- An "ideal" dispersionless lossless medium cannot exist:

$$\chi(\omega) = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi_i(\omega_0)}{\omega_0 - \omega} d\omega_0$$
(1)

$$\chi_i(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi(\omega_0)}{\omega_0 - \omega} \, \mathrm{d}\omega_0 \tag{2}$$

For constant $\chi(f)$, i.e., $\chi(f) = \epsilon_r(f) - 1 = \text{const}_f$, we find $\chi_i(f) = \epsilon_{ri}(f) = 0$ from Eq. (1), which implies $\chi(f) = 0$ and $\epsilon_r(f) = 1$, Eq. (2). Real media always have loss (or gain) in some frequency ranges, and the real part of the susceptibility is always frequency dependent. $\chi(f) = \text{const}_f$ and $\chi_i = 0$ is only possible in certain frequency ranges.



Lorentz oscillator model of bound charges Ε $\left| \vec{e}_{x} \right| \vec{d} = x\vec{e}_{x} \left| \vec{E} \right|$ Equation of motion for bound charges: $m_e \frac{\mathrm{d}^2 x}{\mathrm{d}t^2} = -eE_x - m_e \omega_r^2 x - m_e \gamma_r \frac{\mathrm{d}x}{\mathrm{d}t}$ Complex electric susceptibility: $\underline{\chi}(\omega) = \chi_0 \frac{\omega_r^2}{\omega_r^2 - \omega^2 + j\omega\gamma_r}$ $=\frac{\left(\omega_r^2-\omega^2\right)\omega_r^2}{\left(\omega_r^2-\omega^2\right)^2+\omega^2\gamma_r^2}\chi_0-j\frac{\omega\gamma_r\omega_r^2}{\left(\omega_r^2-\omega^2\right)^2+\omega^2\gamma_r^2}\chi_0, \quad \chi_0=\frac{e^2N}{\epsilon_0m_e\omega_r^2}$ Institute of Photonics



Refractive index and absorption





Real media often have several resonances, each of which contributes to the refractive index and to the absorption:





Complex electric susceptibility far from resonance ($|\omega_r - \omega| >> \gamma_r$):

$$\underline{\chi}(\omega) \approx \frac{\omega_r^2}{\omega_r^2 - \omega^2} \chi_0$$

 $\Rightarrow \chi$ is approximately real, absorption is small. Contributions from multiple resonances lead to so-called Sellmeier equations:

$$n^{2} = 1 + \chi = 1 + \sum_{\nu} \chi_{0\nu} \frac{f_{\nu}^{2}}{f_{\nu}^{2} - f^{2}} = 1 + \sum_{\nu} \chi_{0\nu} \frac{\lambda^{2}}{\lambda^{2} - \lambda_{\nu}^{2}}$$

For Sellmeier coefficients $\chi_{0\nu}$ and λ_{ν} , see reference books on optical materials or material databases, e.g.,

- Palik, E. D. (1998), Handbook of Optical Constants of Solids, Academic Press, San Diego, CA
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Sellmeier coefficients of various materials



Material	Sellmeier Equation	Wavelength
	(Wavelength λ in μ m)	Range (µm)
Fused silica	$n^{2} = 1 + \frac{0.6962\lambda^{2}}{\lambda^{2} - (0.06840)^{2}} + \frac{0.4079\lambda^{2}}{\lambda^{2} - (0.1162)^{2}} + \frac{0.8975\lambda^{2}}{\lambda^{2} - (9.8962)^{2}}$	0.21–3.71
Si	$n^2 = 1 + \frac{10.6684\lambda^2}{\lambda^2 - (0.3015)^2} + \frac{0.0030\lambda^2}{\lambda^2 - (1.1347)^2} + \frac{1.5413\lambda^2}{\lambda^2 - (1104.0)^2}$	1.36–11
GaAs	$n^{2} = 3.5 + \frac{7.4969\lambda^{2}}{\lambda^{2} - (0.4082)^{2}} + \frac{1.9347\lambda^{2}}{\lambda^{2} - (37.17)^{2}}$	1.4–11
BBO	$n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2$	0.22-1.06
	$n_e^2 = 2.3753 + rac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2$	
KDP	$n_o^2 = 1 + \frac{1.2566\lambda^2}{\lambda^2 - (0.09191)^2} + \frac{33.8991\lambda^2}{\lambda^2 - (33.3752)^2}$	0.4–1.06
	$n_e^2 = 1 + \frac{1.1311\lambda^2}{\lambda^2 - (0.09026)^2} + \frac{5.7568\lambda^2}{\lambda^2 - (28.4913)^2}$	
LiNbO ₃	$n_o^2 = 2.3920 + \frac{2.5112\lambda^2}{\lambda^2 - (0.217)^2} + \frac{7.1333\lambda^2}{\lambda^2 - (16.502)^2}$	0.4–3.1
	$n_e^2 = 2.3247 + rac{2.2565\lambda^2}{\lambda^2 - (0.210)^2} + rac{14.503\lambda^2}{\lambda^2 - (25.915)^2}$	

Saleh, B. E. A. & Teich, M. C. (2007), Fundamentals of Photonics, John Wiley & Sons, Hoboken, NJ.



Lecture 3

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and Quantum Electronics

Refractive index and absorption





Real media often have several resonances, each of which contributes to the refractive index and to the absorption:



Example: X-ray lenses





$$\chi(\omega) = \frac{\left(\omega_r^2 - \omega^2\right)\omega_r^2}{\left(\omega_r^2 - \omega^2\right)^2 + \omega^2\gamma_r^2}\chi_0$$
$$\chi_i(\omega) = -\frac{\omega\gamma_r\omega_r^2}{\left(\omega_r^2 - \omega^2\right)^2 + \omega^2\gamma_r^2}\chi_0$$

At very high frequencies ($\omega >> \omega_r$):

n < 1

 \Rightarrow Focussing lenses must have concave form!

n very close to 1 (1 - n \approx 10⁻⁶)

 \Rightarrow Needs lots of lenses to obtain sufficient refraction.



Complex electric susceptibility far from resonance ($|\omega_r - \omega| >> \gamma_r$):

$$\underline{\chi}(\omega) \approx \frac{\omega_r^2}{\omega_r^2 - \omega^2} \chi_0$$

 $\Rightarrow \chi$ is approximately real, absorption is small. Contributions from multiple resonances lead to so-called Sellmeier equations:

$$n^{2} = 1 + \chi = 1 + \sum_{\nu} \chi_{0\nu} \frac{f_{\nu}^{2}}{f_{\nu}^{2} - f^{2}} = 1 + \sum_{\nu} \chi_{0\nu} \frac{\lambda^{2}}{\lambda^{2} - \lambda_{\nu}^{2}}$$

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62 27.06.2018 Christian Koos

Lorentz model (linear optics):

Linear relationship between restoring force and displacement, corresponding to a quadratic potential ("harmonic potential") ⇒ Harmonic oscillator as first-order approximation for small amplitudes.

General case (nonlinear optics):

Higher-order (e.g., cubic) terms in the potential ("anharmonic potential") lead to nonlinear (e.g., quadratic) relationship between restoring force and displacement.

 \Rightarrow Anharmonic oscillator:

$$m_e \frac{\mathrm{d}^2 x}{\mathrm{d}t^2} = -eE_x - m_e \gamma_r \frac{\mathrm{d}x}{\mathrm{d}t} - m_e \omega_r^2 x - \frac{\mathrm{d}x^2}{\mathrm{d}t} - \frac{\mathrm{d}x^2}{\mathrm{d}t^2} - \frac{\mathrm{d}x^2}{\mathrm{$$

For $E_x = \text{Re} \{\underline{E}_x \exp(j \omega_0 t)\}$: Polarization contains new spectral components at $\omega = 0$, $\omega = \omega_0$, $\omega = 2\omega_0$, $\omega = 3\omega_0$...





Nonlinear wave equation: Basic assumptions



• Decompose electric polarization into a strong linear and a weak nonlinear contribution:

 $\mathbf{P}(\mathbf{r},t) = \mathbf{P}_{\mathsf{L}}(\mathbf{r},t) + \mathbf{P}_{\mathsf{NL}}(\mathbf{r},t)$

where $|\mathbf{P}_{\mathsf{NL}}(\mathbf{r},t)| \ll |\mathbf{P}_{\mathsf{L}}(\mathbf{r},t)|$.

- ⇒ The nonlinear polarization can be treated as a small perturbation of the linear polarization, leading to plane-wave-like solutions with weakly space and timedependent amplitudes ("slowly varying envelope approximation", SVEA)
- Assume that the medium is operated far away from any electronic resonances such that the electric susceptibility is real and independent of frequency,

$$\underline{\chi}^{(1)}\left(\mathbf{r},\omega_{l}
ight)pprox\chi^{(1)}\left(\mathbf{r}
ight)\in\mathbb{R}$$

We can then relate the instantaneous linear polarization directly to the instantaneous electric field in the time domain, neglecting any memory of the medium,

$$\mathbf{P}_{\mathsf{L}}(\mathbf{r},t) = \epsilon_0 \boldsymbol{\chi}^{(1)}(\mathbf{r}) \, \mathbf{E}(\mathbf{r},t)$$





 \Rightarrow Wave equation for inhomogeneous nonlinear media:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},t) + \mu_0 \epsilon_0 \epsilon_r \frac{\partial^2 \mathbf{E}(\mathbf{r},t)}{\partial t^2} = -\mu_0 \frac{\partial^2 \mathbf{P}_{\mathsf{NL}}(\mathbf{r},t)}{\partial t^2}$$

Simplification for isotropic homogeneous nonlinear media:

$$\nabla^{2}\mathbf{E}(\mathbf{r},t) - \frac{n^{2}}{c^{2}}\frac{\partial^{2}\mathbf{E}(\mathbf{r},t)}{\partial t^{2}} = \mu_{0}\frac{\partial^{2}\mathbf{P}_{\mathsf{NL}}(\mathbf{r},t)}{\partial t^{2}}.$$

Simplifying assumptions:

- Linearly polarized, "plane-wave-like" fields (Without loss of generality: Polarization along x, propagation along z)
 - \Rightarrow Represent by scalar field quantities E(z,t) and P(z,t)
- Polarization responds instantaneously to applied electric field ("memory-less material")
 - \Rightarrow Expand nonlinear time-domain relationship between *P* and *E* into a power series of scalar quantities:

$$P(z,t) = \epsilon_0 \chi^{(1)} E(z,t) + \epsilon_0 \chi^{(2)} E^2(z,t) + \epsilon_0 \chi^{(3)} E^3(z,t) + \dots$$

= $P_{\text{L}}(z,t) + P_{\text{NL}}(z,t)$
 $P_{\text{L}}(z,t) = \epsilon_0 \chi^{(1)} E(z,t)$
 $P_{\text{NL}}(z,t) = \epsilon_0 \chi^{(2)} E^2(z,t) + \epsilon_0 \chi^{(3)} E^3(z,t) + \dots$



Representation of nonlinear polarization



Example: Second-order nonlinear polarization

Assume superposition of two monochromatic plane waves, oscillating with frequency ω_1 and ω_2 :

$$E(z,t) = \frac{1}{2} \left(\underline{E}(\omega_1) \exp\left(j \left(\omega_1 t - k_1 z\right)\right) + \underline{E}(\omega_2) \exp\left(j \left(\omega_2 t - k_2 z\right)\right) + \text{c.c.} \right).$$

⇒ The second-order nonlinear polarization contains components at all sum and difference frequencies:

$$P_{\mathsf{NL}}(z,t) = \frac{1}{4} \epsilon_0 \chi^{(2)} \left(\underline{E}_1^2 e^{j2(\omega_1 t - k_1 z)} + \text{c.c.} + \underline{E}_2^2 e^{j2(\omega_2 t - k_2 z)} + \text{c.c.} + 2|\underline{E}_1|^2 + 2|\underline{E}_2|^2 + 2|\underline{E}_2|^2 + 2|\underline{E}_2|^2 + 2|\underline{E}_2 e^{j((\omega_1 + \omega_2)t - (k_1 + k_2)z)} + \text{c.c.} + 2\underline{E}_1 \underline{E}_2 e^{j((\omega_1 - \omega_2)t - (k_1 - k_2)z)} + \text{c.c.} + 2\underline{E}_1 \underline{E}_2^* e^{j((\omega_1 - \omega_2)t - (k_1 - k_2)z)} + \text{c.c.} \right)$$
Second-harmonic generation (SHG)
$$Optical rectification (OR)$$

$$Sum and difference frequency generation (SFG, DFG)$$

Note: The individual expressions of the second-order polarization exhibit plane wave-like space and time dependences. However, the wavenumbers differ from that of a plane electromagnetic wave at the same frequency, $k(\omega_1) + k(\omega_2) \neq k(\omega_1 + \omega_2)$.



Solution of nonlinear wave equation: Slowly-varying envelope approximation (SVEA)



Solution ansatz:

- Represent electric field E(z,t) and nonlinear polarization $P_{NL}(z,t)$ as a superposition of plane-wave-like fields, oscillating at different frequencies ω_l

۹

Introduce (weakly) time- and space-dependent amplitudes to account for nonlinear interaction

$$E(z,t) = \frac{1}{2} \left(\sum_{l} \underline{E}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} P_{\text{NL}}(z,t) = \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}z-k_{p,l}z\right)\right) + \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,\omega_$$

omplex conjugate of preceding expression.

where
$$\left| \frac{\partial^2 \underline{E}(z,t,\omega_l)}{\partial t^2} \right| \ll \omega_l \left| \frac{\partial \underline{E}(z,t,\omega_l)}{\partial t} \right|$$

 $\left| \frac{\partial^2 \underline{E}(z,t,\omega_l)}{\partial z^2} \right| \ll k_l \left| \frac{\partial \underline{E}(z,t,\omega_l)}{\partial z} \right|$

Slowly-varying envelope approximation (SVEA): The envelope $\underline{E}(z,t,\omega_l)$ and $\underline{P}(z,t,\omega_l)$ vary much "slower" with z and t than the respective carrier wave.

 \Rightarrow First-order DEq for describing evolution of the envelope <u>E(z,t, ω_l)</u>:

$$\frac{\partial \underline{E}(z,t,\omega_l)}{\partial z} + \frac{n}{c} \frac{\partial \underline{E}(z,t,\omega_l)}{\partial t} = -j \frac{\omega_l}{2\epsilon_0 cn} \underline{P}_{\mathsf{NL}}(z,t,\omega_l) e^{-j(k_{p,l}-k_l)z}.$$



Lecture 4



 \Rightarrow Wave equation for inhomogeneous nonlinear media:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},t) + \mu_0 \epsilon_0 \epsilon_r \frac{\partial^2 \mathbf{E}(\mathbf{r},t)}{\partial t^2} = -\mu_0 \frac{\partial^2 \mathbf{P}_{\mathsf{NL}}(\mathbf{r},t)}{\partial t^2}$$

Simplification for isotropic homogeneous nonlinear media:

$$\nabla^{2}\mathbf{E}(\mathbf{r},t) - \frac{n^{2}}{c^{2}}\frac{\partial^{2}\mathbf{E}(\mathbf{r},t)}{\partial t^{2}} = \mu_{0}\frac{\partial^{2}\mathbf{P}_{\mathsf{NL}}(\mathbf{r},t)}{\partial t^{2}}.$$

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 $P_{\text{L}}(z,t) = \epsilon_0 \chi^{(1)} E(z,t)$
 $P_{\text{NL}}(z,t) = \epsilon_0 \chi^{(2)} E^2(z,t) + \epsilon_0 \chi^{(3)} E^3(z,t) + \dots$



Representation of nonlinear polarization



Example: Second-order nonlinear polarization

Assume superposition of two monochromatic plane waves, oscillating with frequency ω_1 and ω_2 :

$$E(z,t) = \frac{1}{2} \left(\underline{E}(\omega_1) \exp\left(j \left(\omega_1 t - k_1 z\right)\right) + \underline{E}(\omega_2) \exp\left(j \left(\omega_2 t - k_2 z\right)\right) + \text{c.c.} \right).$$

⇒ The second-order nonlinear polarization contains components at all sum and difference frequencies:

$$P_{\mathsf{NL}}(z,t) = \frac{1}{4} \epsilon_0 \chi^{(2)} \left(\underline{E}_1^2 e^{j2(\omega_1 t - k_1 z)} + \text{c.c.} + \underline{E}_2^2 e^{j2(\omega_2 t - k_2 z)} + \text{c.c.} + 2|\underline{E}_1|^2 + 2|\underline{E}_2|^2 + 2|\underline{E}_2|^2 + 2|\underline{E}_2|^2 + 2|\underline{E}_2 e^{j((\omega_1 + \omega_2)t - (k_1 + k_2)z)} + \text{c.c.} + 2\underline{E}_1 \underline{E}_2 e^{j((\omega_1 - \omega_2)t - (k_1 - k_2)z)} + \text{c.c.} + 2\underline{E}_1 \underline{E}_2^* e^{j((\omega_1 - \omega_2)t - (k_1 - k_2)z)} + \text{c.c.} \right)$$
Second-harmonic generation (SHG)
$$Optical rectification (OR)$$

$$Sum and difference frequency generation (SFG, DFG)$$

Note: The individual expressions of the second-order polarization exhibit plane wave-like space and time dependences. However, the wavenumbers differ from that of a plane electromagnetic wave at the same frequency, $k(\omega_1) + k(\omega_2) \neq k(\omega_1 + \omega_2)$.



Solution of nonlinear wave equation: Slowly-varying envelope approximation (SVEA)



Solution ansatz:

- Represent electric field E(z,t) and nonlinear polarization $P_{NL}(z,t)$ as a superposition of plane-wave-like fields, oscillating at different frequencies ω_l

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Introduce (weakly) time- and space-dependent amplitudes to account for nonlinear interaction

$$E(z,t) = \frac{1}{2} \left(\sum_{l} \underline{E}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} P_{\text{NL}}(z,t) = \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}t-k_{p,l}z\right)\right) + \text{c.c.} \right) \int_{V}^{C_{l}} \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,t,\omega_{l}) \exp\left(j\left(\omega_{l}z-k_{p,l}z\right)\right) + \frac{1}{2} \left(\sum_{l} \underline{P}_{\text{NL}}(z,\omega_$$

omplex conjugate of preceding expression.

where
$$\left| \frac{\partial^2 \underline{E}(z,t,\omega_l)}{\partial t^2} \right| \ll \omega_l \left| \frac{\partial \underline{E}(z,t,\omega_l)}{\partial t} \right|$$

 $\left| \frac{\partial^2 \underline{E}(z,t,\omega_l)}{\partial z^2} \right| \ll k_l \left| \frac{\partial \underline{E}(z,t,\omega_l)}{\partial z} \right|$

Slowly-varying envelope approximation (SVEA): The envelope $\underline{E}(z,t,\omega_l)$ and $\underline{P}(z,t,\omega_l)$ vary much "slower" with z and t than the respective carrier wave.

 \Rightarrow First-order DEq for describing evolution of the envelope <u>E(z,t, ω)</u>:

$$\frac{\partial \underline{E}(z,t,\omega_l)}{\partial z} + \frac{n}{c} \frac{\partial \underline{E}(z,t,\omega_l)}{\partial t} = -j \frac{\omega_l}{2\epsilon_0 cn} \underline{P}_{\mathsf{NL}}(z,t,\omega_l) e^{-j(k_{p,l}-k_l)z}.$$



Retarded time frame



Use a retarded time frame to represent electric field and nonlinear polarization:

$$t' = t - \frac{nz}{c},$$

$$z' = z,$$

$$\underline{E}(z, t, \omega_l) = \underline{E}'(z, t - \frac{nz}{c}, \omega_l).$$

 \Rightarrow First-order DEq:

$$\frac{\partial \underline{E}'(z',t',\omega_l)}{\partial z'} = -j \frac{\omega_l}{2\epsilon_0 cn} \underline{P}'_{\mathsf{NL}}(z',t',\omega_l) e^{-j(k_{p,l}-k_l)z'}.$$

- Nonlinear polarization <u>P</u>[•]_{NL}(z',t',ω_l) acts as a source for new frequency components
- Depending of the relative phase between $\underline{P}_{NL}^{\prime}(z',t',\omega_l)$ and $\underline{E}^{\prime}(z',t',\omega_l)$, the nonlinear polarization can cause amplification, absorption or phase shifts.
- Efficient nonlinear interaction requires proper phase matching, $k_{p,l} k_l \approx 0$

\Rightarrow <u>SPM/XPM</u>



Second-order nonlinear processes



Recall: Frequency components of second-order nonlinear polarization for superposition of two monochromatic waves with frequencies ω_1 and ω_2 :

72 27.06.2018 Christian Koos


Second-harmonic generation (SHG):



Optical rectification (OR):
$$\omega_p = 0, k_p = 0$$

 $\underline{P}_{NL}(z, t, 0) = \begin{pmatrix} 1 \\ 2 \\ - \end{pmatrix}_{0} \chi^{(2)} \underline{E}(z, t, \omega_1) \underline{E}(z, t, -\omega_1)$

Frequency coomponents at $\omega = 0$ need special consideration (will be explained later!)

Figures adapted from Boyd, Nonlinear Optics

Institute of Photonics and Quantum Electronics





Summary of third-order nonlinear processes



Process	Abbreviation	Involved frequencies	Degeneracy factor D
Third-harmonic generation	THG	$(+\omega_1,+\omega_1,+\omega_1)$	1
Self-phase modulation	SPM	$(+\omega_1,-\omega_1,+\omega_1)$	3
Cross-phase modulation	XPM	$(+\omega_2,-\omega_2,+\omega_1)$	6
Non-degenerate four-wave mixing	(non-degenerate) FWM	$(+\omega_1,+\omega_2,+\omega_3)$	6
		$(+\omega_1,+\omega_2,-\omega_3)$	6
Degenerate four-wave mixing	(degenerate) FWM	$(+\omega_1,+\omega_1,+\omega_2)$	3
		$(+\omega_1,+\omega_1,-\omega_2)$	3

Note: Degeneracy factor *D* corresponds to the number of distinct permutations of the triple set of involved frequencies



Generation of one new photon (third-order sum frequency generation):





Generation of one new photon (third-order sum frequency generation):

$$\omega_p = 2\omega_1 + \omega_2, \ k_p = 2k_1 + k_2$$
$$\underline{P}_{\text{FWM}}(z, t, \omega_p = 2\omega_1 + \omega_2) = \frac{3}{4}\epsilon_0 \chi^{(3)} \underline{E}^2(z, t, \omega_1) \underline{E}(z, t, \omega_2).$$

Generation of two new photons:

$$\omega_p = 2\omega_1 - \omega_2, \ k_p = 2k_1 - k_2$$

$$\underline{P}_{\text{FWM}}(z, t, \omega_p = 2\omega_1 - \omega_2) = \frac{3}{4}\epsilon_0 \chi^{(3)} \underline{E}^2(z, t, \omega_1) \underline{E}^*(z, t, \omega_2)$$





Third-harmonic generation (THG):

$$\omega_p = 3\omega_1, k_p = 3k_1$$

$$\underline{P}_{\mathsf{THG}}(z,t,3\omega_1) = \frac{1}{4}\epsilon_0 \chi^{(3)} \underline{E}^3(z,t,\omega_1)$$

$$\omega_p = \omega_1, k_p = k_1$$

$$\underline{P}_{\mathsf{SPM}}(z,t,\omega_1) = \frac{3}{4} \epsilon_0 \chi^{(3)} |\underline{E}(z,t,\omega_1)|^2 \underline{E}(z,t,\omega_1) \quad \mathsf{SPM}$$

$$\underline{P}_{\mathsf{XPM}}(z,t,\omega_1) = \frac{6}{4} \epsilon_0 \chi^{(3)} |\underline{E}(z,t,\omega_2)|^2 \underline{E}(z,t,\omega_1) \quad \mathsf{XPM}$$

 \Rightarrow Nonlinear wave equation

78 27.06.2018 Christian Koos

$$\hbar \omega_1$$

 $\hbar \omega_1$
 $\hbar \omega_1$
 $\hbar \omega_1$

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 $\hbar\omega_1$



 $\hbar\omega_2$







The intensity-dependent refractive index leads to modulation of a light beam by itself (SPM) or by another beam of light (XPM). For the same intensity of the modulating beam, the effect of XPM is twice as large as SPM!

Figure adapted from Boyd, Nonlinear Optics



Kerr effect and intensity-dependent refractive index



Consider a single monochromatic wave at frequency ω_1 in a third-order nonlinear medium => SPM is the dominant nonlinear effect!

$$\frac{\partial \underline{E}(z,t,\omega_1)}{\partial z} = -j \frac{3\omega_1 \chi^{(3)}}{8cn(\omega_1)} |\underline{E}(z,t,\omega_1)|^2 \underline{E}(z,t,\omega_1)$$

⇒ SPM causes a negative phase shift that is proportional to the square of the field magnitude, i.e., proportional to the intensity. This can be interpreted as an intensitydependent refractive index.

$$n(z,t,\omega_1) = n_0(\omega_1) + n_2 I(z,t,\omega_1)$$

where $n_2 = \frac{3Z_0}{4n_0^2}\chi^{(3)}$. Kerr coefficient

Note: For a superposition of two waves oscillating at frequencies ω_1 and ω_2 , the refractive index seen by wave 1 will also be influenced by cross-phase modulation (XPM) due to wave 2. Note that XPM has twice the degeneracy factor of SPM.

$$n(z,t,\omega_{1}) = n_{0}(\omega_{1}) + n_{2}(I(z,t,\omega_{1}) + 2I(z,t,\omega_{2})).$$



Third-order nonlinear coefficients of various materials



Material	<i>n</i> ₀	$\chi^{(3)} (m^2/V^2)$	$n_2 ({\rm cm}^2/{\rm W})$
Crystals			
Al ₂ O 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 ₂ S 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}
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}

Material	<i>n</i> ₀	$\chi^{(3)} (m^2/V^2)$	$n_2 ({\rm cm}^2/{\rm W})$
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}	

^{*a*} This table assumes the definition of the third-order susceptibility $\chi^{(3)}$ used in this book, as given for instance by Eq. (1.1.2) or by Eq. (1.3.21). This definition is consistent with that introduced by Bloembergen (1964). Some workers use an alternative definition which renders their values four times smaller. In compiling this table we have converted the literature values when necessary to the present definition.

The quantity n_2 is the coefficient of the intensity-dependent refractive index which is defined such that $n = n_0 + n_2 I$, where n_0 is the linear refractive index and I is the laser intensity. The relation between n_2 and $\chi^{(3)}$ is consequently $n_2 = 12\pi^2 \chi^{(3)}/n_0^2$. When the intensity is measured in W/cm² and $\chi^{(3)}$ is measured in electrostatic units (esu), that is, in cm² statvolt⁻², the relation between n_2 and $\chi^{(3)}$ becomes $n_2(\text{cm}^2/\text{W}) = 0.0395\chi^{(3)}(\text{esu})/n_0^2$. The quantity β is the coefficient describing two-photon absorption.

Note: Even though Boyd uses a different definition for complex electric field amplitudes, the values for $\chi^{(3)}$ and n_2 are consistent with the definitions used in this lecture. Adapted from Boyd, Nonlinear Optics



Lecture 5

Retarded time frame



Use a retarded time frame to represent electric field and nonlinear polarization:

$$t' = t - \frac{nz}{c},$$

$$z' = z,$$

$$\underline{E}(z, t, \omega_l) = \underline{E}'(z, t - \frac{nz}{c}, \omega_l).$$

 \Rightarrow First-order DEq:

$$\frac{\partial \underline{E}'(z',t',\omega_l)}{\partial z'} = -j \frac{\omega_l}{2\epsilon_0 cn} \underline{P}'_{\mathsf{NL}}(z',t',\omega_l) e^{-j(k_{p,l}-k_l)z'}.$$

- Nonlinear polarization <u>P</u>[•]_{NL}(z',t',ω_l) acts as a source for new frequency components
- Depending of the relative phase between $\underline{P}_{NL}^{\prime}(z',t',\omega_l)$ and $\underline{E}^{\prime}(z',t',\omega_l)$, the nonlinear polarization can cause amplification, absorption or phase shifts.
- Efficient nonlinear interaction requires proper phase matching, $k_{p,l} k_l \approx 0$

\Rightarrow <u>SPM/XPM</u>



Kerr effect and intensity-dependent refractive index



Consider a single monochromatic wave at frequency ω_1 in a third-order medium => SPM is the dominant nonlinear effect!

$$\frac{\partial \underline{E}(z,t,\omega_1)}{\partial z} = -j \frac{3\omega_1 \chi^{(3)}}{8cn(\omega_1)} |\underline{E}(z,t,\omega_1)|^2 \underline{E}(z,t,\omega_1)$$

⇒ SPM causes a negative phase shift that is proportional to the square of the field magnitude, i.e., proportional to the intensity. This can be interpreted as an intensitydependent refractive index.

$$n(z,t,\omega_1) = n_0(\omega_1) + n_2 I(z,t,\omega_1)$$

where $n_2 = \frac{3Z_0}{4n_0^2}\chi^{(3)}$. Kerr coefficient

Note: For a superposition of two waves oscillating at frequencies ω_1 and ω_2 , the refractive index seen by wave 1 will also be influenced by cross-phase modulation (XPM) due to wave 2. Note that XPM has twice the degeneracy factor of SPM.

$$n(z,t,\omega_{1}) = n_{0}(\omega_{1}) + n_{2}(I(z,t,\omega_{1}) + 2I(z,t,\omega_{2})).$$



Parametric versus nonparametric processes



Parametric processes:

- Quantum state of the material remains unchanged
- No transfer of energy, momentum, or angular momentum between the optical field and the material => Momentum and energy conservation:

$$\sum_{i} \omega_i = \sum_{f} \omega_f, \ \sum_{i} k_i = \sum_{f} k_f,$$

- Given by real part of complex susceptibility
- Note: Quantum system can be removed from a real energy state only for brief time intervals ∠t, in which it resides in a so-called virtual energy level:

 $\Delta t \Delta W < \hbar$

 \Rightarrow Ultra-fast response!

• Examples: SFG, SHG, DFG, THG, SPM, XPM, FWM

Nonparametric processes:

- Transfer of a quantum system from one real level to another
- Sum of photon energies is not conserved
- Given by imaginary part of complex susceptibility
- Examples: Two-photon absorption (TPA)



Two-photon absorption



Recall: Relations for cross-phase modulation (XPM) and self-phase modulation (SPM) Here: Consider case of complex nonlinear susceptibility $\chi^{(3)}$

$$\frac{\partial \underline{E}(z,t,\omega_{1})}{\partial z} = -j \frac{3\omega_{l}}{8cn} \underline{\chi}^{(3)} |\underline{E}(z,t,\omega_{1})|^{2} \underline{E}(z,t,\omega_{1}),$$

$$\frac{\partial \underline{E}(z,t,\omega_{1})}{\partial z} = -j \frac{3\omega_{l}}{4cn} \underline{\chi}^{(3)} |\underline{E}(z,t,\omega_{2})|^{2} \underline{E}(z,t,\omega_{1}).$$

- \Rightarrow Imaginary part of $\chi^{(3)}$ leads to change of amplitude
- \Rightarrow Two-photon absorption (TPA) and cross-two-photon absorption (XTPA)



Formal definition of the nonlinear optical susceptibility



So far: Time-domain treatment of special case of nonlinear polarization

- Instantaneous response of the nonlinear polarization with respect to the E-field
- Linearly polarized fields that are represented by scalar field quantities
- \Rightarrow Taylor series in the time domain:

$$P(z,t) = \epsilon_0 \chi^{(1)} E(z,t) + \epsilon_0 \chi^{(2)} E^2(z,t) + \epsilon_0 \chi^{(3)} E^3(z,t) + \dots$$

Now: General case

- Non-instantaneous response of the optical field (nonlinear material with memory)
 - \Rightarrow Volterra series in the time domain
- Vectorial field quantities
 - ⇒ Volterra kernel of the *n*-th order nonlinear susceptibility is represented as a tensor of rank n+1

$$P_{q_0}^{(n)}(t) = \epsilon_0 \sum_{q_1, \dots, q_n} \int_{-\infty}^{+\infty} \int_{-\infty} \underbrace{\chi_{q_0:q_1q_2\dots q_n}^{(n)}(\tau_1, \dots, \tau_n) E_{q_1}(t - \tau_1) \dots E_{q_n}(t - \tau_n) \, \mathrm{d}\tau_1 \dots \mathrm{d}\tau_n.$$

 $q_0, q_1 \dots, q_n \in \{x, y, z\}$ denote the various vectorial components of the electric field, the polarization, and the corresponding elements of the susceptibility tensor.



Short-form tensor notation



Replace sum over tensor elements and vectorial components by a short-form notation

Second-order nonlinear susceptibility:

$$P_{q}^{(2)}(t) = \epsilon_{0} \sum_{r,s} \iint_{\tau_{1},\tau_{2}} \chi_{q;r,s}^{(2)}(\tau_{1},\tau_{2}) E_{r}(t-\tau_{1}) E_{s}(t-\tau_{2}) d\tau_{1} d\tau_{2},$$

$$P^{(2)}(t) = \epsilon_{0} \iint_{\tau_{1},\tau_{2}} \chi^{(2)}(\tau_{1},\tau_{2}) : \mathbf{E}(t-\tau_{1}) \mathbf{E}(t-\tau_{2}) d\tau_{1} d\tau_{2},$$
where $\chi^{(2)}: \mathbf{E}\mathbf{E} = \sum_{q,r,s} \mathbf{e}_{q} \chi_{q;r,s}^{(2)} E_{r} E_{s},$
Short-form notation of tensor product contains summation over all vector components and tensor elements

General case:

$$\underline{\chi}^{(n)} : \mathbf{E}(\tau_1) \mathbf{E}(\tau_2) \dots \mathbf{E}(\tau_n) = \sum_{q_0, q_1, \dots, q_n} e_{q_0} \chi^{(n)}_{q_0: q_1 q_2 \dots q_n} E_{q_1}(\tau_1) E_{q_2}(\tau_2) \dots E_{q_n}(\tau_n)$$

where $q_0, q_1 ..., q_n \in \{x, y, z\}$

88 27.06.2018 Christian Koos





Consider simplified case of second-order nonlinearity:

$$P_q^{(2)}(t) = \epsilon_0 \sum_{r,s} \iint_{\tau_1,\tau_2} \chi_{q:r,s}^{(2)}(\tau_1,\tau_2) E_r(t-\tau_1) E_s(t-\tau_2) d\tau_1 d\tau_2,$$

$$\widetilde{P}_{q}^{(2)}(\omega) = \frac{1}{2\pi} \epsilon_{0} \sum_{r,s} \int_{\omega_{1}} \widetilde{\chi}_{q:r,s}^{(2)}(\omega_{1},\omega-\omega_{1}) \widetilde{E}_{r}(\omega_{1}) \widetilde{E}_{s}(\omega-\omega_{1}) d\omega_{1}$$

where
$$\tilde{\chi}_{q:r,s}^{(2)}(\omega_1,\omega_2) = \iint_{\tau_1,\tau_2} \chi_{q:r,s}^{(2)}(\tau_1,\tau_2) e^{-j\omega_1\tau_1} e^{-j\omega_2\tau_2} \mathrm{d}\tau_1 \mathrm{d}\tau_2$$

General case:

$$\widetilde{P}_{q_{0}}^{(n)}(\omega) = \frac{\epsilon_{0}}{(2\pi)^{n-1}} \sum_{q_{1}...,q_{n}} \int \cdots \int \underline{\widetilde{\chi}}_{q_{0}:q_{1},...q_{n}}^{(n)} \left(\omega : \omega_{1}, \ldots, \omega_{n-1}, \omega - \sum_{m=1}^{n-1} \omega_{m} \right) \\ \times \widetilde{E}_{q_{1}}(\omega_{1}) \ldots \widetilde{E}_{q_{n-1}}(\omega_{n-1}) \widetilde{E}_{q_{n}} \left(\omega - \sum_{m=1}^{n-1} \omega_{m} \right) d\omega_{1} \ldots d\omega_{n-1}$$
where
$$+\infty$$

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$$\underline{\tilde{\chi}}_{q_0:q_1q_2\ldots q_n}^{(n)} \left(\sum_{m=1}^n \omega_m : \omega_1, \ldots, \omega_n \right) = \int_{-\infty}^{+\infty} \underbrace{\chi}_{q_0:q_1,\ldots,q_n}^{(n)} \left(\tau_1, \ldots, \tau_n \right) e^{-j \,\omega_1 \tau_1} \ldots e^{-j \,\omega_n \tau_n} \, \mathrm{d}\tau_1 \ldots \, \mathrm{d}\tau_n$$



The nonlinear susceptibility for complex time-domain amplitudes

Example: Second-order nonlinearities

$$\mathbf{P}^{(2)}(t) = \epsilon_0 \iint_{\tau_1,\tau_2} \chi^{(2)}(\tau_1,\tau_2) : \mathbf{E}(t-\tau_1) \mathbf{E}(t-\tau_2) \, \mathrm{d}\tau_1 \mathrm{d}\tau_2$$
$$\mathbf{E}(t) = \frac{1}{2} \sum_{m=-M}^{M} \underline{\mathbf{E}}(\omega_m) e^{j\omega_m t} \quad \text{where} \quad \begin{array}{l} \omega_{-m} = -\omega_m, \quad \omega_0 = 0\\ \underline{\mathbf{E}}(\omega_{-m}) = \underline{\mathbf{E}}^*(\omega_m). \end{array}$$

For now: No involvement of DC fields, $\underline{\mathbf{E}}(\omega_0) = 0$

$$\Rightarrow \mathbf{P}^{(2)}(t) = \frac{1}{4} \epsilon_0 \sum_{l,m} \underline{\widetilde{\chi}}^{(2)}(\omega_{\Sigma} : \omega_l, \omega_m) : \underline{\mathbf{E}}(\omega_l) \underline{\mathbf{E}}(\omega_m) e^{\mathbf{j}(\omega_l + \omega_m)t}$$

Introduce complex time-domain amplitudes for nonlinear polarization:

$$P^{(2)}(t) = \frac{1}{2} \sum_{p=-M}^{M} \underline{P}^{(2)}(\omega_p) e^{j\omega_p t}$$

$$\Rightarrow \qquad \underline{P}^{(2)}(\omega_p) = \frac{1}{2} \epsilon_0 \sum_{\mathbb{S}(\omega_p)} \underline{\chi}^{(2)}(\omega_p : \omega_l, \omega_m) : \underline{\mathbf{E}}(\omega_l) \underline{\mathbf{E}}(\omega_m)$$

where $\mathbb{S}(\omega_p) = \{(l,m) | \omega_l + \omega_m = \omega_p\}$

90 27.06.2018 Christian Koos



Third- and higher-order nonlinearities



Complex time-domain amplitude of third-order nonlinear polarization:

$$\underline{\mathbf{P}}^{(3)}(\omega_p) = \frac{1}{4} \epsilon_0 \sum_{\mathbb{S}(\omega_p)} \underline{\chi}^{(3)}(\omega_p : \omega_l, \omega_m, \omega_o) : \underline{\mathbf{E}}(\omega_l) \underline{\mathbf{E}}(\omega_m) \underline{\mathbf{E}}(\omega_n)$$

where $\mathbb{S}(\omega_p) = \{(l, m, n) | \omega_l + \omega_m + \omega_n = \omega_p\}.$

Complex time-domain amplitude of *n*-th nonlinear polarization:

$$\underline{\mathbf{P}}^{(n)}(\omega_p) = \frac{1}{2^{n-1}} \epsilon_0 \sum_{\mathbb{S}(\omega_p)} \underline{\chi}^{(n)} \left(\omega_p : \omega_{l_1}, \dots, \omega_{l_n} \right) : \underline{\mathbf{E}}(\omega_{l_1}) \dots \underline{\mathbf{E}}(\omega_{l_n}),$$

where
$$\mathbb{S}(\omega_p) = \{(l_1, \dots, l_n) | \omega_{l_1} + \dots + \omega_{l_n} = \omega_p\}$$



Zero frequencies and DC fields...



Problem: Inconsistencies of time-domain representations with respect to zero frequencies $\omega_0 = 0$ and the associated DC fields

 \Rightarrow Use more rigorous definition of time-domain signals and complex amplitudes:

Complex time-domain amplitude of *n*-th nonlinear polarization:

$$\underline{\mathbf{P}}^{(n)}(\omega_p) = \frac{1}{2^{n-1}} \epsilon_0 \sum_{\mathbb{S}(\omega_p)} \frac{\left(1 + \delta_{l_1,0}\right) \dots \left(1 + \delta_{l_n,0}\right)}{1 + \delta_{p,0}} \underline{\chi}^{(n)}\left(\omega_p : \omega_{l_1}, \dots, \omega_{l_n}\right) : \underline{\mathbf{E}}(\omega_{l_1}) \dots \underline{\mathbf{E}}(\omega_{l_n})$$

Examples:

Optical rectification (OR):
$$\underline{\mathbf{P}}^{(2)}(\omega_3 = 0) = \frac{1}{2} \epsilon_0 \underline{\chi}^{(2)}(0 : \omega_1, -\omega_1) : \underline{\mathbf{E}}(\omega_1) \underline{\mathbf{E}}^*(\omega_1)$$

Electro-optic Kerr effect (Quadratic electro-optic effect): $\underline{\mathbf{P}}^{(3)}(\omega_1) = 3\epsilon_0 \underline{\chi}^{(3)}(\omega_1 : \omega_1, 0, 0) : \underline{\mathbf{E}}(\omega_1) \underline{\mathbf{E}}(0) \underline{\mathbf{E}}(0)$



Lecture 6

Zero frequencies and DC fields...



Problem: Inconsistencies of time-domain representations with respect to zero frequencies $\omega_0 = 0$ and the associated static ("DC") fields

 \Rightarrow Use more rigorous definition of time-domain signals and complex amplitudes:

Complex time-domain amplitude of *n*-th nonlinear polarization:

$$\underline{\mathbf{P}}^{(n)}(\omega_p) = \frac{1}{2^{n-1}} \epsilon_0 \sum_{\mathbb{S}(\omega_p)} \frac{\left(1 + \delta_{l_1,0}\right) \dots \left(1 + \delta_{l_n,0}\right)}{1 + \delta_{p,0}} \underline{\chi}^{(n)}\left(\omega_p : \omega_{l_1}, \dots, \omega_{l_n}\right) : \underline{\mathbf{E}}(\omega_{l_1}) \dots \underline{\mathbf{E}}(\omega_{l_n})$$

Examples:

Optical rectification (OR):
$$\underline{\mathbf{P}}^{(2)}(\omega_3 = 0) = \frac{1}{2} \epsilon_0 \underline{\chi}^{(2)}(0 : \omega_1, -\omega_1) : \underline{\mathbf{E}}(\omega_1) \underline{\mathbf{E}}^*(\omega_1)$$

Electro-optic Kerr effect (Quadratic electro-optic effect): $\underline{\mathbf{P}}^{(3)}(\omega_1) = 3\epsilon_0 \underline{\chi}^{(3)}(\omega_1 : \omega_1, 0, 0) : \underline{\mathbf{E}}(\omega_1) \underline{\mathbf{E}}(0) \underline{\mathbf{E}}(0)$



Properties of the nonlinear optical susceptibility tensor



• Causality:

 $\chi_{q_0:q_1q_2...q_n}^{(n)}(\tau_1,\tau_2,\ldots,\tau_n) = 0$ for $\tau_1 < 0 \lor \tau_2 < 0 \cdots \lor \tau_n < 0.$

- ⇒ Frequency-domain relationships (Kramers-Kronig relations) exist for some nonlinear optical processes, but nor for all!
- **Reality of fields:** Electromagnetic field quantities must be represented by real numbers in the time domain!
 - \Rightarrow Positive- and negative-frequency components of the complex susceptibility tensor are the complex conjugate of each other:

$$\underline{\mathbf{E}}(\omega_l) = \underline{\mathbf{E}}^*(-\omega_l) \qquad \underline{\mathbf{P}}(\omega_l) = \underline{\mathbf{P}}^*(-\omega_l)$$

$$\underline{\chi}_{q_0:q_1q_2...q_n}^{(n)}(\omega_{\Sigma}:\omega_1,\omega_2,\ldots,\omega_n) = \left[\underline{\chi}_{q_0:q_1q_2...q_n}^{(n)}(-\omega_{\Sigma}:-\omega_1,-\omega_2,\ldots,-\omega_n)\right]^*$$

Intrinsic permutation symmetry:

Nonlinear susceptibility tensor element remains unchanged if frequency arguments $\omega_1 \dots \omega_n$ (not: ω_{Σ}) and corresponding Cartesian indices are swapped simultaneously

$$\underline{\chi}_{q_0:q_1\ldots q_i q_j\ldots q_n}^{(n)} \left(\omega_{\Sigma}: \omega_1, \ldots, \omega_i, \omega_j, \ldots, \omega_n \right) = \underline{\chi}_{q_0:q_n\ldots q_j q_i\ldots q_1}^{(n)} \left(\omega_{\Sigma}: \omega_n, \ldots, \omega_j, \omega_i, \ldots, \omega_1 \right)$$



Properties of the nonlinear optical susceptibility tensor



Symmetries for lossless media:

All components of the nonlinear susceptibility tensor are real:

$$\underline{\chi}_{q_0:q_1...q_iq_j...q_n}^{(n)}\left(\omega_{\Sigma}:\omega_1,\ldots,\omega_i,\omega_j,\ldots,\omega_n\right)\in\mathbb{R}$$

Permutation symmetry holds also for the resulting frequency ω_{Σ} Note: Signs must be changed appropriately when interchanging the first argument with any other argument.

$$\underline{\tilde{\chi}}_{q_0:q_1\dots q_i\dots q_n}^{(n)} \left(\omega_{\Sigma} : \omega_1, \dots, \omega_i, \dots, \omega_n \right) = \underline{\tilde{\chi}}_{q_i:q_1\dots q_0\dots q_n}^{(n)} \left(\omega_i : -\omega_1, \dots, \omega_{\Sigma}, \dots, -\omega_n \right)$$

• Kleinman's symmetry:

Operated at frequencies far below their lowest resonance frequency

 \Rightarrow The medium is lossless and the nonlinear susceptibility is essentially independent of frequency.

 \Rightarrow The frequency arguments can be permuted without permuting the indices:

$$\underline{\chi}_{q_0:q_1\dots q_i q_j\dots q_n}^{(n)} \left(\omega_{\Sigma} : \omega_1, \dots, \omega_i, \omega_j, \dots, \omega_n \right) = \underline{\chi}_{q_0:q_1\dots q_i q_j\dots q_n}^{(n)} \left(\omega_{\Sigma} : \omega_1, \dots, \omega_j, \omega_i, \dots, \omega_n \right)$$

Contracted notation for second-order nonlinear susceptibility



Assumption: Kleinman symmetry applies, i.e., frequency arguments can be permuted without permuting the corresponding vector indices.

⇒ Exploit permutability of last two frequency arguments to introduce contracted notation:

Represent nonlinear susceptibility tensor as a (3×6) – matrix:

$$\mathbf{d} = \begin{pmatrix} d_{x1} \, d_{x2} \, d_{x3} \, d_{x4} \, d_{x5} \, d_{x6} \\ d_{y1} \, d_{y2} \, d_{y3} \, d_{y4} \, d_{y5} \, d_{y6} \\ d_{z1} \, d_{z2} \, d_{z3} \, d_{z4} \, d_{z5} \, d_{z6} \end{pmatrix}$$

Exploitation of full Kleinman symmetry: Only 10 independent elements

$$\mathbf{d} = \begin{pmatrix} d_{x1} \, d_{x2} \, d_{x3} \, d_{x4} \, d_{x5} \, d_{x6} \\ d_{x6} \, d_{y2} \, d_{y3} \, d_{y4} \, d_{x4} \, d_{x2} \\ d_{x5} \, d_{y4} \, d_{z3} \, d_{y3} \, d_{x3} \, d_{x4} \end{pmatrix}$$





Second-harmonic generation (SHG):

$$\begin{pmatrix} \underline{P}_{x}^{(2)}(2\omega_{1}) \\ \underline{P}_{y}^{(2)}(2\omega_{1}) \\ \underline{P}_{z}^{(2)}(2\omega_{1}) \end{pmatrix} = \epsilon_{0} \begin{pmatrix} d_{x1} d_{x2} d_{x3} d_{x4} d_{x5} d_{x6} \\ d_{x6} d_{y2} d_{y3} d_{y4} d_{x4} d_{x2} \\ d_{x5} d_{y4} d_{z3} d_{y3} d_{x3} d_{x4} \end{pmatrix} \begin{pmatrix} \underline{E}_{x}^{2}(\omega_{1}) \\ \underline{E}_{y}^{2}(\omega_{1}) \\ \underline{E}_{z}^{2}(\omega_{1}) \\ \underline{E}_{z}(\omega_{1}) \\ \underline{E}_{x}(\omega_{1}) \\ \underline{E}_{y}(\omega_{1}) \end{pmatrix}$$

Sum-frequency generation (SFG):

$$\begin{pmatrix} \underline{P}_{x}^{(2)}(\omega_{3}) \\ \underline{P}_{y}^{(2)}(\omega_{3}) \\ \underline{P}_{z}^{(2)}(\omega_{3}) \end{pmatrix} = 2\epsilon_{0} \begin{pmatrix} d_{x1} d_{x2} d_{x3} d_{x4} d_{x5} d_{x6} \\ d_{x6} d_{y2} d_{y3} d_{y4} d_{x4} d_{x2} \\ d_{x5} d_{y4} d_{z3} d_{y3} d_{x3} d_{x4} \end{pmatrix} \begin{pmatrix} \underline{E}_{x}(\omega_{1}) \underline{E}_{x}(\omega_{2}) \\ \underline{E}_{z}(\omega_{1}) \underline{E}_{z}(\omega_{2}) \\ \underline{E}_{x}(\omega_{1}) \underline{E}_{z}(\omega_{2}) + \underline{E}_{z}(\omega_{1}) \underline{E}_{y}(\omega_{2}) \\ \underline{E}_{x}(\omega_{1}) \underline{E}_{z}(\omega_{2}) + \underline{E}_{z}(\omega_{1}) \underline{E}_{x}(\omega_{2}) \\ \underline{E}_{x}(\omega_{1}) \underline{E}_{z}(\omega_{2}) + \underline{E}_{z}(\omega_{1}) \underline{E}_{x}(\omega_{2}) \\ \underline{E}_{x}(\omega_{1}) \underline{E}_{y}(\omega_{2}) + \underline{E}_{y}(\omega_{1}) \underline{E}_{x}(\omega_{2}) \end{pmatrix}$$



Exploitation of spatial symmetries



Neumann's principle: If a crystal is invariant with respect to certain geometric transformations, any of its physical properties must also be invariant with respect to the same transformations.

 \Rightarrow Investigate influence of spatial symmetries on susceptibility tensor Idea:

- Consider coordinate transformation from a coordinate system (x,y,z) to a coordinate system (x',y',z') that leaves the crystal lattice unchanged
- Since this does not change the physical situation, the tensor elements must be invariant under this coordinate transformation.
 Finstein notation: Comprise

Coordinate transformation:

$$\begin{pmatrix} E'_{x'} \\ E'_{y'} \\ E'_{z'} \end{pmatrix} = \begin{pmatrix} T_{x'x} T_{x'y} T_{x'z} \\ T_{y'x} T_{y'y} T_{y'z} \\ T_{z'x} T_{z'y} T_{z'z} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

Einstein notation: Comprises summation over all unpaired subscripts on the right-hand side.

$$E_{q'}' = T_{q'q} E_q$$

For orthogonal transformations (reflection, inversion, rotation): $\mathbf{T}^{-1} = \mathbf{T}^T \qquad (T^{-1})_{qq'} = T_{q'q}.$

$$\Rightarrow \chi_{q'_0:q'_1...q'_n}^{\prime(n)} = T_{q'_0q_0}T_{q'_1q_1}\dots T_{q'_nq_n}\chi_{q_0:q_1...q_n}^{(n)}$$

i.e., susceptibility tensor of rank (n+1) transforms like (n+1)-fold product of coordinates

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Centro-symmetric media and second-order nonlinear effects



Centro-symmetric media are invariant with respect to an inversion of coordinates:

$$T_{q'q} = -\delta_{q'q}$$

$$\Rightarrow \chi_{q_0:q_1...q_n}^{(n)} = (-1)^{n+1} \chi_{q_0:q_1...q_n}^{(n)}.$$

i.e., for even orders n all susceptibility tensor elements must vanish. Centrosymmetric media do not exhibit any second-order (even-order) nonlinearity!

Note: The same applies to amorphous materials with randomly oriented molecules that do not feature any preferential direction. Even though the microscopic structure of the material is not centrosymmetric, the macroscopic structural properties are defined as an average over random orientations of molecules and do hence not change upon inversion of coordinates. **Inversion point:** Mid-point of nearest-neighbour bonds!



Example: Diamond lattice (e.g., silicon); features inversion symmetry.



Nonlinear susceptibility tensors for different spatial symmetries



Needed: Coordinate transformations with respect to which the crystal lattice remains unchanged

 \Rightarrow Categorization of crystals by their symmetry properties:

- 32 crystal classes, characterized by 32 point groups
- 5 cubic point groups, 27 non-cubic point groups
- Point group: Set of symmetry operations that leave the crystal lattice unchanged
- Nomenclature of point groups by Schoenflies notation or international notation

Figure adapted from Ashcroft/ Mermin, Solid State Physics

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5 cubic crystallographic point groups





Figure adapted from Ashcroft/ Mermin, Solid State Physics



Lecture 7

Exploitation of spatial symmetries



Neumann's principle: If a crystal is invariant with respect to certain geometric transformations, any of its physical properties must also be invariant with respect to the same transformations.

 \Rightarrow Investigate influence of spatial symmetries on susceptibility tensor Idea:

- Consider coordinate transformation from a coordinate system (x,y,z) to a coordinate system (x',y',z') that leaves the crystal lattice unchanged
- Since this does not change the physical situation, the tensor elements must be invariant under this coordinate transformation.
 Finstein notation: Comprise

Coordinate transformation:

$$\begin{pmatrix} E'_{x'} \\ E'_{y'} \\ E'_{z'} \end{pmatrix} = \begin{pmatrix} T_{x'x} T_{x'y} T_{x'z} \\ T_{y'x} T_{y'y} T_{y'z} \\ T_{z'x} T_{z'y} T_{z'z} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

Einstein notation: Comprises summation over all unpaired subscripts on the right-hand side.

$$E_{q'}' = T_{q'q} E_q$$

For orthogonal transformations (reflection, inversion, rotation): $\mathbf{T}^{-1} = \mathbf{T}^T \qquad (T^{-1})_{qq'} = T_{q'q}.$

$$\Rightarrow \chi_{q'_0:q'_1...q'_n}^{\prime(n)} = T_{q'_0q_0}T_{q'_1q_1}\dots T_{q'_nq_n}\chi_{q_0:q_1...q_n}^{(n)}$$

i.e., susceptibility tensor of rank (n+1) transforms like (n+1)-fold product of coordinates

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Centro-symmetric media and second-order nonlinear effects



Centro-symmetric media are invariant with respect to an inversion of coordinates:

$$T_{q'q} = -\delta_{q'q}$$

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i.e., for even orders n all susceptibility tensor elements must vanish. Centrosymmetric media do not exhibit any second-order (even-order) nonlinearity!

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Example: Diamond lattice (e.g., silicon); features inversion symmetry.



Nonlinear susceptibility tensors for different spatial symmetries



Needed: Coordinate transformations with respect to which the crystal lattice remains unchanged

 \Rightarrow Categorization of crystals by their symmetry properties:

- 32 crystal classes, characterized by 32 point groups
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Figure adapted from Ashcroft/ Mermin, Solid State Physics

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5 cubic crystallographic point groups





Figure adapted from Ashcroft/ Mermin, Solid State Physics



27 noncubic crystallographic point groups







Schoenflies notation:

 C_n : *n*-fold rotation axis

- C_{nh}: *n*-fold rotation axis + horizontal mirror plane
- C_{nv} : *n*-fold rotation axis + *n* vertical mirror planes
- S_{2n}: 2*n*-fold rotation-reflection axis
- ... => see Ashcroft Mermin for more information!

Figure adapted from Ashcroft/ Mermin, Solid State Physics


Second-order susceptibility for the 32 crystal classes



Crystal System	Crystal Class	Nonvanishing Tensor Elements	Crystal System	Crystal Class	Nonvanishing Tensor Elements
Triclinic	$1 = C_1$ $\overline{1} = S_2$	All elements are independent and nonzero Each element vanishes	Cubic	432 = O $\bar{4}3m = T_d$	xyz = -xzy = yzx = -yxz = zxy = -zyx $xyz = xzy = yzx = yxz = zxy = zyx$
Monoclinic	$2 = C_2$	xyz, xzy, xxy, xyx, yxx, yyy, yzz, yzx, yxz, zyz, zzy, zxy, zyx (twofold axis parallel to ŷ)		23 = T $m3 = T_h, m3m = O_h$	xyz = yzx = zxy, xzy = yxz = zyx Each element vanishes
	$m = C_{1h}$	$xxx, xyy, xzz, xzx, xxz, yyz, yzy, yxy, yyx, zxx, zyy, zzz, zzx, zxz (mirror plane perpendicular to \hat{y})$	Trigonal	$3 = C_3$	xxx = -xyy = -yyz = -yxy, xyz = -yxz, xzy = xzx = yzy, xxz = yyz, yyy = -yxx = -xxy = -
	$2/m = C_{2h}$	Each element vanishes			zxx = zyy, zzz, zxy = -zyx
Orthorhombic	$222 = D_2$	xyz, xzy, yzx, yxz, zxy, zyx		$32 = D_3$	xxx = -xyy = -yyx = -yxy, xyz = -yxz,
	$mm2 = C_{2v}$	xzx, xxz, yyz, yzy, zxx, zyy, zzz			xzy = -yzx, zxy = -zyx
	$mmm = D_{2h}$	Each element vanishes		$3m = C_{3v}$	xzx = yzy, xxz = yyz, zxx = zyy, zzz, yyy = -yx
Tetragonal	$4 = C_4$	xyz = -yxz, xzy = -yzx, xzx = yzy, xxz = yyz,		$\bar{3} = S_6, \bar{3}m = D_{3d}$	$-xxy = -xyx$ (mirror plane perpendicular to \hat{x}) Each element vanishes
	$\bar{4} = S_4$	$z_{XX} = z_{YY}, z_{ZZ}, z_{XY} = -z_{YX}$ $x_{YZ} = y_{XZ}, x_{ZY} = y_{ZX}, x_{ZX} = -y_{ZY}, x_{XZ} = -y_{YZ}$	Hexagonal	$6 - C_{c}$	$rv_7 = -vr_7 r_7v = -v_7r r_7r - v_7v r_7r_7 - v_7r_7$
		zxx = -zyy, zxy = zyx	yx		$z_{XX} = z_{YX}, z_{ZZ}, z_{XY} = -z_{YX}$
	$422 = D_4$	xyz = -yxz, xzy = -yzx, zxy = -zyx		$\bar{6} = C_{3h}$	xxx = -xyy = -yxy = -yyx,
	$4mm = C_{4v}$	xzx = yzy, xxz = yyz, zxx = zyy, zzz			yyy = -yxx = -xyx = -xxy
	$\overline{4}2m = D_{2d}$	xyz = yxz, xzy = yzx, zxy = zyx		$622 = D_6$	xyz = -yxz, xzy = -yxz, zxy = -zyx
	$4/m = C_{4h}$	Each element vanishes		$6mm = C_{6v}$	xzx = yzy, xxz = yyz, zxx = zyy, zzz
	$4/mmm = D_{4h}$	Each element vanishes		$\overline{6}m2 = D_{3h}$	yyy = -yxx = -xxy = -xyx
				$6/m = C_{6h}$	Each element vanishes



Diamond structure, e.g., Si $m3 = O_h$ \Rightarrow All second-order elements vanish!



Zincblende structure, e.g., GaAs 43m

 ⇒ 6 nonzero elements, which are all identical, xyz = xzy = yzx = yxz
 = zxy = zyx

IPC

Figures adapted from Boyd, Nonlinear Optics



Second-order susceptibility in contracted notation







Material	Point Group	$d_{il} (pm/V)$	Watch out for
Ag3AsS3 (proustite)	$3m = C_{3v}$	$d_{22} = 18$ $d_{15} = 11$	 unspecified wavelengths
AgGaSe ₂	$\overline{4}2m = D_{2d}$	$d_{36} = 33$	• typos
AgSbS ₃ (pyrargyrite)	$3m = C_{3v}$	$d_{15} = 8$ $d_{22} = 9$	
beta-BaB ₂ O ₄ (BBO) (beta barium borate)	$3m = C_{3v}$	$d_{22} = 2.2$	
CdGeAs ₂	$\overline{4}2m = D_{2d}$	$d_{36} = 235$	
CdS	$6mm = C_{6v}$	$d_{33} = 78$ $d_{31} = -40$	🌳 👩 🌪 📍 👘 мь
GaAs	43 <i>m</i>	$d_{36} = 370$	
KH2PO4 (KDP)	2m	$d_{36} = 0.43$	
KD ₂ PO ₄ (KD*P)	2m	$d_{36} = 0.42$	
LiIO ₃	$6 = C_6$	$d_{15} = -5.5$ $d_{31} = -7$	LiNbO ₃
LiNbO3	$3m = C_{3v}$	$d_{32} = -30$ $d_{31} = -5.9$	
Quartz	$32 = D_3$	$d_{11} = 0.3$ $d_{14} = 0.008$	

Notes: Values are obtained from a variety of sources. Some of the more complete tabulations are those of R.L. Sutherland (1996), that of A.V. Smith, and the data sheets of Cleveland Crystals, Inc.

To convert to the gaussian system, multiply each entry by $(3 \times 10^{-8})/4\pi = 2.386 \times 10^{-9}$ to obtain *d* in esu units of cm/statvolt.

In any system of units, $\chi^{(2)} = 2d$ by convention.

Figures adapted from Boyd, Nonlinear Optics

Other values from the literature: d_{22} \approx 3pm/V; d_{31} \approx -5 pm/V; d_{33} \approx -25 pm/V

111 27.06.2018 Christian Koos



Isotropic

There are 21 nonzero elements, of which only 3 are independent. They are:

yyzz = zzyy = zzxx = xxzz = xxyy = yyxx, yzyz = zyzy = zxzx = xzxz = xyxy = yxyx,yzzy = zyyz = zxxz = xzzx = xyyx = yxxy;

and

xxxx = yyyy = zzzz = xxyy + xyxy + xyyx.

Cubic

For the two classes 23 and m3, there are 21 nonzero elements, of which only 7 are independent. They are:

$$xxxx = yyyy = zzzz,$$

$$yyzz = zzxx = xxyy,$$

$$zzyy = xxzz = yyxx,$$

$$yzyz = zxzx = xyxy,$$

$$zyzy = xzxz = yxyx,$$

$$yzzy = zxxz = xyyx,$$

$$zyyz = xzzx = yxyx.$$

For the three classes 432, $\overline{4}3m$, and m3m, there are 21 nonzero elements, of which only 4 are independent. They are:

$$xxxx = yyyy = zzzz,$$

$$yyzz = zzyy = zzxx = xxzz = xxyy = yyxx,$$

$$yzyz = zyzy = zxzx = xzxz = xyxy = yxyx,$$

$$yzzy = zyyz = zxxz = xzzx = xyyx = yxxy.$$



Figure adapted from Boyd, Nonlinear Optics





Hexagonal

For the three classes 6, $\overline{6}$, and 6/m, there are 41 nonzero elements, of which only 19 are independent. They are:

$$\begin{aligned} zzzz, \\ xxxx = yyyy = xxyy + xyyx + xyxy, \\ yyzz = xxzz, & xyzz = -yxzz, \\ zyyz = zxxz, & zzxy = -zzyx, \\ zyyz = zxxz, & zxyz = -zyxz, \\ yzzy = xzzx, & xzyz = -yzzx, \\ yzyz = xzxz, & xzyz = -yzxz, \\ yzyz = xzxz, & zxzy = -yzxz, \\ yzyy = zxzx, & zxzy = -yzxz, \\ yyyy = -xyxx, \\ xyyy = -xyxx, \\ xyyy = -yxxx. \end{aligned}$$

For the four classes 622, 6mm, 6/mmm, and $\overline{6}m2$, there are 21 nonzero elements, of which only 10 are independent. They are:

$$zzzz, xxxx = yyyy = xxyy + xyyx + xyxy, \begin{cases} xxyy = yyxx \\ xyyx = yxxy \\ xyyz = yxyx \\ xyyz = yxyx \end{cases}$$
$$yyzz = xzzz, \\ zzyy = zzxz, \\ yzyz = xzzz, \\ yzyz = xzzz, \\ zyzy = zxzz, \\ zyzy = zxzz. \end{cases}$$



Figure adapted from Boyd, Nonlinear Optics



trigonal

Trigonal

For the two classes 3 and $\overline{3}$, there are 73 nonzero elements, of which only 27 are independent. They are:



a monoclinic С tetragonal а Ă iC triclinic а orthorhombic С С hexagonal а

 \triangleleft

cubic

For the three classes 3m, $\bar{3}m$, and 32, there are 37 nonzero elements, of which only 14 are independent. They are:

> zzzz,xxxx = yyyy = xxyy + xyyx + xyxy,xyy = yxxy,xyy = yxyx,xyy = yxyx,xyy = yxyx,xyy = yxyx,xyy = yxyx,xyy = yyxx,xyy = yyxx,xyy = yyxx,xyy = yyxx,xyy = yxxy,xyy = yxyx,xyy = yxyy,xyy = yxyx,xyy = yxyy,xyy = yxyy,xyy = yxyx,xyy = yxyy,xyy = yxyy = yxyy,xyy = yxyy,xyy = yxyy,xyy = yxyy,xyy = yyy = yxyy,xyy = yyy = yyyy = yyy

Figure adapted from Boyd, Nonlinear Optics





```
yyzz = xxzz, \quad xxxz = -xyyz = -yxyz = -yyxz,

zzyy = zzxx, \quad xxzx = -xyzy = -yxzy = -yyzx,

zyyz = zxxz, \quad xzxx = -xzyy = -yzxy = -yzyx,

yzzy = xzzx, \quad zxxx = -zxyy = -zyxy = -zyyx,

yzyz = xzxz,

zyzy = zxzx,
```

Tetragonal

For the three classes 4, $\overline{4}$, and 4/m, there are 41 nonzero elements, of which only 21 are independent. They are:

xxxx = yyyy, zzzz,

zzxx = zzyy,	xyzz = -yxzz,	xxyy = yyxx,	xxxy = -yyyx,
xxzz = zzyy,	zzxy = -zzyx,	xyxy = yxyx,	xxyx = -yyxy,
zxzx = zyzy,	xzyz = -yzxz,	xyyx = yxxy,	xyxx = -yxyy,
xzxz = yzyz,	zxzy = -zyzx,		yxxx = -xyyy,
zxxz = zyyz,	zxyz = -zyxz,		
xzzx = yzzy,	xzzy = -yzzx.		

For the four classes 422, 4mm, 4/mmm, and $\overline{4}2m$, there are 21 nonzero elements, of which only 11 are independent. They are:

```
xxxx = yyyy, zzzz,

yyzz = xxzz, yzzy = xzzx xxyy = yyxx,

zzyy = zzxx, yzyz = xzxz xyxy = yxyx,

zyyz = zxxz, zyzy = zxzx xyyx = yxxy.
```

Monoclinic

For the three classes 2, m, and 2/m, there are 41 independent nonzero elements, consisting of:

3 elements with indices all equal,
18 elements with indices equal in pairs,
12 elements with indices having two y's one x, and one z,
4 elements with indices having three x's and one z,
4 elements with indices having three z's and one x.

Orthorhombic

For all three classes, 222, mm2, and mmm, there are 21 independent nonzero elements, consisting of:

3 elements with indices all equal, 18 elements with indices equal in pairs.

Triclinic

For both classes, 1 and 1, there are 81 independent nonzero elements.

115 27.06.2018 Christian Koos



Figure adapted from Boyd, Nonlinear Optics



Second-order nonlinear effects

Permeability and impermeability tensors of anisotropic media



Note: Second-order nonlinear effects predominantly exist in anisotropic materials! \Rightarrow Need to study wave propagation in linear anisotropic materials first!

Representation by permittivity tensor: (3x3)-matrix

$$\underline{\mathbf{D}} = \epsilon_0 \underline{\epsilon}_r \underline{\mathbf{E}}, \quad \underline{\epsilon}_r = \mathbf{I} + \underline{\chi}$$

For lossless reciprocal media (no magneto-optic effect): $\underline{\epsilon}_{ij} = \underline{\epsilon}_{ji} \in \mathbb{R}$ \Rightarrow Representation in diagonal form with respect to principal axes of the crystal:

$$\begin{pmatrix} \underline{D}x\\ \underline{D}y\\ \underline{D}z \end{pmatrix} = \epsilon_0 \begin{pmatrix} \epsilon_{xx} & 0 & 0\\ 0 & \epsilon_{yy} & 0\\ 0 & 0 & \epsilon_{zz} \end{pmatrix} \begin{pmatrix} \underline{E}x\\ \underline{E}y\\ \underline{E}z \end{pmatrix} = \epsilon_0 \begin{pmatrix} n_1^2 & 0 & 0\\ 0 & n_2^2 & 0\\ 0 & 0 & n_3^2 \end{pmatrix} \begin{pmatrix} \underline{E}x\\ \underline{E}y\\ \underline{E}z \end{pmatrix},$$

Alternatively: Representation by impermeability tensor η :

$$\underline{\mathbf{E}} = \frac{1}{\epsilon_0} \underline{\eta} \underline{\mathbf{D}}.$$

$$\begin{pmatrix} \underline{E}x\\ \underline{E}y\\ \underline{E}z \end{pmatrix} = \frac{1}{\epsilon_0} \begin{pmatrix} \eta_{xx} & 0 & 0\\ 0 & \eta_{yy} & 0\\ 0 & 0 & \eta_{zz} \end{pmatrix} \begin{pmatrix} \underline{D}x\\ \underline{D}y\\ \underline{D}z \end{pmatrix} = \frac{1}{\epsilon_0} \begin{pmatrix} \frac{1}{n_1^2} & 0 & 0\\ 0 & \frac{1}{n_2^2} & 0\\ 0 & 0 & \frac{1}{n_3^2} \end{pmatrix} \begin{pmatrix} \underline{D}x\\ \underline{D}y\\ \underline{D}z \end{pmatrix}$$



Biaxial, uniaxial, and isotropic crystals

Form of permittivity and impermeability tensor is constrained by symmetry of the crystal \Rightarrow 3 different categories; depending on their representation with respect to the principal axes:

Biaxial crystals:

• Three different principal refractive indices, $n_1 \neq n_2 \neq n_3$

Uniaxial crystals:

Two orthogonal directions, along which refractive • indices are equal.

=> Ordinary indices $n_0 = n_1 = n_2$

- Third index: Extraordinary index $n_e = n_3$
- Positive uniaxial: $n_e > n_o$
- Negative uniaxial: $n_{e} < n_{o}$ ٠
- Note: Uniaxial crystals exhibit a single axis with • threefold, four-fold, or six-fold symmetry.

Isotropic crystals:

- Higher symmetry, e.g., due to a cubic unit cell.
- All three indices are equal

$$\underline{\epsilon}_{r} = \begin{pmatrix} n_{o}^{2} & 0 & 0 \\ 0 & n_{o}^{2} & 0 \\ 0 & 0 & n_{e}^{2} \end{pmatrix}$$

$$\underline{\epsilon}_r = \begin{pmatrix} n^2 & 0 & 0 \\ 0 & n^2 & 0 \\ 0 & 0 & n^2 \end{pmatrix}$$





 $\underline{\epsilon}_{r} = \begin{pmatrix} n_{1}^{2} & 0 & 0\\ 0 & n_{2}^{2} & 0\\ 0 & 0 & n_{2}^{2} \end{pmatrix}$

Permeability tensors for different crystal classes



Representation of $\underline{\epsilon}_r$ with respect to the coordinates of the unit cell (not necessarily the principal axes of the permittivity tensor!)



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The seven crystal classes and their

trigonal

cubic

hierarchy:



Index ellipsoid





Index ellipsoid (optical indicatrix): Quadratic representation of the electric impermeability tensor.

$$\sum_{i,j} \eta_{ij} X_i X_j = 1$$

Representation with respect to the principal axes of the crystal:

$$\frac{X_1^2}{n_1^2} + \frac{X_2^2}{n_2^2} + \frac{X_3^2}{n_3^2} = 1$$

 X_1 , X_2 , X_3 = principal axes of the crystal n_1 , n_2 , n_3 = principal refractive indices

Figure adapted from Saleh/Teich, Fundamentals of Photonics

Wave propagation in anisotropic crystals





Propagation along a principal axis – arbitrary polarization direction:



Decomposition into a superposition of two linearly polarized modes; difference in wavenumber k leads to phase delay between the two polarizations,

$$\Delta \Phi = -k_0 \left(n_2 - n_1 \right) z$$

Figures adapted from Saleh/Teich, Fundamentals of Photonics

 n_2

nı

Wave propagation in anisotropic crystals



Propagation in arbitrary direction – fundamental properties:

Isotropic media $\underbrace{\mathbf{H}}_{\mathbf{E}} \parallel \underbrace{\mathbf{B}}_{\mathbf{D}}$

<u>k</u>, <u>**E**</u>, <u>**H**</u> are mutually orthogonal and form a right-handed set

 $\underline{k} \parallel \underline{S}$

 $\begin{array}{c}
\underline{\mathbf{H}} \parallel \underline{\mathbf{B}} \\
\underline{\mathbf{E}} \parallel \underline{\mathbf{D}} \quad \underline{\mathbf{D}} = \epsilon_0 \underline{\epsilon}_r \underline{\mathbf{E}}, \\
\underline{\mathbf{E}} = \frac{1}{\epsilon_0} \underline{\eta} \underline{\mathbf{D}}.
\end{array}$ $\underline{\mathbf{k}}, \underline{\mathbf{D}}, \underline{\mathbf{H}} \text{ are mutually}$

orthogonal and form a right-handed set

Anisotropic media



Figure adapted from Saleh/Teich, Fundamentals of Photonics



Normal modes for propagation in arbitrary direction



Plane-wave ansatz for the fields:

$$\underline{\mathbf{E}} = \underline{\mathbf{E}}_0 e^{-\mathbf{j}\mathbf{k}\mathbf{r}} \qquad \underline{\mathbf{H}} = \underline{\mathbf{H}}_0 e^{-\mathbf{j}\mathbf{k}\mathbf{r}} \qquad \underline{\mathbf{D}} = \underline{\mathbf{D}}_0 e^{-\mathbf{j}\mathbf{k}\mathbf{r}}.$$

Insert into Maxwell's equations:

$$\begin{aligned} -j\mathbf{k} \times \underline{\mathbf{E}} &= -j\,\omega\mu_0\underline{\mathbf{H}}, \qquad \underline{\mathbf{E}} = \frac{1}{\epsilon_0}\underline{\eta}\underline{\mathbf{D}}. \\ -j\mathbf{k} \times \underline{\mathbf{H}} &= j\,\omega\underline{\mathbf{D}}. \qquad \text{Note: } \mathbf{A} \times \mathbf{B} \times \mathbf{C} = \mathbf{B}\left(\mathbf{A}^{\mathrm{T}}\mathbf{C}\right) - \mathbf{C}\left(\mathbf{A}^{\mathrm{T}}\mathbf{B}\right). \end{aligned}$$

Wave equation for **D**:

$$-\mathbf{k} \times \left(\mathbf{k} \times \left(\underline{\eta}\underline{\mathbf{D}}\right)\right) = k_0^2 \underline{\mathbf{D}}$$

Transform to implicit equation that relates the dielectric displacement vector \underline{D} to the corresponding propagation constant *k*:

$$\underline{\mathbf{D}}^T \underline{\boldsymbol{\eta}} \underline{\mathbf{D}} = \frac{k_0^2}{\mathbf{k}^T \mathbf{k}} \underline{\mathbf{D}}^T \underline{\mathbf{D}}$$

$$\frac{X^2}{n_1^2} + \frac{Y^2}{n_2^2} + \frac{Z^2}{n_3^2} = 1$$

Index ellipsoid / optical indicatrix

where
$$X = \frac{k}{k_0} \frac{D_x}{D}$$
,
rix $D = \sqrt{\mathbf{D}^T \mathbf{D}}$

$$k = \sqrt{\mathbf{k}^T \mathbf{k}}$$

 $\frac{D_x}{D}, \qquad Y = \frac{k}{k_0} \frac{D_y}{D}, \qquad Z = \frac{k}{k_0} \frac{D_z}{D},$

123 27.06.2018 Christian Koos

Determining normal modes from the index ellipsoid





Figure adapted from Saleh/Teich, Fundamentals of Photonics

Given: Propagation direction of the optical wave,

$$\mathbf{u} = \frac{\mathbf{k}}{|\mathbf{k}|}$$

Wanted: Effective refractive indices n_a and n_b of the normal modes and corresponding displacement vectors $\underline{\mathbf{D}}_a$ and $\underline{\mathbf{D}}_b$ (or their normalized counterparts $(X, Y, Z)^T$)

Solution:

- Draw a plane passing through the origin of the index ellipsoid, normal to **u**. The intersection of the plane with the ellipsoid produces the index ellipse.
- The half-lengths of the major and minor axes of the index ellipse are the refractive indexes n_a and n_b of the two normal modes, that propagate like plane waves (without derivation).
- The directions of the major and minor axes of the index ellipse are the directions of the vectors <u>D</u>_a and <u>D</u>_b for the normal modes. These directions are orthogonal.
- The vectors $\underline{\mathbf{E}}_{a}$ and $\underline{\mathbf{E}}_{b}$ are derived from $\underline{\mathbf{D}}_{a}$ and $\underline{\mathbf{D}}_{b}$

$$\underline{\mathbf{D}} = \epsilon_0 \underline{\epsilon}_r \underline{\mathbf{E}}, \qquad \underline{\mathbf{E}} = \frac{1}{\epsilon_0} \underline{\eta} \underline{\mathbf{D}}.$$

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Uniaxial crystals



 $n_1 = n_2 = n_0$ and $n_3 = n_e$

 \Rightarrow Index ellipsoid is rotationally symmetric with respect to the Z-axis



Normal modes:

- Ordinary mode: n_a = n_o
- Extraordinary mode: $n_b = n (\theta)$, where

$$\frac{1}{n^2(\theta)} = \frac{\sin^2(\theta)}{n_e^2} + \frac{\cos^2(\theta)}{n_o^2}$$

• Walk-off angle ρ between the electric field $\underline{\mathbf{E}}_{e}$ and the dielectric displacement $\underline{\mathbf{D}}_{e}$: $\cos(\rho) = \frac{\underline{\mathbf{E}}_{b}^{T}\underline{\mathbf{D}}_{b}}{|\underline{\mathbf{E}}_{b}| |\underline{\mathbf{D}}_{b}|} = \frac{n_{e}^{2}\cos^{2}(\theta) + n_{o}^{2}\sin^{2}(\theta)}{\sqrt{n_{e}^{4}\cos^{2}(\theta) + n_{o}^{4}\sin^{2}(\theta)}}$

Lecture 8

Wave propagation in anisotropic crystals





Propagation along a principal axis – arbitrary polarization direction:



Decomposition into a superposition of two linearly polarized modes; difference in wavenumber k leads to phase delay between the two polarizations,

$$\Delta \Phi = -k_0 \left(n_2 - n_1 \right) z$$

Figures adapted from Saleh/Teich, Fundamentals of Photonics

 n_2

nı

Determining normal modes from the index ellipsoid





Figure adapted from Saleh/Teich, Fundamentals of Photonics Given: Propagation direction of the optical wave,

$$\mathbf{u} = \frac{\mathbf{k}}{|\mathbf{k}|}$$

Wanted: Effective refractive indices n_a and n_b of the normal modes and corresponding displacement vectors \underline{D}_a and \underline{D}_b

Solution:

- Draw a plane passing through the origin of the index ellipsoid, normal to u. The intersection of the plane with the ellipsoid produces the index ellipse.
- The half-lengths of the major and minor axes of the index ellipse are the refractive indexes n_a and n_b of the two normal modes, that propagate like plane waves (without derivation).
- The directions of the major and minor axes of the index ellipse are the directions of the vectors \underline{D}_a and \underline{D}_b for the normal modes. These directions are orthogonal.
- The vectors \underline{E}_a and \underline{E}_b are derived from \underline{D}_a and \underline{D}_b

$$\underline{\mathbf{D}} = \epsilon_0 \underline{\epsilon}_r \underline{\mathbf{E}}, \qquad \underline{\mathbf{E}} = \frac{1}{\epsilon_0} \underline{\eta} \underline{\mathbf{D}}.$$



Uniaxial crystals



 $n_1 = n_2 = n_0$ and $n_3 = n_e$

 \Rightarrow Index ellipsoid is rotationally symmetric with respect to the Z-axis



Normal modes:

- Ordinary mode: n_a = n_o
- Extraordinary mode: $n_b = n(\theta)$, where

$$\frac{1}{n^2(\theta)} = \frac{\sin^2(\theta)}{n_e^2} + \frac{\cos^2(\theta)}{n_o^2}$$

• Walk-off angle ρ between the electric field <u>**E**</u>_e and the dielectric displacement <u>**D**</u>_e: $\cos(\rho) = \frac{\underline{\mathbf{E}}_b^T \underline{\mathbf{D}}_b}{|\mathbf{E}| + |\mathbf{D}|}$

$$= \frac{\underline{\mathbf{E}}_{b}^{T} \underline{\mathbf{D}}_{b}}{|\underline{\mathbf{E}}_{b}| |\underline{\mathbf{D}}_{b}|} = \frac{n_{e}^{2} \cos^{2}(\theta) + n_{o}^{2} \sin^{2}(\theta)}{\sqrt{n_{e}^{4} \cos^{2}(\theta) + n_{o}^{4} \sin^{2}(\theta)}}$$

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Consider gradient of implicit representation of index ellipsoid:

$$\mathbf{n} = \operatorname{grad}\left(\frac{X^2}{n_1^2} + \frac{Y^2}{n_2^2} + \frac{Z^2}{n_3^2}\right) = 2\left(\frac{\frac{X}{n_1^2}}{\frac{Y}{n_2^2}}{\frac{Z}{n_3^2}}\right) = \frac{2k}{k_0 D}\left(\frac{\frac{D_x}{n_1^2}}{\frac{D_y}{n_2^2}}{\frac{D_z}{n_3^2}}\right) = \frac{2k\epsilon_0}{k_0 D}\underline{E},$$

=> Surface normal of the index ellipsoid defines direction of the electric field \underline{E}



Note: For uniaxial crystals, E || D for ordinary modes, but not for extraordinary modes

⇒ For a given direction of **k**, the walk-off angle ρ between E and D corresponds also to the walk-off between the Poynting vector of the ordinary and the extraordinary beam



Linear electro-optic effect / Pockels effect



Index ellipsoid in coordinate system of crystal unit cell:

Nonlinear interaction: Express elements of the impermeability tensor as a power series in the strength of the external electric components E_k

$$\eta_{ij} = \eta_{ij}^{(0)} + \sum_{k} r_{ijk} E_k + \sum_{k,l} s_{ijkl} E_k E_l + \dots$$

Using contracted notation, the third-rank electro-optic tensor r_{ijk} can be expressed as a twodimensional (6×3)-matrix r_{hk} :

$$\begin{pmatrix} \Delta (1/n^2)_1 \\ \Delta (1/n^2)_2 \\ \Delta (1/n^2)_3 \\ \Delta (1/n^2)_4 \\ \Delta (1/n^2)_5 \\ \Delta (1/n^2)_6 \end{pmatrix} = \begin{pmatrix} r_{11} r_{12} r_{13} \\ r_{21} r_{22} r_{23} \\ r_{31} r_{32} r_{33} \\ r_{41} r_{42} r_{43} \\ r_{51} r_{52} r_{53} \\ r_{61} r_{62} r_{63} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}.$$



Electro-optic tensor for different crystal classes



is restricted by the symmetry of the underlying crystal lattice:			Material	Point Group	Electrooptic Coefficients (10 ⁻¹² m/V)	Refractive Index
$r_{ij} = \begin{bmatrix} 0\\0\\r_{41}\\0 \end{bmatrix}$		(for class $\overline{4}2m$),	Potassium dihydrogen phosphate, KH ₂ PO ₄ (KDP)	42 <i>m</i>	$r_{41} = 8.77$ $r_{63} = 10.5$	$n_0 = 1.514$ $n_e = 1.472$ (at 0.5461 μ m)
	$ \begin{array}{ccc} 0 & 0 \\ 0 & 0 \\ r_{41} & 0 \end{array} $		Potassium dideuterium phosphate, KD ₂ PO ₄ (KD*P)	42 <i>m</i>	$r_{41} = 8.8$ $r_{63} = 26.4$	$n_0 = 1.508$ $n_e = 1.468$ (at 0.5461 μ m)
L o F o	$0 r_{63}$		Lithium niobate, LiNbO3	3m	$r_{13} = 9.6$ $r_{22} = 6.8$ $r_{33} = 30.9$ $r_{42} = 32.6$	$n_0 = 2.3410$ $n_e = 2.2457$ (at 0.5 μ m)
$r_{ij} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ r_{42} \\ r_{22} \end{bmatrix}$	$\begin{array}{ccc} r_{22} & r_{13} \\ 0 & r_{33} \\ r_{42} & 0 \\ 0 & 0 \end{array}$	(for class 3 <i>m</i>),	Lithium tantalate, LiTaO3	3m	$r_{13} = 8.4$ $r_{22} = -0.2$ $r_{33} = 30.5$ $r_{51} = 20$	$n_0 = 2.176$ $n_e = 2.180$ (at 0.633 nm)
	0 0		Barium titanate, BaTiO ₃ ^b	4 <i>mm</i>	$r_{13} = 19.5$ $r_{33} = 97$ $r_{42} = 1640$	$n_0 = 2.488$ $n_e = 2.424$ (at 514 nm)
$r_{ij} = \begin{bmatrix} 0\\0\\0\\0 \end{bmatrix}$	$\begin{array}{ccc} 0 & r_{13} \\ 0 & r_{13} \end{array}$		Strontium barium niobate, Sr _{0.6} Ba _{0.4} NbO ₆ (SBN:60)	4 <i>mm</i>	$r_{13} = 55$ $r_{33} = 224$ $r_{42} = 80$	$n_0 = 2.367$ $n_e = 2.337$ (at 514 nm)
	$\begin{array}{ccc} 0 & r_{33} \\ r_{42} & 0 \end{array}$	(for class 4mm).	Zinc telluride, ZnTe	43 <i>m</i>	$r_{41} = 4.0$	$n_0 = 2.99$ (at 0.633 μ m)
$r_{ij} = \begin{bmatrix} 0\\0\\0\\r_{42}\\0 \end{bmatrix}$	$\begin{bmatrix} 0 & r_{13} \\ 0 & r_{13} \\ 0 & r_{33} \\ r_{42} & 0 \\ 0 & 0 \\ 0 & 0 \end{bmatrix}$	(for class 4mm).	Strontium barium niobate, Sr _{0.6} Ba _{0.4} NbO ₆ (SBN:60) Zinc telluride, ZnTe	4mm 43m r example, Thompso	$r_{33} = 97$ $r_{42} = 1640$ $r_{13} = 55$ $r_{33} = 224$ $r_{42} = 80$ $r_{41} = 4.0$ on and Hartfield (1978)	$n_e = 2.4$ (at 514 n $n_0 = 2.3$) $n_e = 2.3$ (at 514 n $n_0 = 2.9$ (at 0.633)) and Cook a

Figures adapted from Boyd, Nonlinear Optics

Form of the electro-ontic tensor r

^{*a*} From a variety of sources. See, for example, Thompson and Hartfield (1978) and Cook and Jaffe (1979). The electrooptic coefficients are given in the MKS units of m/V. To convert to the cgs units of cm/statvolt each entry should be multiplied by 3×10^4 .

$$\epsilon_{dc}^{\parallel} = 135, \epsilon_{dc}^{\perp} = 3700.$$

Electro-optic modulators



Electro-optic modulators: Exploit second-order nonlinearities to modulate a beam of light by means of an electric signal.





Longitudinal modulator: The Pockels cell





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Applied modulating field E_z transforms uniaxial crystal into a biaxial medium:

$$\eta (E_z) = \begin{pmatrix} \frac{1}{n_o^2} & r_{63}E_z & 0\\ r_{63}E_z & \frac{1}{n_o^2} & 0\\ 0 & 0 & \frac{1}{n_e^2} \end{pmatrix}$$

Transform indicatrix to new principal axes (X', Y', Z'):

$$X = \frac{1}{\sqrt{2}} \left(X' + Y' \right) \qquad n_{X'} = n_o \left(1 + \frac{1}{2} r_{63} n_o^2 E_z \right) \\ Y = \frac{1}{\sqrt{2}} \left(-X' + Y' \right) \qquad n_{Y'} = n_o \left(1 - \frac{1}{2} r_{63} n_o^2 E_z \right) \\ Z = Z' \qquad n_{Z'} = n_e$$

⇒ Normal modes are polarized at 45° with respect to x and y and experience a phase delay with respect to each other \vec{E}_x

during propagation:

$$\Delta \Phi = \pi \frac{U}{U_{\pi}}$$
$$U_{\pi} = \frac{\lambda}{2r_{63}n_o^3}$$



Deformation of indicatrix by external electric field







Turning the Pockels into an amplitude modulator



Insert polarizer after the Pockels cell:





$$T(U) = \sin^2\left(\frac{\pi}{2}\frac{U}{U_{\pi}}\right)$$

Adjust operating point by quarter-wave plate:





Transverse modulator





The workhorse of optical communications: Integrated $LiNbO_3$ modulator





Proton-exchanged (PE) waveguides



Note: Diffusion length along y-direction much larger (and much more uncertain) than along *x*- and *z*-direction

 \Rightarrow Waveguides usually oriented along *y*-direction!



Analysis of a transverse LiNbO₃ modulator





External voltage along z: Material remains uniaxial!

$$\left(\frac{1}{n_o^2} + r_{13}E_z\right)X^2 + \left(\frac{1}{n_o^2} + r_{13}E_z\right)Y^2 + \left(\frac{1}{n_c^2} + r_{33}E_z\right)Z^2 = 1$$

 \Rightarrow Refractive index change for plane wave polarized along z / phase shift / π -voltage:

$$\Delta n = -\frac{1}{2}n_e^3 r_{33}E_z \qquad \Delta \Phi = \frac{1}{2}n_e^3 r_{33}E_z k_0 L \qquad U_\pi = \frac{d}{L}\frac{\lambda_0}{r_{33}n_e^3}$$



Lecture 9

Electro-optic modulators



Electro-optic modulators: Exploit second-order nonlinearities to modulate a beam of light by means of an electric signal.





Analysis of a transverse LiNbO₃ modulator





External voltage along z: Material remains uniaxial!

$$\left(\frac{1}{n_o^2} + r_{13}E_z\right)X^2 + \left(\frac{1}{n_o^2} + r_{13}E_z\right)Y^2 + \left(\frac{1}{n_c^2} + r_{33}E_z\right)Z^2 = 1$$

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143 27.06.2018 Christian Koos



Technical realization: *x*-cut and *z*-cut geometry



z-cut geometry:



x-cut geometry:



- Wafer surface normal to z-direction
- Modulating field oriented vertically (along z-direction)
- Waveguide oriented along ydirection
- "TM-polarization", i.e., dominant electric field component of the optical mode oriented along zdirection
- Wafer surface normal to x-direction
- Modulating field oriented horizontally (along z-direction)
- Waveguide oriented along ydirection
- "TE-polarization", i.e., dominant electric field component of the optical mode oriented along zdirection


The Mach-Zehnder Modulator (MZM)



So far: Phase modulator



Turning a phase modulator into an amplitude modulator: Mach-Zehnder Modulator (MZM)



Here: Push-pull configuration, i.e., antisymmetric phase shifts in both arms of the interferometer (same magnitude, opposite sign)

 \Rightarrow Chirp-free operation

Figures adapted from Sumitomo Modulator Application Note.



Dual-drive Mach-Zehnder Modulator



Individual transmission line for each modulator arm \Rightarrow Dual-drive / dual-electrode configuration:



Figures adapted from Sumitomo Modulator Application Note.

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Combine push-pull modulator with coplanar transmission line:



z-cut:

- Electrodes placed on top of the waveguide
- Electric field flux concentrated for center electrode ("hot electrode")
- \Rightarrow Good overlap between optical field
- er an RF field; high efficiency
 - Reduced overlap for ground electrode
 - ⇒ Asymmetric modulation; slightly chirped output signal



x-cut:

- Slightly decreased efficiency, but equal overlap for both arms
- ⇒ Antisymmetric modulation; Layer unchirped output signal

Figures adapted from Sumitomo Modulator Application Note.



Two more examples...





z-cut dual-drive modulator:

- Good overlap of modulating RFfield and optical field
- \Rightarrow Low switching voltage, good electro-optic efficiency
- Push-pull-operation requires two RF-signals of identical amplitude but opposite sign



x-cut push-pull modulator:

- Push-pull operation with a single RF signal
- \Rightarrow Chirp-free modulation
- Needs approx. 20% higher voltage as compared to z-cut configuration

Ed L. Wooten et al., IEEE J. Sel. Topics Quant. Electron. 6, 69-82 (2000)



Ongoing research at IPQ: Silicon-based modulators



Silicon-organic hybrid (SOH) device: Silicon slot waveguide + organic electro-optic cladding



- Good overlap between optical field and RF field
- Small electrode spacing

Push-pull configuration:



SOH Mach-Zehnder Modulators: Implementation





- Basic silicon photonic waveguide structures fabricated in standard silicon photonic line along with full portfolio of other devices (pn modulators, SiGe detectors *etc.*)
 ⇒ High integration density, high yield
- Organic EO materials deposited and poled in a post-processing step ⇒ No compatibility issues, efficient large-area processing



SOH Mach-Zehnder Modulators: Implementation







SOH Mach-Zehnder Modulators: Implementation





Chirp-free push-pull operation!

Koos *et al.*, J. Lightw. Technol. **34**, 256-268 (2016) Koeber *et al.*, Light Sci Appl **4**, (2015)



Finally: A real device ...







Sum-frequency generation and impact of phase mismatch



Recall: Evolution of complex field amplitudes during propagation through a nonlinear medium (SVEA)

$$\frac{\partial \underline{E}'(z',t',\omega_l)}{\partial z'} = -j \frac{\omega_l}{2\epsilon_0 cn} \underline{P}'_{\mathsf{NL}}(z',t',\omega_l) e^{-j(k_{p,l}-k_l)z'}.$$

Now: Investigate impact of phase mismatch on interaction of three waves oscillating at frequencies ω_1 , ω_2 , and $\omega_3 = \omega_1 + \omega_2$.

Assume fixed polarizations along unit vectors **e**_i:

$$\underline{\mathbf{E}}(z,t,\omega_{i}) = \underline{\mathbf{E}}(z,t,\omega_{i}) \mathbf{e}_{i}, \quad \underline{\mathbf{P}}(z,t,\omega_{3}) = \underline{\mathbf{P}}(z,t,\omega_{3}) \mathbf{e}_{3},$$

$$\Rightarrow \frac{\partial \underline{\mathbf{E}}(z,t,\omega_{3})}{\partial z} = -\mathbf{j}\frac{\omega_{3}}{cn(\omega_{3})}d_{\text{eff}}\underline{\mathbf{E}}(z,t,\omega_{1})\underline{\mathbf{E}}(z,t,\omega_{2})e^{-\mathbf{j}\Delta kz},$$

$$\frac{\partial \underline{\mathbf{E}}(z,t,\omega_{1})}{\partial z} = -\mathbf{j}\frac{\omega_{1}}{cn(\omega_{1})}d_{\text{eff}}\underline{\mathbf{E}}(z,t,\omega_{3})\underline{\mathbf{E}}^{*}(z,t,\omega_{2})e^{\mathbf{j}\Delta kz},$$

$$\frac{\partial \underline{\mathbf{E}}(z,t,\omega_{2})}{\partial z} = -\mathbf{j}\frac{\omega_{2}}{cn(\omega_{2})}d_{\text{eff}}\underline{\mathbf{E}}(z,t,\omega_{3})\underline{\mathbf{E}}^{*}(z,t,\omega_{1})e^{\mathbf{j}\Delta kz}.$$
where $\Delta k = k_{1} + k_{2} - k_{3}$ Wave vector mismatch
$$d_{\text{eff}} = \frac{1}{2}\sum_{q,r,s}\mathbf{e}_{3,q}\underline{\chi}_{q;r,s}^{(2)}(\omega_{3}:\omega_{1},\omega_{2})\mathbf{e}_{1,r}\mathbf{e}_{2,s}$$
Effective 2nd-order nonlinear susceptibility





Zero phase mismatch: Linear increase of converted amplitude (depletion at ω_1 and ω_2 neglected): $\underline{E}(L,t,\omega_{3})|_{\Delta k=0} \approx -j \frac{\omega_{3}}{cn(\omega_{3})} d_{\text{eff}} \underline{E}(0,t,\omega_{1}) \underline{E}(0,t,\omega_{2}) L$

Non-zero phase mismatch: Oscillatory behavior

$$\underline{E}(L,t,\omega_3)|_{\Delta k\neq 0} = -j\frac{\omega_3}{cn(\omega_3)}d_{\text{eff}}\underline{E}(0,t,\omega_1)\underline{E}(0,t,\omega_2)\frac{2}{\Delta k}\sin\left(\frac{\Delta kL}{2}\right)e^{-j\frac{\Delta kL}{2}}.$$

 \Rightarrow Considerable decrease of the power conversion efficiency:



Lecture 10

Sum-frequency generation and impact of phase mismatch



Recall: Evolution of complex field amplitudes during propagation through a nonlinear medium (SVEA)

$$\frac{\partial \underline{E}'(z',t',\omega_l)}{\partial z'} = -j \frac{\omega_l}{2\epsilon_0 cn} \underline{P}'_{\mathsf{NL}}(z',t',\omega_l) e^{-j(k_{p,l}-k_l)z'}.$$

Now: Investigate impact of phase mismatch on interaction of three waves oscillating at frequencies ω_1 , ω_2 , and $\omega_3 = \omega_1 + \omega_2$.

Assume fixed polarization directions along unit vectors **e**_i:

$$\underline{\mathbf{E}}(z,t,\omega_{i}) = \underline{\mathbf{E}}(z,t,\omega_{i}) \mathbf{e}_{i}, \quad \underline{\mathbf{P}}(z,t,\omega_{3}) = \underline{\mathbf{P}}(z,t,\omega_{3}) \mathbf{e}_{3},$$

$$\Rightarrow \frac{\partial \underline{\mathbf{E}}(z,t,\omega_{3})}{\partial z} = -\mathbf{j}\frac{\omega_{3}}{cn(\omega_{3})}d_{\text{eff}}\underline{\mathbf{E}}(z,t,\omega_{1})\underline{\mathbf{E}}(z,t,\omega_{2})e^{-\mathbf{j}\Delta kz},$$

$$\frac{\partial \underline{\mathbf{E}}(z,t,\omega_{1})}{\partial z} = -\mathbf{j}\frac{\omega_{1}}{cn(\omega_{1})}d_{\text{eff}}\underline{\mathbf{E}}(z,t,\omega_{3})\underline{\mathbf{E}}^{*}(z,t,\omega_{2})e^{\mathbf{j}\Delta kz},$$

$$\frac{\partial \underline{\mathbf{E}}(z,t,\omega_{2})}{\partial z} = -\mathbf{j}\frac{\omega_{2}}{cn(\omega_{2})}d_{\text{eff}}\underline{\mathbf{E}}(z,t,\omega_{3})\underline{\mathbf{E}}^{*}(z,t,\omega_{1})e^{\mathbf{j}\Delta kz}.$$
where $\Delta k = k_{1} + k_{2} - k_{3}$ Wave vector mismatch
$$d_{\text{eff}} = \frac{1}{2}\sum_{q,r,s}\mathbf{e}_{3,q}\underline{\chi}_{q;r,s}^{(2)}(\omega_{3}:\omega_{1},\omega_{2})\mathbf{e}_{1,r}\mathbf{e}_{2,s}$$
Effective 2nd-order nonlinear susceptibility

157 27.06.2018 Christian Koos





Zero phase mismatch: Linear increase of converted amplitude (depletion at ω_1 and ω_2 neglected): $\underline{E}(L,t,\omega_{3})|_{\Delta k=0} \approx -j \frac{\omega_{3}}{cn(\omega_{3})} d_{\text{eff}} \underline{E}(0,t,\omega_{1}) \underline{E}(0,t,\omega_{2}) L$

Non-zero phase mismatch: Oscillatory behavior

$$\underline{E}(L,t,\omega_3)|_{\Delta k\neq 0} = -j\frac{\omega_3}{cn(\omega_3)}d_{\text{eff}}\underline{E}(0,t,\omega_1)\underline{E}(0,t,\omega_2)\frac{2}{\Delta k}\sin\left(\frac{\Delta kL}{2}\right)e^{-j\frac{\Delta kL}{2}}.$$

 \Rightarrow Considerable decrease of the power conversion efficiency:



Phase matching concepts



Phase matching conditions:

SFG:
$$\omega_1 n(\omega_1) + \omega_2 n(\omega_2) - \omega_3 n(\omega_3) = 0$$

SHG: $n(\omega_1) = n(2\omega_1)$

Mostly: Materials operated below resonance frequencies

 \Rightarrow Normal (phase velocity) dispersion, i.e., refractive index increases with frequency

 \Rightarrow Phase matching conditions cannot be fulfilled.

Idea: Exploit birefringence for phase matching!





Type-1 phase matching in negative-uniaxial crystal



Type-1 phase matching: Lower-frequency components have the same polarization

Second-harmonic generation (SHG): $n_e(2\omega_1) = n_o(\omega_1)$ [$oo \rightarrow e$]



Sum-frequency generation (SFG): (1207)

$$\omega_{3}n_{e}(\omega_{3}) = \omega_{1}n_{o}(\omega_{1}) + \omega_{2}n_{o}(\omega_{2}) \qquad [oo \to e]$$

Figures adapted from Stegeman, Nonlinear Optics and Saleh-Teich, Fundamentals of Photonics

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Sum-frequency generation (SFG):

 $\omega_{3}n_{o}(\omega_{3}) = \omega_{1}n_{e}(\omega_{1}) + \omega_{2}n_{e}(\omega_{2}) \qquad [ee \to o]$

Figures adapted from Stegeman, Nonlinear Optics and Saleh-Teich, Fundamentals of Photonics

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Type-1 phase matching: Tuning options



Temperature tuning:

- Birefringence is temperature-dependent
 Phase-matching by varying the temperature of the crystal!
- Example LiNbO₃ : Strong temperature dependence of birefringence.

Angle tuning:

- Adjust propagation direction of the involved light beams to obtain phase matching
- Example: Second-harmonic generation in a positive-uniaxial crystal





Figures adapted from Stegeman, Nonlinear Optics and Saleh-Teich, Fundamentals of Photonics



Type-1 phase matching: Calculation of propagation angle



Example: Type-1 phase matching for second-harmonic $[00] \rightarrow e$ generation in a negative-uniaxial crystal $n_e(2\omega_1,\Theta_n)=n_o(\omega_1)$ $n(\omega)$ $n_e(\theta)$ $\Rightarrow \tan \Theta_p = \frac{n_e (2\omega_1)}{n_o (2\omega_1)} \sqrt{\frac{n_o^2 (2\omega_1) - n_o^2 (\omega_1)}{n_o^2 (\omega_1) - n_o^2 (2\omega_1)}}.$ n_o ne Problem: Walk-off between ordinary and extraordinary ray 2ω ω Recall: Poynting vector and wave vector of the extraordinary ray are not parallel, and hence the Poynting vector of z_{∎n_e} the fundamental and the second harmonic are not parallel! Extraordinary $D(2\omega_1)$ Θ wave $D(\omega$ n k! H,B $n_{\rm o}$ y $\cos\left(\rho\right) = \frac{n_e^2\left(2\omega_1\right)\cos^2\left(\Theta_p\right) + n_o^2\left(2\omega_1\right)\sin^2\left(\Theta_p\right)}{\sqrt{n_e^4\left(2\omega_1\right)\cos^2\left(\Theta_p\right) + n_o^4\left(2\omega_1\right)\sin^2\left(\Theta_p\right)}}$ Figures adapted from Stegeman, Nonlinear Optics and Saleh-Teich, Fundamentals of Photonics

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Х

Type-2 phase matching



For strong birefringence: Type-1 phase-matching would overcompensate phase velocity mismatch and require large propagation angles

Type-2 phase matching: Lower-frequency components have different polarizations



Quasi-phase-matching



In some cases we cannot use birefringence to achieve phase matching:

• Nonlinear material is not birefringent



• Birefringence too weak / dispersion too strong, e.g., at short wavelengths



• Exploit strong d₃₃ coefficient; this requires waves that are polarized in the same direction

LiNbO₃: $d_{22} \approx 3pm/V$ $d_{31} \approx -5 pm/V$ $d_{33} \approx -25 pm/V$

frequency, ω Li Nb O

Figures adapted from Boyd, Nonlinear Optics

The principle of quasi-phase-matching (QPM)



Re

 $\Delta kz = 0$

Problem of phase mismatch: Continuous change of phase of new contributions along z.

$$\frac{\partial \underline{E}(z,t,\omega_{3})}{\partial z} = -j \frac{\omega_{3}}{cn(\omega_{3})} d_{eff} \underline{E}(z,t,\omega_{1}) \underline{E}(z,t,\omega_{2}) e^{-j\Delta kz},$$
Phase matching: $\Delta k = 0$
Phase mismatch: $\Delta k \neq 0$
Phase mismatch:

Quasi-phase-matching: Periodic sign reversal of the second-order nonlinearity d_{eff} , whenever a phase shift of π is accumulated



 $\Delta kz = 2\pi'$

Quasi-phase-matching (QPM)









Evolution of converted wave amplitude $E(z,t,\omega)$ along z:



Figure adapted from Boyd, Nonlinear Optics

168 27.06.2018 Christian Koos

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Evolution of converted wave along z:

$$\frac{\partial \underline{E}(z,t,\omega_{3})}{\partial z} = -j \frac{\omega_{3}}{cn(\omega_{3})} d_{\text{eff}} \underline{E}(z,t,\omega_{1}) \underline{E}(z,t,\omega_{2}) e^{-j\Delta kz},$$

Introduce periodic effective second-order nonlinearity: $d_{eff}(z + \Lambda) = d_{eff}(z)$

$$d_{\text{eff}}(z) = \sum_{m} d_{m} e^{jm\frac{2\pi}{\Lambda}z},$$

$$\frac{\partial \underline{E}(z,t,\omega_{3})}{\partial z} = -j \frac{\omega_{3}}{cn(\omega_{3})} \sum_{m} d_{m} \underline{E}(z,t,\omega_{1}) \underline{E}(z,t,\omega_{2}) e^{-j\left(\Delta k - m\frac{2\pi}{\Lambda}\right)z},$$

Phase-matching by first-order interaction (m=1): $\Lambda = \frac{2\pi}{\Delta k}$,



Figure adapted from Saleh-Teich, Fundamentals of Photonics



Technology: Periodically poled lithium niobate (PPLN)



Principle:

- Lithium Niobate is a ferroelectric crystal, i.e., each unit cell in the crystal has a small electric dipole moment, depending on the positions of the niobium and lithium atoms in the unit cell.
- A strong electric field (~ 22 kV/mm) can locally invert the crystal structure and flip the orientation of the dipole moment and of the second-order nonlinear susceptibility tensor.

Fabrication of PPLN:



Figure adapted from Thorlabs, Tutorial on Periodically Poled Lithium Niobate (PPLN), www.thorlabs.com

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Consider interaction of 3 waves oscillating at frequencies ω_1, ω_2 , and $\omega_3 = \omega_1 + \omega_2$: $\frac{\partial \underline{E}(z, t, \omega_3)}{\partial z} = -j \frac{\omega_3}{cn(\omega_3)} d_{\text{eff}} \underline{E}(z, t, \omega_1) \underline{E}(z, t, \omega_2) e^{-j\Delta kz},$ $\frac{\partial \underline{E}(z, t, \omega_1)}{\partial z} = -j \frac{\omega_1}{cn(\omega_1)} d_{\text{eff}} \underline{E}(z, t, \omega_3) \underline{E}^*(z, t, \omega_2) e^{j\Delta kz},$ $\frac{\partial \underline{E}(z, t, \omega_2)}{\partial z} = -j \frac{\omega_2}{cn(\omega_2)} d_{\text{eff}} \underline{E}(z, t, \omega_3) \underline{E}^*(z, t, \omega_1) e^{j\Delta kz}.$

Intensity:
$$I(z,t,\omega_i) = \frac{1}{2} \epsilon_0 cn(\omega_i) |\underline{E}(z,t,\omega_i)|^2$$
.

 \Rightarrow Evolution of intensities along z:

$$\frac{\partial I(z,t,\omega_{3})}{\partial z} = -\epsilon_{0}\omega_{3}d_{\text{eff}} \operatorname{Im}\left\{\underline{E}^{*}(z,t,\omega_{1})\underline{E}^{*}(z,t,\omega_{2})\underline{E}(z,t,\omega_{3})e^{j\Delta kz}\right\}$$
$$\frac{\partial I(z,t,\omega_{2})}{\partial z} = \epsilon_{0}\omega_{2}d_{\text{eff}} \operatorname{Im}\left\{\underline{E}^{*}(z,t,\omega_{1})\underline{E}^{*}(z,t,\omega_{2})\underline{E}(z,t,\omega_{3})e^{j\Delta kz}\right\}$$
$$\frac{\partial I(z,t,\omega_{1})}{\partial z} = \epsilon_{0}\omega_{1}d_{\text{eff}} \operatorname{Im}\left\{\underline{E}^{*}(z,t,\omega_{1})\underline{E}^{*}(z,t,\omega_{2})\underline{E}(z,t,\omega_{3})e^{j\Delta kz}\right\}$$



The Manley-Rowe relations



Conclusions:

• Total intensity does not change along z

$$\frac{\partial}{\partial z}\left(I\left(z,t,\omega_{1}\right)+I\left(z,t,\omega_{2}\right)+I\left(z,t,\omega_{3}\right)\right)=0.$$

• Change of photon fluxes are connected:

$$\frac{\partial}{\partial z} \left(\frac{I(z, t, \omega_1)}{\hbar \omega_1} \right) = \frac{\partial}{\partial z} \left(\frac{I(z, t, \omega_2)}{\hbar \omega_2} \right) = -\frac{\partial}{\partial z} \left(\frac{I(z, t, \omega_3)}{\hbar \omega_3} \right)$$

⇒ Generation of a photon at ω_1 is always accompanied by generation of a photon at ω_2 and annihilation of a photon at ω_3 (and vice versa)





Lecture 11



Consider interaction of 3 waves oscillating at frequencies ω_1, ω_2 , and $\omega_3 = \omega_1 + \omega_2$: $\frac{\partial \underline{E}(z, t, \omega_3)}{\partial z} = -j \frac{\omega_3}{cn(\omega_3)} d_{\text{eff}} \underline{E}(z, t, \omega_1) \underline{E}(z, t, \omega_2) e^{-j\Delta kz},$ $\frac{\partial \underline{E}(z, t, \omega_1)}{\partial z} = -j \frac{\omega_1}{cn(\omega_1)} d_{\text{eff}} \underline{E}(z, t, \omega_3) \underline{E}^*(z, t, \omega_2) e^{j\Delta kz},$ $\frac{\partial \underline{E}(z, t, \omega_2)}{\partial z} = -j \frac{\omega_2}{cn(\omega_2)} d_{\text{eff}} \underline{E}(z, t, \omega_3) \underline{E}^*(z, t, \omega_1) e^{j\Delta kz}.$

Intensity:
$$I(z,t,\omega_i) = \frac{1}{2} \epsilon_0 cn(\omega_i) |\underline{E}(z,t,\omega_i)|^2$$
.

 \Rightarrow Evolution of intensities along z:

$$\frac{\partial I(z,t,\omega_{3})}{\partial z} = -\epsilon_{0}\omega_{3}d_{\text{eff}} \operatorname{Im}\left\{\underline{E}^{*}(z,t,\omega_{1})\underline{E}^{*}(z,t,\omega_{2})\underline{E}(z,t,\omega_{3})e^{j\Delta kz}\right\}$$
$$\frac{\partial I(z,t,\omega_{2})}{\partial z} = \epsilon_{0}\omega_{2}d_{\text{eff}} \operatorname{Im}\left\{\underline{E}^{*}(z,t,\omega_{1})\underline{E}^{*}(z,t,\omega_{2})\underline{E}(z,t,\omega_{3})e^{j\Delta kz}\right\}$$
$$\frac{\partial I(z,t,\omega_{1})}{\partial z} = \epsilon_{0}\omega_{1}d_{\text{eff}} \operatorname{Im}\left\{\underline{E}^{*}(z,t,\omega_{1})\underline{E}^{*}(z,t,\omega_{2})\underline{E}(z,t,\omega_{3})e^{j\Delta kz}\right\}$$



The Manley-Rowe relations



Conclusions:

• Total intensity does not change along z

$$\frac{\partial}{\partial z}\left(I\left(z,t,\omega_{1}\right)+I\left(z,t,\omega_{2}\right)+I\left(z,t,\omega_{3}\right)\right)=0.$$

• Change of photon fluxes are connected:

$$\frac{\partial}{\partial z} \left(\frac{I(z, t, \omega_1)}{\hbar \omega_1} \right) = \frac{\partial}{\partial z} \left(\frac{I(z, t, \omega_2)}{\hbar \omega_2} \right) = -\frac{\partial}{\partial z} \left(\frac{I(z, t, \omega_3)}{\hbar \omega_3} \right)$$

⇒ Generation of a photon at ω_1 is always accompanied by generation of a photon at ω_2 and annihilation of a photon at ω_3 (and vice versa)





Parametric amplification



Consider difference frequency generation with strong pump wave at $\omega_3 = \omega_1 + \omega_2$

Assumptions: - Phase matching: $\Delta k = 0$

- Strong pump at frequency ω_3
 - \Rightarrow Pump depletion can be neglected, $\underline{E}(z, t, \omega_3) = \underline{E}(0, t, \omega_3)$

Coupled differential equations:

$$\frac{\partial \underline{E}(z,t,\omega_{1})}{\partial z} = -j \frac{\omega_{1}}{cn(\omega_{1})} d_{\text{eff}} \underline{E}(0,t,\omega_{3}) \underline{E}^{*}(z,t,\omega_{2}),$$
$$\frac{\partial \underline{E}(z,t,\omega_{2})}{\partial z} = -j \frac{\omega_{2}}{cn(\omega_{2})} d_{\text{eff}} \underline{E}(0,t,\omega_{3}) \underline{E}^{*}(z,t,\omega_{1}).$$

General solution:

$$\underline{E}(z,t,\omega_{1}) = \underline{E}_{a} \cosh(\kappa z) + \underline{E}_{b} \sinh(\kappa z),$$

$$\underline{E}(z,t,\omega_{2}) = -j \sqrt{\frac{\omega_{2}n(\omega_{1})}{\omega_{1}n(\omega_{2})}} \frac{\underline{E}(0,t,\omega_{3})}{|\underline{E}(0,t,\omega_{3})|} (\underline{E}_{a}^{*} \sinh(\kappa z) + \underline{E}_{b}^{*} \cosh(\kappa z))$$
where $\kappa^{2} = \frac{\omega_{1}\omega_{2}d_{\text{eff}}^{2}}{c^{2}n(\omega_{1})n(\omega_{2})} |\underline{E}(0,t,\omega_{3})|^{2}.$



Parametric amplifier and oscillator



Optical parametric amplifier (OPA): Launch signal at frequency ω_1 :

> $E(0, t, \omega_1) = E_1$ $\underline{E}(0,t,\omega_2)=0.$

 \Rightarrow Parametric amplification of signal at ω_1 and generation of idler wave at ω_2 :

$$\underline{E}(z,t,\omega_1)$$

$$\underline{E}(z,t,\omega_2)$$

Signal: $\underline{E}(z, t, \omega_1) = \underline{E}_1 \cosh(\kappa z)$,

Idler:
$$\underline{E}(z,t,\omega_2) = -j \sqrt{\frac{\omega_2 n(\omega_1)}{\omega_1 n(\omega_2)}} \frac{\underline{E}(0,t,\omega_3)}{|\underline{E}(0,t,\omega_3)|} \underline{E}_1^* \sinh(\kappa z).$$

Optical parametric oscillator (OPO): Add mirrors to form an optical resonator



 \Rightarrow Wavelength can be adjusted by tuning phase-matching conditions!



Acousto-optics and photon-phonon interactions

Elasto-optic effect and strain





Visualization of the elasto-optic effect: The medium becomes birefringent under strain, leading to interference fringes in a polarized-light image.

Quantitative description: Strain tensor

$$\sigma_{kl} = \frac{1}{2} \left(\frac{\partial u_k}{\partial x_l} + \frac{\partial u_l}{\partial x_k} \right)$$

where $u = (u_1, u_2, u_3)$ denotes the vectorial displacement of a volume element (dx,dy,dz) at a position $(x_1, x_2, x_3) = (x, y, z)$.

A few examples $(x_1 = x, x_2 = y, x_3 = z, u_1 = 0, u_2 = V)$:



Figure adapted from lizuka, Elements of Photonics

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Elasto-optic tensor



Recall: Anisotropic media are characterized by the electric impermeability tensor η_{ij} and the associated index ellipsoid



$$\sum_{i,j} \eta_{ij} X_i X_j = 1$$

Strain-induced changes $\Delta \eta_{ij}$ of the impermeability tensor elements are related to the various elements of the strain tensor by the fourth-rank elasto-optic tensor p_{iikl}

$$\eta_{ij}(\sigma_{kl}) = \eta_{ij}(0) + \sum_{kl} p_{ijkl}\sigma_{kl}$$

Contracted notation:

ij/kl	11	22	33	23, 32	13, 31	12, 21
I/K	1	2	3	4	5	6

 \Rightarrow Elasto-optic tensor can be written as a 6 x 6 matrix

$$\Delta \eta_I = p_{IK} \sigma_K$$

Figure adapted from Saleh-Teich, Fundamentals of Photonics

Crystal symmetries further restrict elements of p_{IK} .


Elasto-optic tensors of different materials



Name of Substance	Chemical Symbol	Photoelastic Constant	Index of Refraction	Wavelength (µm)	Crystal Symmetry	Elastooptic Tensor					
Fused sili ca	Si O ₂	$p_{11} = 0.121$ $p_{12} = 0.270$ $p_{44} = p_{55} = p_{66}$ $= \frac{1}{2}(p_{11} - p_{12})$	<i>n</i> = 1.457	0.63	Isotropic	$\begin{bmatrix} p_{11} \\ p_{12} \\ p_{12} \\ p_{12} \end{bmatrix}$	<i>p</i> ₁₂ <i>p</i> ₁₁ <i>p</i> ₁₂	<i>p</i> ₁₂ <i>p</i> ₁₂ <i>p</i> ₁₁	0 0 0		
Water	$\rm H_2O$	$p_{11} = 0.31 p_{12} = 0.31 p_{44} = p_{55} = p_{66} = \frac{1}{2}(p_{11} - p_{12})$	<i>n</i> = 1.33	0.63	Isotropic	0	0 0	0 0	р ₄₄ 0 р 0	$p_{0} = 0$, , ,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
Galli um arsenide	GaAs	$p_{11} = -0.165$ $p_{12} = -0.140$ $p_{44} = -0.061$	$n_x = n_y = n_z = 3.42$	1.15	43m	$\begin{bmatrix} p_{11} \\ p_{12} \\ p_{12} \\ p_{12} \end{bmatrix}$	p_{12} p_{11} p_{12}	<i>p</i> ₁₂ <i>p</i> ₁₂ <i>p</i> ₁₁	0 0 0		<u>}</u>]
Zinc sulfide	β-ZnS	$p_{11} = 0.091 p_{12} = -0.01 p_{44} = 0.075$	$n_x = n_y = n_z = 2.352$	0.63		0	0	0 0	0 p 0 0) () 44 () 0 p	, ,] ,]
Lithium niobate	LiNbO3	$p_{11} = -0.02$ $p_{12} = 0.08$ $p_{13} = 0.13$ $p_{14} = -0.08$ $p_{31} = 0.17$ $p_{33} = 0.07$ $p_{41} = -0.15$ $p_{44} = 0.12$ $p_{66} = \frac{1}{2}(p_{11} - p_{12})$	$n_x = n_y = 2.286$ $n_z = 2.20$	0.63	3m	$\begin{bmatrix} p_{11} \\ p_{12} \\ p_{31} \\ p_{41} \\ 0 \\ 0 \end{bmatrix}$	p_{12} p_{11} p_{31} $-p_{41}$ 0 0	p_{13} p_{13} p_{33} 0 0 0	p_{14} $-p_{14}$ 0 p_{44} 0 0	0 0 0 <i>p</i> 44 <i>p</i> 14	$\begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ p_{41} \\ p_{66} \end{bmatrix}$

Figure adapted from lizuka, Elements of Photonics

Elasto-optic tensors of different materials



Name of Substance	Che mic al Sym bol	Photoelastic Constant	Index of Refraction	Wave lengt h (µm)	Crystal Symmetry	Elastooptic Tensor					
Lit hium tanta late	LiTaO3	$p_{11} = -0.08$ $p_{12} = -0.08$ $p_{13} = 0.09$ $p_{14} = -0.03$ $p_{31} = 0.09$ $p_{33} = -0.044$ $p_{41} = -0.085$ $p_{44} = 0.02$ $p_{66} = \frac{1}{2}(p_{11} - p_{12})$	$n_x = n_y = 2.1$ $n_z = 2.180$	76 0.63	3т						
Rutile	TiO ₂	$p_{11} = -0.011$ $p_{12} = 0.172$ $p_{13} = -0.168$ $p_{31} = -0.096$ $p_{33} = -0.058$ $p_{44} = 0.0095$ $p_{66} = \pm 0.072$	$n_x = n_y = 2.5$ $n_z = 2.875$	85 0.63 0.51 0.63	<u>4</u> 2m	$\begin{bmatrix} p_{11} \\ p_{12} \end{bmatrix}$	$p_{12} \\ p_{11}$	<i>p</i> ₁₃ <i>p</i> ₁₃	0	0	0
Potassium dihydrogen phosphate (KDP)	KH ₂ PO ₄	$p_{11} = 0.251$ $p_{12} = 0.249$ $p_{13} = 0.246$ $p_{31} = 0.225$ $p_{33} = 0.221$ $p_{44} = -0.019$ $p_{66} = -0.058$	$n_x = n_y = 1.5$ $n_z = 1.47$	1 0.63 0.59 0.63	42 <i>m</i>	$ \begin{bmatrix} p_{31} \\ 0 \\ 0 \\ 0 \end{bmatrix} $			<i>p</i> ₄₄ 0 0	0 <i>P</i> 44 0	$\begin{bmatrix} 0 \\ 0 \\ p_{66} \end{bmatrix}$

Figure adapted from lizuka, Elements of Photonics

Elasto-optic tensors of different materials



Name of Substance	Chemi cal Symbol	Photoelastic Constant	Index of Refraction	Wavelength (µm)	Crystal Symmetry	Elast ooptic Tensor			
Ammonium dihydrogen phosphate (ADP)	NH4H2PO4 or ADP	$p_{11} = 0.302$ $p_{12} = 0.246$ $p_{13} = 0.236$ $p_{31} = 0.195$ $p_{33} = 0.263$ $p_{44} = -0.058$ $p_{66} = -0.075$	$n_x = n_y = 1.52$ $n_z = 1.48$	0.63 0.59 0.59	<u>4</u> 2m	$\begin{bmatrix} p_{11} & p_{12} & p_1 \\ p_{12} & p_{11} & p_1 \\ p_{31} & p_{31} & p_3 \end{bmatrix}$	3 0 0 3 0 0 3 0 0	$\begin{bmatrix} 0\\0\\0\\0\\p_{66}\end{bmatrix}$	
Tellurium dioxide	TeO ₂	$p_{11} = 0.0074$ $p_{12} = 0.187$ $p_{13} = 0.340$ $p_{31} = 0.090$ $p_{33} = 0.240$ $p_{44} = -0.17$ $p_{66} = -0.046$	$n_x = n_y = n_z = 2.35$	0.63	42 <i>m</i>		$ \begin{array}{ccc} p_{44} & 0 \\ 0 & p_{44} \\ 0 & 0 \end{array} $		

Figure adapted from lizuka, Elements of Photonics



Acousto-optic modulator



Acousto-optic effect: Strain induced by an acoustic wave => "Wave-like" variation of refractive index

Example: Surface-acoustic wave (SAW) modulator based on TeO₂





Acousto-optic modulator







Dielectric displacement for small index perturbation: $\underline{\mathbf{D}}(\mathbf{r},t) \approx \epsilon_0 \left(n_0^2 \underline{\mathbf{E}}(\mathbf{r},t) + 2n_0 \Delta n \left(\mathbf{r},t\right) \underline{\mathbf{E}}(\mathbf{r},t) \right).$

Wave equation for acousto-optic interaction:

$$\nabla^{2}\underline{\mathbf{E}}(\mathbf{r},t) - \frac{n_{0}^{2}}{c^{2}}\frac{\partial^{2}\underline{\mathbf{E}}(\mathbf{r},t)}{\partial t^{2}} = \frac{2n_{0}}{c^{2}}\frac{\partial^{2}\left(\Delta n\left(\mathbf{r},t\right)\underline{\mathbf{E}}(\mathbf{r},t)\right)}{\partial t^{2}}$$

Slowly varying envelope approximation (in space only!):

$$\underline{\mathbf{E}}(\mathbf{r},t) = \sum_{l} \underline{E}(\mathbf{r},\omega_{l}) \, \mathbf{e}_{l} \, e^{\mathbf{j}(\omega_{l}t - \mathbf{k}_{l}\mathbf{r})}.$$
where $\left| \nabla^{2} \underline{E}(\mathbf{r},\omega_{l}) \right| \ll |\mathbf{k}_{l} \cdot \nabla \underline{E}(\mathbf{r},\omega_{l})|$

Coupled-wave equation for space-dependent wave amplitudes:

$$\sum_{l} \left[-2j\mathbf{k}_{l} \cdot \nabla \underline{E}(\mathbf{r}, \omega_{l})\right] \mathbf{e}_{l} e^{j(\omega_{l}t - \mathbf{k}_{l}\mathbf{r})} = \frac{2n_{0}}{c^{2}} \sum_{l} \frac{\partial^{2}}{\partial t^{2}} \left(\Delta n\left(\mathbf{r}, t\right) \underline{E}(\mathbf{r}, \omega_{l}) \mathbf{e}_{l} e^{j(\omega_{l}t - \mathbf{k}_{l}\mathbf{r})}\right).$$

Launch wave at frequency ω_0 , consider evolution of amplitude at $\omega_1 = \omega_0 + \Omega$:

$$\mathbf{k}_1 \cdot \nabla \underline{E}(\mathbf{r}, \omega_1) = -j \frac{1}{2} (\mathbf{e}_1 \cdot \mathbf{e}_0) \frac{\omega_1^2}{c^2} n_0 \Delta n_0 \underline{E}(\mathbf{r}, \omega_0) e^{-j(\mathbf{k}_0 + \mathbf{q} - \mathbf{k}_1)\mathbf{r}}.$$



Lecture 12

Acousto-optic modulator



Acousto-optic effect: Strain induced by an acoustic wave => "Wave-like" variation of refractive index

Example: Surface-acoustic wave (SAW) modulator based on TeO₂ (strong $p_{13} = p_{23}$)





Acousto-optic modulator







Dielectric displacement for small index perturbation: $\underline{\mathbf{D}}(\mathbf{r},t) \approx \epsilon_0 \left(n_0^2 \underline{\mathbf{E}}(\mathbf{r},t) + 2n_0 \Delta n \left(\mathbf{r},t\right) \underline{\mathbf{E}}(\mathbf{r},t) \right).$

Wave equation for acousto-optic interaction:

$$\nabla^{2}\underline{\mathbf{E}}(\mathbf{r},t) - \frac{n_{0}^{2}}{c^{2}}\frac{\partial^{2}\underline{\mathbf{E}}(\mathbf{r},t)}{\partial t^{2}} = \frac{2n_{0}}{c^{2}}\frac{\partial^{2}\left(\Delta n\left(\mathbf{r},t\right)\underline{\mathbf{E}}(\mathbf{r},t)\right)}{\partial t^{2}}$$

Slowly varying envelope approximation:

$$\underline{\mathbf{E}}(\mathbf{r},t) = \sum_{l} \underline{E}(\mathbf{r},\omega_{l}) \, \mathbf{e}_{l} \, e^{\mathbf{j}(\omega_{l}t - \mathbf{k}_{l}\mathbf{r})}.$$
where $\left|\nabla^{2}\underline{E}(\mathbf{r},\omega_{l})\right| \ll |\mathbf{k}_{l} \cdot \nabla \underline{E}(\mathbf{r},\omega_{l})|$

Coupled-wave equation for space-dependent wave amplitudes:

$$\sum_{l} \left[-2j\mathbf{k}_{l} \cdot \nabla \underline{E}(\mathbf{r}, \omega_{l})\right] \mathbf{e}_{l} e^{j(\omega_{l}t - \mathbf{k}_{l}\mathbf{r})} = \frac{2n_{0}}{c^{2}} \sum_{l} \frac{\partial^{2}}{\partial t^{2}} \left(\Delta n\left(\mathbf{r}, t\right) \underline{E}(\mathbf{r}, \omega_{l}) \mathbf{e}_{l} e^{j(\omega_{l}t - \mathbf{k}_{l}\mathbf{r})}\right).$$

Launch wave at frequency ω_0 , consider evolution of amplitude at $\omega_1 = \omega_0 + \Omega$:

$$\mathbf{k}_1 \cdot \nabla \underline{E}(\mathbf{r}, \omega_1) = -j \frac{1}{2} (\mathbf{e}_1 \cdot \mathbf{e}_0) \frac{\omega_1^2}{c^2} n_0 \Delta n_0 \underline{E}(\mathbf{r}, \omega_0) e^{-j(\mathbf{k}_0 + \mathbf{q} - \mathbf{k}_1)\mathbf{r}}.$$



Phase matching and Bragg condition – up-conversion





Phase matching:

$$\mathbf{k}_1 = \mathbf{k}_0 + \mathbf{q}$$
 where $|\mathbf{k}_0| \approx |\mathbf{k}_1|$

$$\sin \Theta_B = \frac{|\mathbf{q}|}{2|\mathbf{k}_0|} = \frac{\pi/\pi_0}{2\Lambda}.$$

Bragg angle: Reflections from neighboring wavefronts of the acoustic wave experience a relative phase delay of 2π and hence interfere constructively

Figure adapted from Saleh-Teich, Fundamentals of Photonics



Down-conversion and deflection from a standing wave







Bragg diffraction of beams



Diffraction of an optical beam from an acoustic plane wave



Only one plane-wave component satisfies the Bragg condition.

Diffraction of an optical beam from an acoustic beam



Acousto-optic modulators







Analogue acousto-optic modulator:

For weak acoustic waves, the intensity of the refracted light is proportional to the intensity of the acoustic wave.

Acousto-optic switch:

At high acoustic intensities, total reflection occurs, and the reflected beam can be turned on and off by switching the sound wave on and off.

Figures adapted from Saleh-Teich, Fudamentals of Photonics





Acousto-optic beam scanners





Acousto-optic space switches





Splitting of incident optical beam by using acoustic drive signals that comprise various frequency components

Figure adapted from lizuka, Elements of Photonics



Frequency shifter ("Bragg cell")







Quantum interpretation:

- Light wave of angular frequency ω and wavevector **k** corresponds to stream of photons of energy $\hbar \omega$ and momentum $\hbar \mathbf{k}$.
- Acoustic wave of angular frequency Ω and wavenumber q can be regarded as a stream of phonons of energy ħΩ and momentum ħq.
- Acousto-optic effects correspond to interaction photons with phonons, whereby new photons with frequency ω_s and wavevectors k_s can be generated.
- Energy and momentum conservation require

$$\omega_s = \omega + \Omega_s$$
$$\mathbf{k}_s = \mathbf{k} + \mathbf{q}$$



Acoustic and optical phonons Simple model: ₩€00000 M €00000 M €00000 M €00000 M €00000 G-spring 0000 **Diatomic linear chain** K-spring mass M M Optical phonons: $\omega(k)$ Motion of neighbouring atoms ,,out of phase" Localized vibrational normal modes of molecules $\frac{2K}{M}$ Bigger energy than acoustic phonons 2(K+G) Energy approximately independent of momentum $\frac{2G}{M}$ KI+VV+600001+VV+600001+VV+600001+VV+600001+ Acoustic phonons: Neighbouring atoms move "in phase" "Traveling sound waves" • Energy small; approximately proportional to momentum <u>π</u> 0 77 п a 000000 MM C00000 MM C00000 MM C00000 MM C00000

Figure adapted from Ashcroft/Mermin, Solid State Physics



Interaction of photons and phonons in optical fibers





Figure adapted from Saleh-Teich, Fundamentals of Photonics

Rayleigh scattering:

- Scattering due to localized fluctuations of optical density
- No frequency shift

Brillouin scattering:

- Scattering due to interactions with acoustic phonons
- Small frequency shift (~ 11 GHz in silica fibers)
- Only in backward direction

Raman scattering:

- Scattering due to interactions with optical phonons
- Large frequency shift (~ 13 THz in silica fibers)
- Scattering in forward and backward direction

Stokes process:

Photon loses energy during interaction

Anti-Stokes process:

Photon gains energy during interaction



Brillouin scattering



a)

11.4

11.2

- Interaction of light with a traveling sound wave (acoustic phonons)
- Only in backward direction
- Brillouin shift:

$$f_B = \frac{\Omega_B}{2\pi} = 2n\frac{v_s}{c}f$$

• Silica fiber:

 $f_{\rm B} \approx$ 11 GHz,

gain bandwidth 50 MHz ... 100 MHz

Brillouin shift depends on local strain and temperature of the fiber

SIGNAL

 \Rightarrow Application in "distributed sensing"

Stimulated Brillouin scattering (SBS):

• Interference of incident and scattered wave lead to beat signal and generates an acoustic wave with frequency $f_{\rm B}$ via the process of electrostriction

10.6

 \Rightarrow Positive feedback leads to "stimulated scattering"

• Quantitative model:

$$rac{dI_p}{dz} = -g_B I_p I_s - \alpha I_p,$$
 $g_B(\Omega) = g_B(\omega_p - \omega_s)$ Brillouin gain
 $rac{dI_s}{dz} = g_B I_p I_s + \alpha I_s,$ Figure adapted from Agrawal, Nonlinear Fiber Optics

c)

10.8

b)

11.0

FREQUENCY (GHz)



Raman scattering



- Interaction of light with vibrational states of the material molecules or atoms (optical phonons)
- Scattering in both forward and backward direction
- Silica fibers:
 - Raman shift ~ 13 THz
 - Large gain bandwidth (~ 10 THz) due to amorphous structure of the fibers (local inhomogeneities => Broadening of vibrational energy states)



Figure adapted from Agrawal, Fiber-Optic Communication Systems

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Stimulated Raman scattering and Raman amplifier

7 T



- \Rightarrow New phonons generated by electrostriction, which further enhance Raman scattering
- \Rightarrow Positive feedback

• Quantitative model:
$$\frac{dI_s}{dz} = g_R I_p I_s - \alpha_s I_s, \qquad g_R(\Omega) = g_R(\omega_p - \omega_s)$$

$$\frac{dI_p}{dz} = -\frac{\omega_p}{\omega_s} g_R I_p I_s - \alpha_p I_p, \qquad \text{Raman gain}$$

Raman amplifier:

Small-signal approximation: Neglect Raman-induced depletion of the pump wave



Example: Raman laser on silicon



Challenge: Light emission on silicon (indirect bandgap!) \Rightarrow Exploit stimulated Raman scattering in a high-Q silicon microring resonator: Pump-power monitor Polarization controller WDM filter Pump at 1.550 nm 99/1 Tap coupler Laser output at 1,686 nm -10 -20 Ring laser cavity LP filter -30 Optical 10 -40 spectrum -50 Output spectrum 0 analyser -60 Here: Pump laser -10 --70 -80 Relative spectral power (dB) at 1430.5 nm -20 -90/10 1,684 1,685 1,686 1,687 1,688 1,689 1,690 1,691 1,692 -30 Tap coupler 80 dB Laser output -40 power meter -50 -60 -70 · Rong, H.; Xu, S.; Kuo, Y.-H.; Sih, V.; Cohen, O.; Raday, -80 O. & Paniccia, M. (2007), 'Low-threshold continuous-wave -90 1,545 1,546 1,543 1.544 1.547 1.548 Raman silicon laser', Nat. Photon 1(4), 232-237. Wavelength (nm) Institute of Photonics Christian Koos 204 27.06.2018 and Quantum Electronics

Lecture 13



and Quantum Electronics

IPQ Lab Tour



Tuesday, 17. July 2018, 10:30 – 11:15 AM (after the NLO tutorial) Meeting point: Building 30.10, Room 3.42 (IPQ seminar room)

1µm

Au

Photonic integration, plasmonics and THz devices:



Muehlbrandt, S. et al., Optica 3, 741 (2016). 3D-Nanoprinting by two-photon Vanguard lithography/ Photonic wire bonds:



Billah, M. R. et al., ECOC'17, Th.PDP.C.1 (2017)

Frequency combs and optical communications:



Trocha, P. et al., Science **359**, 887-891 (2018)

207 27.06.2018

Note: Some third-order nonlinear optical effects are inherently phase-matched (SPM, XPM)!

Examples:

Nanophotonic waveguides:

Third-order nonlinearities

- Strong confinement: Small effective cross section (diameter < 1 μm)
- \Rightarrow Large intensities!
- Typically mm-scale interaction lengths



Optical fibers:

• Dominate in many materials (e.g., if second-order nonlinear effects are absent)

Can be strong for high intensities and/or large interaction lengths

- Core diameter \approx 10 μm
- Interaction over several kilometres!



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Large interaction lengths in optical fibers...









Capacity of optical fibers is eventually limited by nonlinearities!



Capacity / spectral efficiency of optical fibers is limited by third-order nonlinearities



Essiambre *et al.*, 'Capacity Limits of Optical Fiber Networks', *Journal of Lightwave Technology* **28(4)**, **662 -701** (2010)

Record transmission speed over a single fiber core: 101.7 Tbit/s

- Transmission distance: 3 x 55 km
- Spectral efficiency: 11 bit/s/Hz
- Total power < 100 mW



Qian *et al.,*, 'High Capacity/Spectral Efficiency 101.7-Tb/s WDM Transmission Using PDM-128QAM-OFDM Over 165-km SSMF Within C- and L-Bands', *Lightwave Technology, Journal of* **30(10), 1540-1548** (2012).



Propagation of monochromatic signals in optical waveguides



Waveguide modes for monochromatic waves:

Lossless *z*-invariant dielectric structure ("lossless homogeneous waveguide"):

n=n(x,y)

where Im{<u>n</u>} = 0 throughout space. **Eigenmodes:** A lossless homogenous waveguide features a set of electromagnetic wave patterns which do not change their transverse shapes during propagation along z, so-called eigenmodes:

$$\underline{\mathbf{E}}(\mathbf{r},t) = \underline{\mathcal{E}}(x,y,\omega) e^{j(\omega t - \beta(\omega)z)},$$

$$\underline{\mathbf{H}}(\mathbf{r},t) = \underline{\mathcal{H}}(x,y,\omega) e^{j(\omega t - \beta(\omega)z)}$$

$$\beta(\omega) \quad \text{Dispersion relation}$$



Further information: Lecture "Optical waveguides and fibers (OWF)" ⇒ Winter term

 \Rightarrow Maxwell's equations for monochromatic guided modes (to be solved numerically...):

$$(\nabla \times \underline{\mathcal{E}}(x, y, \omega)) - j\beta(\omega) e_z \times \underline{\mathcal{E}}(x, y, \omega) = -j\omega\mu_0 \underline{\mathcal{H}}(x, y, \omega)$$

$$(\nabla \times \underline{\mathcal{H}}(x, y, \omega)) - j\beta(\omega) e_z \times \underline{\mathcal{H}}(x, y, \omega) = j\omega\epsilon_0 n^2 \underline{\mathcal{E}}(x, y, \omega).$$

Propagation of monochromatic signals in optical waveguides



Mode expansion: For a given frequency, *any* field pattern propagating along a waveguide can expressed as a superposition of eigenmodes (completeness)

$$\underline{\mathbf{E}}(\mathbf{r},t) = \sum_{\mu} \underline{A}_{\mu} \underline{\mathcal{E}}_{\mu}(x,y,\omega) e^{\mathbf{j}(\omega t - \beta_{\mu}(\omega)z)},$$
$$\underline{\mathbf{H}}(\mathbf{r},t) = \sum_{\mu} \underline{A}_{\mu} \underline{\mathcal{H}}_{\mu}(x,y,\omega) e^{\mathbf{j}(\omega t - \beta_{\mu}(\omega)z)},$$



Orthogonality relation:

$$\frac{1}{4} \iint_{-\infty}^{\infty} \left(\underline{\mathcal{E}}_{\nu}(x,y) \times \underline{\mathcal{H}}_{\mu}^{\star}(x,y) + \underline{\mathcal{E}}_{\mu}^{\star}(x,y) \times \underline{\mathcal{H}}_{\nu}(x,y) \right) \cdot \mathbf{e}_{z} \, \mathrm{d} \, x \, \mathrm{d} \, y = \mathcal{P}_{\mu} \delta_{\nu\mu},$$

where
$$\mathcal{P}_{\mu} = \frac{1}{2} \iint_{-\infty}^{\infty} \operatorname{Re} \left\{ \underline{\mathcal{E}}_{\mu}(x, y) \times \underline{\mathcal{H}}_{\mu}^{\star}(x, y) \right\} \cdot \mathbf{e}_{z} \, \mathrm{d} \, x \, \mathrm{d} \, y.$$

Further information: Lecture "Optical waveguides and fibers (OWF)" \Rightarrow Winter term

212 27.06.2018 Christian Koos



Propagation of time-dependent signals in a linear waveguide



Time-domain description, based on slowly-varying envelopes:

$$\underline{\mathbf{E}}(\mathbf{r},t) = \underline{A}(z,t) \underline{\mathcal{E}}(x,y,\omega_c) e^{\mathbf{j}(\omega_c t - \beta(\omega_c)z)},$$

$$\underline{\mathbf{H}}(\mathbf{r},t) = \underline{A}(z,t) \underline{\mathcal{H}}(x,y,\omega_c) e^{\mathbf{j}(\omega_c t - \beta(\omega_c)z)}$$

Maxwell's equations:

$$\underline{\widetilde{A}}(z,\omega-\omega_{c})\left(\nabla\times\underline{\mathcal{E}}(x,y,\omega_{c})\right) + \left(\frac{\partial\underline{\widetilde{A}}(z,\omega-\omega_{c})}{\partial z} - j\beta\left(\omega_{c}\right)\underline{\widetilde{A}}(z,\omega-\omega_{c})\right)e_{z}\times\underline{\mathcal{E}}(x,y,\omega_{c}) = -j\omega\mu_{0}\underline{\mathcal{H}}(x,y,\omega_{c}).$$

Differential equation in frequency domain:

$$\frac{\partial \underline{\widetilde{A}}(z,\omega-\omega_{c})}{\partial z}+j\left(\beta\left(\omega\right)-\beta\left(\omega_{c}\right)\right)\underline{\widetilde{A}}(z,\omega-\omega_{c})=0$$

Use Taylor expansion of dispersion relation about ω_c :

$$\beta(\omega) \approx \beta_c^{(0)} + (\omega - \omega_c)\beta_c^{(1)} + \frac{(\omega - \omega_c)^2}{2!}\beta_c^{(2)} + \frac{(\omega - \omega_c)^3}{3!}\beta_c^{(3)} + \dots ,$$

where $\beta_c^{(i)} = \frac{d^i\beta(\omega)}{d\omega^i}\Big|_{\omega = \omega_c}$.

213 27.06.2018 Christian Koos



Propagation of time-dependent signals in a linear waveguide



Differential equation in the time domain:

$$\frac{\partial \underline{A}(z,t)}{\partial z} + \beta_c^{(1)} \frac{\partial \underline{A}(z,t)}{\partial t} - j \frac{1}{2} \beta_c^{(2)} \frac{\partial^2 \underline{A}(z,t)}{\partial t^2} + \ldots = 0.$$

Introduce retarded time frame:

$$t' = t - \beta_c^{(1)} z,$$

$$z' = z,$$

$$\underline{A}(z,t) = \underline{A}'(z,t - \beta_c^{(1)} z).$$

Simplified differential equation:

$$\frac{\partial \underline{A}'(z',t')}{\partial z'} - j\frac{1}{2}\beta_c^{(2)}\frac{\partial^2 \underline{A}(z',t')}{\partial t'^2} + \ldots = 0.$$

Example: Propagation of a Gaussian impulse through a dispersive waveguide

Solve corresponding DEq. in the frequency domain:

$$\underline{\widetilde{A}}(z,\omega) = \underline{\widetilde{A}}(0,\omega) e^{-j\frac{1}{2}\beta_c^{(2)}\omega^2 z}.$$





Lecture 14
IPQ Lab Tour



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1µm

Au

Photonic integration, plasmonics and THz devices:



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Frequency combs and optical communications:



Trocha, P. et al., Science **359**, 887-891 (2018)

217 27.06.2018

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Propagation of time-dependent signals in a linear waveguide



Differential equation in the time domain:

$$\frac{\partial \underline{A}(z,t)}{\partial z} + \beta_c^{(1)} \frac{\partial \underline{A}(z,t)}{\partial t} - j \frac{1}{2} \beta_c^{(2)} \frac{\partial^2 \underline{A}(z,t)}{\partial t^2} + \ldots = 0.$$

Introduce retarded time frame:

$$t' = t - \beta_c^{(1)} z,$$

$$z' = z,$$

$$\underline{A}(z,t) = \underline{A}'(z,t - \beta_c^{(1)} z).$$

Simplified differential equation:

$$\frac{\partial \underline{A}'(z',t')}{\partial z'} - j\frac{1}{2}\beta_c^{(2)}\frac{\partial^2 \underline{A}(z',t')}{\partial t'^2} + \ldots = 0.$$

Example: Propagation of a Gaussian impulse through a dispersive waveguide

Solve corresponding DEq. in the frequency domain:

$$\underline{\widetilde{A}}(z,\omega) = \underline{\widetilde{A}}(0,\omega) e^{-j\frac{1}{2}\beta_c^{(2)}\omega^2 z}.$$





Signal propagation in third-order nonlinear waveguides



Now: Consider joint influence of dispersion and optical nonlinearities

Maxwell's equations:
$$\nabla \times \mathbf{H}(\mathbf{r},t) = \epsilon_0 n^2 \frac{\partial}{\partial t} \mathbf{E}(\mathbf{r},t) + \frac{\partial}{\partial t} \mathbf{P}_{\mathsf{NL}}(\mathbf{r},t),$$

 $\nabla \times \mathbf{E}(\mathbf{r},t) = -\mu_0 \frac{\partial}{\partial t} \mathbf{H}(\mathbf{r},t).$

General SVEA mode ansatz:

$$\begin{split} \mathbf{E}(\mathbf{r},t) &= \frac{1}{2} \sum_{m=-M}^{M} \sum_{\mu} \underline{A}_{\mu} \left(z,t,\omega_{m}\right) \frac{\underline{\mathcal{E}}_{\mu}(x,y,\omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} e^{\mathbf{j}(\omega_{m}t-\beta_{\mu}(\omega_{m})z)},\\ \mathbf{H}(\mathbf{r},t) &= \frac{1}{2} \sum_{\substack{m=-M \\ \neq}}^{M} \sum_{\mu} \underline{A}_{\mu} \left(z,t,\omega_{m}\right) \frac{\underline{\mathcal{H}}_{\mu}(x,y,\omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} e^{\mathbf{j}(\omega_{m}t-\beta_{\mu}(\omega_{m})z)},\\ \text{Various frequencies (coupled by optical nonlinearities)} & \text{Power normalization (coupled by optical nonlinearities)} \\ \text{where } \mathcal{P}_{\mu} &= \frac{1}{2} \iint_{-\infty}^{\infty} \operatorname{Re} \left\{ \underline{\mathcal{E}}_{\mu}(x,y) \times \underline{\mathcal{H}}_{\mu}^{\star}(x,y) \right\} \cdot \mathbf{e}_{z} \operatorname{d} x \operatorname{d} y.\\ \mathbf{P}_{\mathsf{NL}}(\mathbf{r},t) &= \frac{1}{2} \sum_{m=-M}^{+M} \underline{\mathbf{P}}_{\mathsf{NL}}(\mathbf{r},t,\omega_{m}) e^{\mathbf{j}\omega_{m}t}, \end{split}$$

220 27.06.2018 Christian Koos





Insert mode ansatz into Maxwell's equations in the frequency domain: Recall: $\nabla \times (\Phi \mathbf{F}) = \Phi (\nabla \times \mathbf{F}) + (\nabla \Phi \times \mathbf{F})$

$$\Rightarrow \sum_{\mu} \underline{\widetilde{A}}_{\mu} (z, \omega - \omega_m, \omega_m) e^{-j\beta_{\mu}(\omega_m)z} \left[\nabla \times \frac{\underline{\mathcal{H}}_{\mu}(x, y, \omega_m)}{\sqrt{\mathcal{P}_{\mu}}} \right] \\ + \frac{\partial}{\partial z} \left[\underline{\widetilde{A}}_{\mu} (z, \omega - \omega_m, \omega_m) e^{-j\beta_{\mu}(\omega_m)z} \right] \mathbf{e}_z \times \frac{\underline{\mathcal{H}}_{\mu}(x, y, \omega_m)}{\sqrt{\mathcal{P}_{\mu}}} \\ - j\omega\epsilon_0 n^2 \underline{\widetilde{A}}_{\mu} (z, \omega - \omega_m, \omega_m) \frac{\underline{\mathcal{E}}_{\mu}(x, y, \omega_m)}{\sqrt{\mathcal{P}_{\mu}}} e^{-j\beta_{\mu}(\omega_m)z} = j\omega \underline{\widetilde{P}}_{\mathsf{NL}}(\mathbf{r}, \omega - \omega_m, \omega_m)$$

$$\begin{split} \sum_{\mu} \underline{\widetilde{A}}_{\mu} \left(z, \omega - \omega_{m}, \omega_{m} \right) e^{-j\beta_{\mu}(\omega_{m})z} \left[\nabla \times \frac{\underline{\mathcal{E}}_{\mu}(x, y, \omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} \right] \\ + \frac{\partial}{\partial z} \left[\underline{\widetilde{A}}_{\mu} \left(z, \omega - \omega_{m}, \omega_{m} \right) e^{-j\beta_{\mu}(\omega_{m})z} \right] \mathbf{e}_{z} \times \frac{\underline{\mathcal{E}}_{\mu}(x, y, \omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} \\ + j\omega\mu_{0} \underline{\widetilde{A}}_{\mu} \left(z, \omega - \omega_{m}, \omega_{m} \right) \frac{\underline{\mathcal{H}}_{\mu}(x, y, \omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} e^{-j\beta_{\mu}(\omega_{m})z} = 0. \end{split}$$

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Signal propagation in third-order nonlinear waveguides



$$\begin{aligned} & \operatorname{Recall:} \ \underline{\mathcal{E}}_{\mu}(x,y,\omega_{m}) \ \text{and} \ \underline{\mathcal{H}}_{\mu}(x,y,\omega_{m}) \ \text{are guided} \\ & \operatorname{modes of the linear waveguide!} \\ \Rightarrow \quad \left(\nabla \times \frac{\underline{\mathcal{E}}_{\mu}(x,y,\omega)}{\sqrt{\mathcal{P}_{\mu}}} \right) = j\beta_{\mu}(\omega) \ \mathbf{e}_{z} \times \frac{\underline{\mathcal{E}}_{\mu}(x,y,\omega)}{\sqrt{\mathcal{P}_{\mu}}} - j\omega\mu_{0} \frac{\underline{\mathcal{H}}_{\mu}(x,y,\omega)}{\sqrt{\mathcal{P}_{\mu}}}, \\ & \left(\nabla \times \frac{\underline{\mathcal{H}}_{\mu}(x,y,\omega)}{\sqrt{\mathcal{P}_{\mu}}} \right) = j\beta_{\mu}(\omega) \ \mathbf{e}_{z} \times \frac{\underline{\mathcal{H}}_{\mu}(x,y,\omega)}{\sqrt{\mathcal{P}_{\mu}}} + j\omega\epsilon_{0}n^{2} \frac{\underline{\mathcal{E}}_{\mu}(x,y,\omega)}{\sqrt{\mathcal{P}_{\mu}}}. \end{aligned}$$

$$\begin{aligned} & \operatorname{Insert into derived relations:} \qquad \operatorname{Note:} \ \underline{\mathcal{E}}_{\mu}(x,y,\omega) \approx \underline{\mathcal{E}}_{\mu}(x,y,\omega_{m}) \ \text{for } \omega \approx \omega_{m} \\ & \sum_{\mu} \left[\frac{\partial \underline{\tilde{A}}_{\mu}(z,\omega-\omega_{m},\omega_{m})}{\partial z} + j\left(\beta_{\mu}(\omega) - \beta_{\mu}(\omega_{m})\right) \underline{\tilde{A}}_{\mu}(z,\omega-\omega_{m},\omega_{m}) \right] \mathbf{e}_{z} \\ & \times \frac{\underline{\mathcal{H}}_{\mu}(x,y,\omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} e^{-j\beta_{\mu}(\omega_{m})z} = j\omega \underline{\tilde{P}}_{\mathrm{NL}}(\mathbf{r},\omega-\omega_{m},\omega_{m}) \\ & \sum_{\mu} \left[\frac{\partial \underline{\tilde{A}}_{\mu}(z,\omega-\omega_{m},\omega_{m})}{\partial z} + j\left(\beta_{\mu}(\omega) - \beta_{\mu}(\omega_{m})\right) \underline{\tilde{A}}_{\mu}(z,\omega-\omega_{m},\omega_{m}) \right] \mathbf{e}_{z} \\ & \times \frac{\underline{\mathcal{E}}_{\mu}(x,y,\omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} e^{-j\beta_{\mu}(\omega_{m})z} = 0 \end{aligned}$$

$$\begin{aligned} & \operatorname{Recall: Othogonality relation} \\ & 1 \le c\infty \end{aligned}$$

 $\frac{1}{4} \iint_{-\infty}^{\infty} \left(\underline{\mathcal{E}}_{\mu}(x,y) \times \underline{\mathcal{H}}_{\nu}^{\star}(x,y) + \underline{\mathcal{E}}_{\nu}^{\star}(x,y) \times \underline{\mathcal{H}}_{\mu}(x,y) \right) \cdot \mathbf{e}_{z} \, \mathrm{d} \, x \, \mathrm{d} \, y = \mathcal{P}_{\mu} \delta_{\nu\mu},$

222 27.06.2018 Christian Koos



Dot-multiply with
$$[-\underline{\mathcal{E}}_{\nu}^{*}(x, y, \omega_{m})]$$
 and $\underline{H}_{\nu}^{*}(x, y, \omega_{m})$ and add resulting relations

$$\Rightarrow \sum_{\mu} \left[\frac{\partial \underline{\widetilde{A}}_{\mu} \left(z, \omega - \omega_{m}, \omega_{m} \right)}{\partial z} + j \left(\beta_{\mu} \left(\omega \right) - \beta_{\mu} \left(\omega_{m} \right) \right) \underline{\widetilde{A}}_{\mu} \left(z, \omega - \omega_{m}, \omega_{m} \right) \right]$$

$$\left[\frac{\underline{\mathcal{E}}_{\mu}(x, y, \omega_{m}) \times \underline{H}_{\nu}^{*}(x, y, \omega_{m}) + \underline{\mathcal{E}}_{\nu}^{*}(x, y, \omega_{m}) \times \underline{\mathcal{H}}_{\mu}(x, y, \omega_{m})}{\sqrt{\mathcal{P}_{\mu}}} \right] \mathbf{e}_{z} e^{-j\beta_{\mu}(\omega_{m})z}$$

$$= -j\omega \underline{\widetilde{P}}_{\mathsf{NL}}(\mathbf{r}, \omega - \omega_{m}, \omega_{m}) \cdot \underline{\mathcal{E}}_{\nu}^{*}(x, y, \omega_{m})$$

Integrate over the entire (x,y)-plane and make use of orthogonality relation

$$\Rightarrow \left[\frac{\partial \underline{\widetilde{A}}_{\nu}\left(z,\omega-\omega_{m},\omega_{m}\right)}{\partial z}+j\left(\beta_{\nu}\left(\omega\right)-\beta_{\nu}\left(\omega_{m}\right)\right)\underline{\widetilde{A}}_{\nu}\left(z,\omega-\omega_{m},\omega_{m}\right)\right]e^{-j\beta_{\nu}\left(\omega_{m}\right)z}$$
$$=-\frac{j\omega}{4\sqrt{\mathcal{P}_{\nu}}}\iint_{-\infty}^{\infty}\underline{\widetilde{P}}_{\mathsf{NL}}(\mathbf{r},\omega-\omega_{m},\omega_{m})\cdot\underline{\mathcal{E}}_{\nu}^{*}(x,y,\omega_{m})\,\mathrm{d}x\,\mathrm{d}y$$

Use Taylor expansion of the propagation constant $\beta_{\nu}(\omega)$ about the carrier frequency ω_m

$$\beta(\omega) \approx \beta_c^{(0)} + (\omega - \omega_c)\beta_c^{(1)} + \frac{(\omega - \omega_c)^2}{2!}\beta_c^{(2)} + \frac{(\omega - \omega_c)^3}{3!}\beta_c^{(3)} + \dots,$$

223 27.06.2018 Christian Koos





Transform back to the time domain ...

$$\Rightarrow \left[\frac{\partial \underline{A}_{\nu}\left(z,t,\omega_{m}\right)}{\partial z} + \beta_{c}^{\left(1\right)}\frac{\partial \underline{A}_{\nu}\left(z,t,\omega_{m}\right)}{\partial t} - j\frac{1}{2}\beta_{c}^{\left(2\right)}\frac{\partial^{2}\underline{A}_{\nu}\left(z,t,\omega_{m}\right)}{\partial t^{2}}\right]e^{j\left(\omega_{m}t-\beta_{\nu}\left(\omega_{m}\right)z\right)}$$
$$= -\frac{1}{4\sqrt{\mathcal{P}_{\nu}}}\frac{\partial}{\partial t}\left(\iint_{-\infty}^{\infty}\underline{P}_{\mathsf{NL}}(\mathbf{r},t,\omega_{m})\cdot\underline{\mathcal{E}}_{\nu}^{*}(x,y,\omega_{m})\,\mathrm{d}x\,\mathrm{d}y\,e^{j\omega_{m}t}\right)$$

Introduce retarded time frame:

$$t' = t - \beta_c^{(1)} z,$$

$$z' = z,$$

$$\underline{A}(z,t) = \underline{A}'(z,t - \beta_c^{(1)} z).$$

$$\Rightarrow \qquad \left[\frac{\partial \underline{A}_{\nu}(z,t,\omega_{m})}{\partial z} - j\frac{1}{2}\beta_{c}^{(2)}\frac{\partial^{2}\underline{A}_{\nu}(z,t,\omega_{m})}{\partial t^{2}}\right]e^{-j\beta_{\nu}(\omega_{m})z} \\ = -\frac{j\omega_{m}}{4\sqrt{\mathcal{P}_{\nu}}}\iint_{-\infty}^{\infty}\underline{P}_{\mathsf{NL}}(\mathbf{r},t,\omega_{m})\cdot\underline{\mathcal{E}}_{\nu}^{*}(x,y,\omega_{m})\,\mathrm{d}x\,\mathrm{d}y.$$

(primes skipped ...)



Description of nonlinear polarization



Recall:
$$\underline{\mathbf{P}}^{(3)}(\omega_m) = \frac{1}{4} \epsilon_0 \sum_{\mathbb{S}(\omega_m)} \underline{\chi}^{(3)}(\omega_m : \omega_l, \omega_p, \omega_o) : \underline{\mathbf{E}}(\omega_l) \underline{\mathbf{E}}(\omega_p) \underline{\mathbf{E}}(\omega_o)$$

where $\mathbb{S}(\omega_m) = \left\{ (l_1, \dots, l_n) | \omega_{l_1} + \dots + \omega_{l_n} = \omega_m \right\}.$

 \Rightarrow For SPM (one frequency component at ω_m only, excitation only by dominant mode ν):

$$\underline{\mathbf{P}}_{\mathsf{NL}}(\mathbf{r},t,\omega_m) = \frac{3}{4} \epsilon_0 \left(\underline{\chi}^{(3)}(\omega_m : \omega_m, -\omega_m, \omega_m) : \underline{\mathcal{E}}_{\nu}(x, y, \omega_m) \underline{\mathcal{E}}_{\nu}^*(x, y, \omega_m) \underline{\mathcal{E}}_{\nu}(x, y, \omega_m) \right) \\ \frac{\underline{A}_{\nu}(z, t, \omega_m)}{\sqrt{\mathcal{P}_{\nu}}} \frac{\underline{A}_{\nu}^*(z, t, \omega_m)}{\sqrt{\mathcal{P}_{\nu}}} \frac{\underline{A}_{\nu}(z, t, \omega_m)}{\sqrt{\mathcal{P}_{\nu}}} e^{-\mathbf{j}\beta_{\nu}(\omega_m)z}$$

 \Rightarrow Nonlinear Schrödinger Equation (NLSE):

$$\frac{\partial \underline{A}_{\nu}\left(z,t,\omega_{m}\right)}{\partial z} - j\frac{1}{2}\beta_{c}^{(2)}\frac{\partial^{2}\underline{A}_{\nu}\left(z,t,\omega_{m}\right)}{\partial t^{2}} = -j\gamma \left|\underline{A}_{\nu}\left(z,t,\omega_{m}\right)\right|^{2}\underline{A}_{\nu}\left(z,t,\omega_{m}\right),$$
where $\gamma_{\nu}\left(\omega_{m}\right) = \frac{3\omega_{m}\epsilon_{0}}{16}\frac{\int\int_{-\infty}^{\infty}\left[\underline{\chi}^{(3)}:\underline{\mathcal{E}}_{\nu}\underline{\mathcal{E}}_{\nu}^{*}\underline{\mathcal{E}}_{\nu}\right]\cdot\underline{\mathcal{E}}_{\nu}^{*}\,\mathrm{d}x\,\mathrm{d}y}{\mathcal{P}_{\nu}^{2}},$
Nonlinearity parameter

 \Rightarrow Include waveguide losses:

$$\frac{\partial \underline{A}_{\nu}(\omega_{m})}{\partial z} - j\frac{1}{2}\beta_{c}^{(2)}\frac{\partial^{2}\underline{A}_{\nu}(\omega_{m})}{\partial t^{2}} = -\frac{\alpha}{2}\underline{A}_{\nu}(\omega_{m}) - j\gamma |\underline{A}_{\nu}(\omega_{m})|^{2} \underline{A}_{\nu}(\omega_{m}),$$

(



Simplifications for optical fibers



Optical fibers:

- Low index contrast, i.e., index *n* of the cladding is usually very similar, $n \approx n_{core} \approx n_{clad}$
- Isotropic material
- Homogeneous nonlinearity, which does not change over the cross section
- $\Rightarrow \text{Transverse components of the mode fields may be approximated by a scalar function:} \quad \underline{\mathcal{E}}_{\nu}(x, y, \omega_m) \approx F_{\nu}(x, y, \omega_m) \, \mathbf{e}_x, \\ \underline{\mathcal{H}}_{\nu}(x, y, \omega_m) \approx \frac{n}{Z_0} F_{\nu}(x, y, \omega_m) \, \mathbf{e}_y.$
- \Rightarrow Simplified representation of nonlinearity parameter:

$$\begin{split} \gamma_{\nu}\left(\omega_{m}\right) &\approx \frac{\omega_{m}n_{2}}{cA_{\text{eff}}}, \\ \text{where} \quad n_{2} &= \frac{3Z_{0}}{4n^{2}}\chi^{(3)}, \quad \text{Kerr coefficient} \\ A_{\text{eff}} &\approx \frac{\left(\iint_{-\infty}^{\infty}|F_{\nu}(x,y,\omega_{m})|^{2} \text{ d}x \text{ d}y\right)^{2}}{\iint_{-\infty}^{\infty}|F_{\nu}(x,y,\omega_{m})|^{4} \text{ d}x \text{ d}y} \quad \text{Effective cross section} \end{split}$$

226 27.06.2018 Christian Koos





Neglect dispersion:
$$\frac{\partial \underline{A}(z,t)}{\partial z} = -j\gamma |\underline{A}(z,t)|^2 \underline{A}(z,t) - \frac{\alpha}{2} \underline{A}(z,t) ,$$

Solution ansatz:
$$\underline{A}(z,t) = \underline{A}_0(t) e^{j\Phi_{NL}(z,t)} e^{-\frac{\alpha}{2}z},$$
$$\Rightarrow \text{Nonlinear phase shift:} \quad \Phi_{NL}(L,t) = -\gamma |\underline{A}_0(t)|^2 L_{\text{eff}},$$

Nonlinear phase shift:
$$\Phi_{NL}(L,t) = -$$

where
$$L_{\text{eff}} = \frac{1 - e^{-\alpha L}}{\alpha}$$
.

Effective length (slightly shorter than geometrical length due to waveguide losses)

Note:

- SPM leaves the temporal pulse power envelope unchanged, but leads to spectral broadening
- Instantaneous frequency offset Ω negative near the leading edge of the pulse (red-shift) and positive near the trailing edge (blue-shift)
- Interplay of SPM and anomalous GVD can lead to pulse forms that do not change their • envelope during propagation, so-called solitons (see next section!)



Figure adapted from: Saleh, B. E. A. & Teich, M. C. (2007), Fundamentals of Photonics, John Wiley & Sons, Hoboken, NJ.

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Spectral broadening and supercontinuum generation



Spectral broadening of a Gaussian pulse for different maximum nonlinear phase shifts at the pulse peak



Frequency





Frequency comb generation



Mode-locked laser + spectral broadening in highly nonlinear fiber (HNLF) Problem: Spectral dips for SPM-induced phase shifts of $\Delta \Phi \ge 1.5 \pi$



 \Rightarrow 325 carriers with 12.5 GHz spacing, linewidth < 10 kHz

Hillerkuss *et al.*, J. Opt. Commun. Netw. 4, 715–723 (2012)

Optical solitons





Mathematical analysis: Start from NLSE, neglect losses

$$\frac{\partial \underline{A}(z,t)}{\partial z} - j\frac{1}{2}\beta_c^{(2)}\frac{\partial^2 \underline{A}(z,t)}{\partial t^2} = -j\gamma |\underline{A}(z,t)|^2 \underline{A}(z,t).$$

Solution ansatz: Real, z-independent envelope + z-dependent "global" phase shift

$$\underline{A}(z,t) = A_0(t) e^{j\Phi(z)}$$





Separation of variables:

$$\Phi(z) = -Kz,$$

$$\frac{\partial^2 A_0(t)}{\partial t^2} = \frac{2}{\beta_c^{(2)}} \left(-K + \gamma A_0^2(t)\right) A_0(t),$$
Solution ansatz for time dependence:

$$A_0(t) = A_1 \operatorname{sech}\left(\frac{t}{T}\right),$$

$$\Rightarrow \operatorname{Total solution:} \quad \underline{A}(z,t) = A_1 \operatorname{sech}\left(\frac{t}{T}\right) e^{-jKz},$$
where

$$A_1^2 = \frac{-\beta_c^{(2)}}{\gamma T^2}, \quad \operatorname{Peak power}\left(\operatorname{Note:} \beta_c^{(2)} < 0!\right)$$

$$K = \frac{1}{2}\gamma A_1^2. \quad \operatorname{Global nonlinear phase shift}_{2}$$
Note: Soliton duration *T* and peak power |A_1|^2 are linked to each other. The smaller *T*, the bigger the impact of dispersion, and the bigger the peak power |A_1|^2 must be to compensate the dispersion by SPM.
Fluxe adapted from: Sateh. B. E. A. & Teich. M. C. (2007). Fundamentals of Photonics. John Wiley & Sons. Hoboken, NJ.

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Higher-order solitons





Fundamental soliton: Real, z-invariant envelope Higher-order soliton: Complex envelope that reproduces itself periodically during propagation.

Figure adapted from: Saleh, B. E. A. & Teich, M. C. (2007), Fundamentals of Photonics, John Wiley & Sons, Hoboken, NJ.



Modulation instability



Modulation instability:

Nonlinear interaction transfers power from a strong continuous-wave (cw) signal to spectral sidebands. This may lead to the break-up of the cw signal into a train of pulses.



Solution: $\Phi_{NL}(z) = -\gamma |A_0|^2 z$

But: Is this solution stable? What happens to small perturbations $\Delta \underline{A}(z,t)$ of the solution? Do they increase or decay?



Figures adapted from Nature Photonics 6, 415–416 (2012), and from Agrawal, Nonlinear Fiber Optics

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Stability analysis



Ansatz: Continuous-wave solution which is perturbed by a small amplitude perturbation $\Delta \underline{A}(z,t)$ $\underline{A}(z,t) = (A_0 + \Delta \underline{A}(z,t)) e^{-j\gamma |A_0|^2 z}$

 \Rightarrow Linearize NLSE to obtain linear differential equation for the amplitude perturbation:

$$\frac{\partial \Delta \underline{A}(z,t)}{\partial z} = j \frac{1}{2} \beta_c^{(2)} \frac{\partial^2 \Delta \underline{A}(z,t)}{\partial t^2} - j\gamma |\underline{A}_0|^2 \left(\Delta \underline{A}(z,t) + \Delta \underline{A}^*(z,t) \right)$$

Time- and space-harmonic ansatz:

$$\Delta \underline{A}(z,t) = C_1 e^{\mathbf{j}(\Omega t - Kz)} + C_2 e^{-\mathbf{j}(\Omega t - Kz)}$$

 \Rightarrow Linear equations for wave amplitudes C₁ and C₂:

$$\begin{pmatrix} -K + \frac{1}{2}\Omega^{2}\beta_{c}^{(2)} + \gamma |\underline{A}_{0}|^{2} & \gamma |\underline{A}_{0}|^{2} \\ \gamma |\underline{A}_{0}|^{2} & K + \frac{1}{2}\Omega^{2}\beta_{c}^{(2)} + \gamma |\underline{A}_{0}|^{2} \end{pmatrix} \begin{pmatrix} C_{1} \\ C_{2} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$

Nontrivial solutions only if the determinant vanishes:

$$K = \pm \sqrt{\left(\frac{1}{2}\Omega^2 \beta_c^{(2)}\right)^2 + \Omega^2 \beta_c^{(2)} \gamma |\underline{A}_0|^2}$$

Dispersion relation for the evolution of the perturbation



Dispersion and gain spectrum

Normal dispersion ($\beta_c^{(2)} > 0$): K real \Rightarrow Solution always stable

Anomalous dispersion ($\beta_c^{(2)} < 0$): K imaginary for $|\Omega| < \Omega_g = \sqrt{\frac{4\gamma |\underline{A}_0|^2}{\rho^{(2)}}}$

 \Rightarrow Solution unstable

Associated gain spectrum:

$$g(\Omega) = \left|\beta_c^{(2)}\right| \Omega \sqrt{\Omega_g^2 - \Omega^2}$$

Gain spectra for three power levels $L_{NL} = 1 \text{ km}$ of the cw signal. The so-called "nonlinear length" is given by Instability Gain (km⁻¹) 5.1 $L_{\rm NL} = \left(\gamma \left|\underline{A}_0\right|^2\right)^{-1}$ 2 km 0.5 5 km 0 -150 -100-50 0 50 100 150 Figure adapted from Agrawal, Nonlinear Fiber Optics Frequency Shift (GHz) Institute of Photonics **235** 27.06.2018 Christian Koos and Quantum Electronics



Stability analysis



Ansatz: Continuous-wave solution which is perturbed by a small amplitude perturbation ΔA (z,t)

 $\underline{A}(z,t) = (A_0 + \Delta \underline{A}(z,t)) e^{-j\gamma |A_0|^2 z}$

 \Rightarrow NLSE leads to differential equation for the amplitude perturbation:

$$\frac{\partial \Delta \underline{A}(z,t)}{\partial z} = j \frac{1}{2} \beta_c^{(2)} \frac{\partial^2 \Delta \underline{A}(z,t)}{\partial t^2} - j\gamma |\underline{A}_0|^2 \left(\Delta \underline{A}(z,t) + \Delta \underline{A}^*(z,t) \right)$$

Time- and space-harmonic ansatz:

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$$\begin{pmatrix} -K + \frac{1}{2}\Omega^2 \beta_c^{(2)} + \gamma |\underline{A}_0|^2 & \gamma |\underline{A}_0|^2 \\ \gamma |\underline{A}_0|^2 & K + \frac{1}{2}\Omega^2 \beta_c^{(2)} + \gamma |\underline{A}_0|^2 \end{pmatrix} \begin{pmatrix} C_1 \\ C_2 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$

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 \Rightarrow Solution unstable

Associated gain spectrum:

$$g(\Omega) = \left|\beta_c^{(2)}\right| \Omega \sqrt{\Omega_g^2 - \Omega^2}$$

Gain spectra for three power levels $L_{NL} = 1 \text{ km}$ of the cw signal. The so-called "nonlinear length" is given by Instability Gain (km⁻¹) 5.1 $L_{\rm NL} = \left(\gamma \left|\underline{A}_0\right|^2\right)^{-1}$ 2 km 0.5 5 km 0 -150 -100-50 0 50 100 150 Figure adapted from Agrawal, Nonlinear Fiber Optics Frequency Shift (GHz) Institute of Photonics **237** 27.06.2018 Christian Koos and Quantum Electronics



Modulation instability in optical fibers



- Gain spectrum of modulation instability is (approximately) symmetric with respect to the cw signal
- Our analysis: Modulation instability gain always present to the optical carrier
- Real fiber: Modulation instability gain only visible if it overcomes propagation loss



Figures adapted from Agrawal, Nonlinear Fiber Optics

Modulation instability and Kerr comb generation in optical microresonators





Figures adapted from Nature Photonics 6, 415–416 (2012)



Kerr soliton comb generation and data transmission





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Summary

Summary I



Linear and nonlinear optics

- Maxwell's equations in linear and nonlinear optics
- Linear and nonlinear dielectric polarization
- Kramers-Kronig relations
- Wave propagation in linear and nonlinear optics
- Slowly-varying envelope approximation
- Retarded time frames
- Overview of various second- and third-order nonlinear processes
- Kerr effect and intensity-dependent refractive index
- Parametric and nonparametric processes

The nonlinear optical susceptibility

- Formal definition and tensor notation
- Properties of the nonlinear optical susceptibility tensor
- Spatial symmetry and Neumann's principle
- Contracted notation

Second-order nonlinear effects

- Permittivity and impermeability tensor
- Biaxial, uniaxial, and isotropic crystals



Summary II



- Index ellipsoid
- Wave propagation in anisotropic crystals
- Linear electro-optic effect / Pockels effect
- Electro-optic modulators
- LiNbO₃ modulator: Principle and technical realization
- Mach-Zehnder modulators
- Impact of phase mismatch in various second-order nonlinear effects
- Phase matching concepts: Type-1, type-2, quasi-phase-matching
- The Manley-Rowe relations
- Parametric amplifiers and oscillators

Acousto-optics and photon-phonon interactions

- Elasto-optic effect and tensor representation
- Acousto-optic modulators and related devices
- Acoustic and optical phonons
- Brillouin and Raman scattering
- Raman amplifier and laser

Third-order nonlinearities

- Impact of third-order nonlinearities on transmission links
- Signal propagation in linear and nonlinear optical waveguides



Summary III



- Nonlinear Schrödinger equation and interplay of nonlinearity and dispersion
- Spectral broadening in optical fibers
- Optical solitons





Lecture 14







The walk-off angle can be derived from the indikatrix.

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Frequency Doubling Franken, 1961


Different phase matching strategies





Abb. 4.7.: Typ I - kritische Phasenanpassung

IP



Index ellipsoid / optical indicatrix





 $x_1^2/n_1^2 + x_2^2/n_2^2 + x_3^2/n_3^2 = 1.$

Figure 20.2-1 The index ellipsoid. The coordinates (x_1, x_2, x_3) are the principal axes and n_1 , n_2 , n_3 are the principal refractive indexes. The refractive indexes of the normal modes of a wave traveling in the direction **k** are n_a and n_b .

