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Nonlinear Effects in Optical Fibers and Applications

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Nonlinear Effects in Optical Fibers and Applications

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Nonlinear Optics:

Optical phenomena resulting from interactions of matter with radiation at high optical intensities

Goals:

To provide a background on fundamentals and overview of basic nonlinear optical processes that occur in glasses, emphasizing those most relevant to optical fiber communications and fiber-optic devices

Nonlinear Optical Phenomena

- **Nonlinear optical phenomena arise when the optical field is comparable in magnitude to the internal fields of matter**

  - **Coulomb field in a typical atom: 10V/1Å = 10^9 V/cm**
  - **E-field of day light ($I = ½\ \varepsilon_0 nc|E|^2$ = 1-100 mW/cm^2): E = 5-500 V/cm**
  - **E-field of focused laser beam (1- 10^4 MW/cm^2): $E = 10^6$ - 10^8 V/cm**

    - **In response to large fields, displacement of electrons is not proportional to the field.**
      - Electrons radiate waves at new frequencies
      - Refractive index depends on light intensity
    - **NLO effects are described by a nonlinear polarization**

Optical Polarization

- **Dipole moment per unit volume**

$$\vec{P} = N\vec{\mu} = \sum q_j \vec{r}_j / \mathrm{Vol}$$

Source term of wave equation

$$\left(\vec{\nabla}\times\vec{\nabla}\times + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E}(\vec{r},t) = -\mu_0 \frac{\partial^2}{\partial t^2}\vec{P}(\vec{r},t)$$

The polarization describes all optical phenomena

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Nonlinear polarization

$$P = \varepsilon_0[\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots]$$

$$P = P_L + P_{NL} = P^{(1)} + P^{(2)} + P^{(3)} + \cdots$$

- **Optical susceptibilities**
  - **Linear polarization:** $P_L = P^{(1)} = \varepsilon_0\chi^{(1)}E$
  - **Second order:** $P^{(2)} = \varepsilon_0\chi^{(2)}E^2$
  - **Third order:** $P^{(3)} = \varepsilon_0\chi^{(3)}E^3$
  - ***n*th order:** $P^{(n)} = \varepsilon_0\chi^{(n)}E^n$

In centrosymmetric media

$$\vec{P}(-\vec{E}) = -\vec{P}(\vec{E}) \Rightarrow \chi^{(2n)} = 0$$

$\chi^{(3)}$ is the lowest order nonlinearity in glasses, liquids,…

$\chi^{(n)}$ **: Optical susceptibility of nth order**

(Rigorous definitions will be presented later)

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A quick tour through the catalog of third order nonlinear optical effects

Third Order Nonlinear Optics:

Effects that occur in any material (not necessarily non-centro-symmetric crystals) such as glasses, liquids, gases...

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Third order nonlinearities

Third harmonic generation & optical Kerr effect

$E = \frac{1}{2} E_0 e^{i\omega t} + c.c.$

$P_{3\omega}^{(3)} = \frac{1}{8} \varepsilon_0 \chi^{(3)} E_0^3 e^{i3\omega t} + c.c.$ → 3ω

$P_{\omega}^{(3)} = \frac{3}{8} \varepsilon_0 \chi^{(3)} |E_0|^2 E_0 e^{i\omega t} + c.c.$ → ω

$\chi^{(3)}$ medium

refractive index change

Sum and difference frequency generation

$E = \frac{1}{2}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c.)$

$\chi^{(3)}$ medium

$|2\omega_2 - \omega_1|$

$2\omega_2 + \omega_1$

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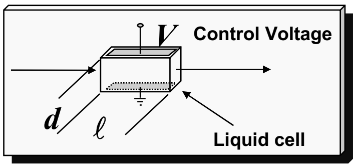
dc Kerr effect

- E_{dc}: **dc or low frequency (<< optical frequency ω)**
- **Quadratic electro-optic effect**

$$E = E_{dc} + (\tfrac{1}{2} E_0 e^{i\omega t} + c.c.)$$

$$P_\omega = \tfrac{1}{2}\varepsilon_0(\chi^{(1)} + 3\chi^{(3)} E_{dc}^2) E_0 e^{i\omega t} + c.c.$$



$$E_{dc} = V/d$$

$$n = n_0 + \Delta n$$

$$\Delta n = 3\chi^{(3)} E_{dc}^2 / 2n_0$$

Discovered by J. Kerr (1875) and used since 1940's in high speed photography
(nanosecond switching time, but needs kVolts)

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Optical Kerr effect

Self effect

$$E = \tfrac{1}{2} E_0 e^{i\omega t} + c.c.$$

$$P = \tfrac{1}{2}\varepsilon_0(\chi^{(1)} + \tfrac{3}{4}\chi^{(3)} |E_0^2|) E_0 e^{i\omega t} + c.c.$$

$$\boxed{\Delta n = \tfrac{3}{8n_0}\chi^{(3)} |E_0^2| \equiv n_2 I}$$

More convenient description $\boxed{n = n_0 + n_2 I}$

Irradiance (W/cm²) $I = \tfrac{1}{2}\varepsilon_0 n_0 c |E_0^2|$

Example: glasses $n_2 = 10^{-15}$ cm²/W, $n_0 = 1.5$, $I = 100$ MW/cm² $\Rightarrow$ $\Delta n = \mathbf{10^{-7}}$

Note: $\Delta\phi = 2\pi\Delta n\,(l/\lambda)$ **and factor** (l/λ) **can be very large**

Pump induced effect

$$E = \tfrac{1}{2}(E_p e^{i\omega_p t} + E_0 e^{i\omega t} + c.c.)$$

$$P_\omega = \tfrac{1}{2}\varepsilon_0(\chi^{(1)} + \tfrac{3}{2}\chi^{(3)} |E_p^2|) E_0 e^{i\omega t} + c.c.$$

$$\boxed{\Delta n = \tfrac{3}{4n_0}\chi^{(3)} |E_p^2| = 2n_2 I_p}$$

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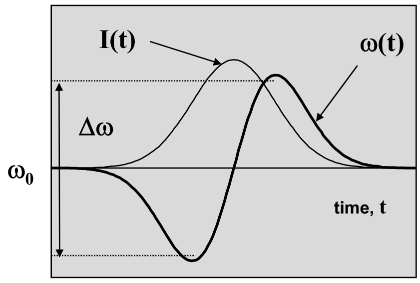
Self phase modulation

- **Spectral broadening of short pulses**

Phase shift $\Delta\phi = -\frac{\omega_0 n_2 z}{c} I(t)$

Instantaneous frequency

$$\omega(t) = \omega_0 - \frac{2\pi n_2 z}{\lambda}\frac{\partial I}{\partial t}$$



Spectral broadening

$$\Delta\lambda \cong \frac{\lambda \ell n_2 I_{max}}{c t_p}$$

Example: SiO_2, λ = 1 µm, $n_2 = 3\times10^{-16}$ cm²/W, ℓ = 1 cm, t_p = 50 fs, $I_{max} = 2\times10^{11}$ W/cm² → **Δλ = 40 nm**

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Four wave mixing

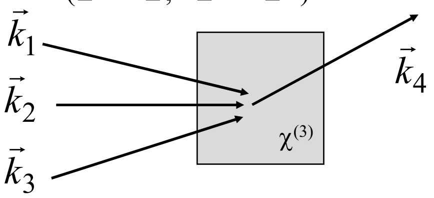
Three incident waves + one generated

$$P^{(3)} = \tfrac{3}{4}\varepsilon_0\chi^{(3)}E_1^{\pm}E_2^{\pm}E_3^{\pm}e^{i(\omega_4 t - \vec{k}_4\cdot\vec{r})} + c.c.$$

$$(E^+ = E; \quad E^- = E^*)$$

$$\omega_4 = \pm\omega_1 \pm \omega_2 \pm \omega_3$$

$$\vec{k}_4 = \pm\vec{k}_1 \pm \vec{k}_2 \pm \vec{k}_3$$



degenerated FWM: $\omega_4 = \omega_1 = \omega_2 = \omega_3 = \omega$

$$P^{(3)} = \tfrac{3}{8}\varepsilon_0\chi^{(3)}E_1E_2E_3^*e^{i(\omega t - \vec{k}_4\cdot\vec{r})}$$

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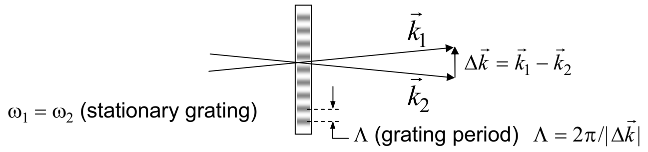
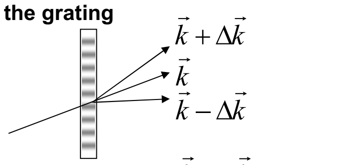
Laser induced gratings

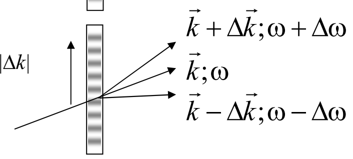
- **Two interfering waves induce a grating (refractive or absorptive)**



- **A third wave scatters (diffracts) in the grating**



- Moving grating: $\Delta\omega = \omega_1 - \omega_2 \neq 0$
  - **grating moves with velocity** $\Delta\omega/|\Delta k|$
  - **inelastic scattering**



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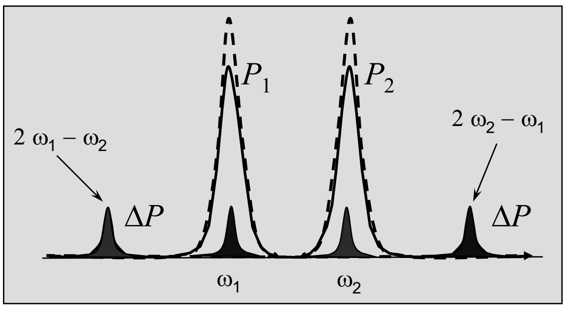
Four Wave Mixing (FWM) in fibers

- **Important effect for WDM systems**



- **Reduces the transmitted power in each channel**
- **Produces cross talk**

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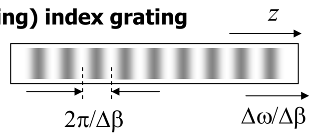
FWM as self-diffraction

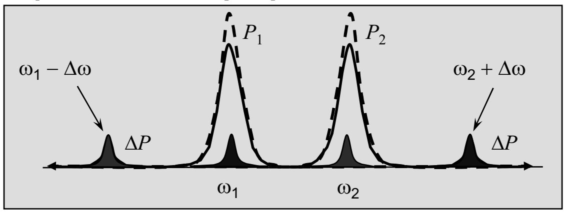
- **Beating of input waves induces a (moving) index grating**

$$\Delta n = \tfrac{3}{4n_0}\chi^{(3)}\left[|E_1^2| + |E_2^2| + (E_1 E_2^* e^{i(\Delta\omega t - \Delta\beta z)} + c.c.)\right]$$



- **A third wave suffers inelastic diffraction**

- **The pump waves are also (Self) diffracted**



FWM and Phase Matching

Wavevector (including SPM and XPM):

Generic case of FWM: $\omega_{ijk} = \omega_i + \omega_j - \omega_k$

Propagation constant $\beta(\omega_i) + \gamma\left(P_i + 2\sum_{k\neq i} P_k\right) L_{eff} / L$

$L_{eff} = (1 - e^{-\alpha L})/\alpha$
(α = loss coefficient)

Wavevector mismatch

$$\Delta\beta = \left[\beta(\omega_k) + \beta(\omega_{ijk}) - \beta(\omega_j) - \beta(\omega_i)\right] + \gamma\left(P_i + 2\sum_{k\neq i} P_k\right) L_{eff} / L$$

FWM efficiency: $$\eta = \frac{\alpha^2}{\alpha^2 + \Delta\beta^2}\left[1 + \frac{4e^{-\alpha L}\sin^2(\Delta\beta L/2)}{\left(1 - e^{-\alpha L}\right)^2}\right]$$

- Very useful for physical insights and rather good estimates
- One of the input tones can be noise: MI, noise amplification,...

Phase conjugation

- **Degenerate four wave mixing with two counter-propagating pump beams and a weak signal beam**

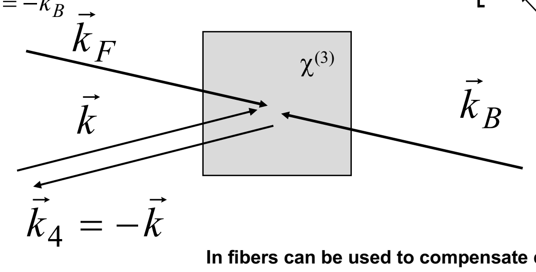
$$P^{(3)} = \frac{3}{8}\varepsilon_0 \chi^{(3)} E_F E_B E^* e^{i[(-\omega+\omega_F+\omega_B)t-(\vec{k}_F+\vec{k}_B-\vec{k}).\vec{r}]} + c.c.$$

$\omega_F = \omega_B = \omega$

$\vec{k}_F = -\vec{k}_B$

$$P^{(3)} = \frac{3}{8}\varepsilon_0 \chi^{(3)} E_F E_B \left[E^* e^{i\vec{k}\cdot\vec{r}} \right] e^{i\omega t} + c.c.$$

Spatial phase conjugate of signal field



In fibers can be used to compensate dispersion and other time-phase distortions

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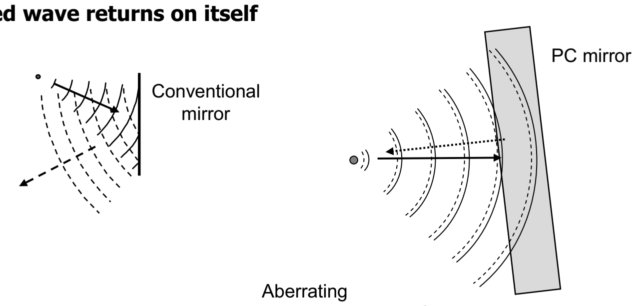
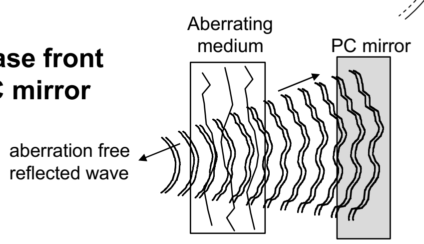
Phase conjugate mirror

- **Reflected wave returns on itself**



Correction of phase front distortions by PC mirror



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Nonlinear absorption

Time averaged absorbed power per unit volume

$$\left\langle \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} \right\rangle = -\frac{\omega \varepsilon_0}{2} |E|^2 (\chi''^{(1)} + \tfrac{3}{4}\chi''^{(3)} |E|^2 + \cdots)$$

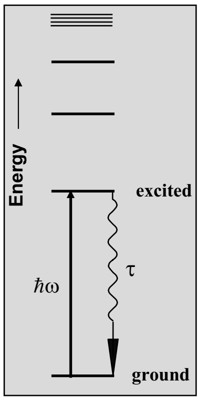
Near resonances $\chi^{(n)}$ is complex ($\chi = \chi' + i\chi''$)

Absorption is described by $\chi''^{(2n-1)}$ (odd orders)

Absorption coefficient

$$\alpha + \Delta\alpha = -\frac{\omega}{nc}(\chi''^{(1)} + \tfrac{3}{4}\chi''^{(3)}|E|^2 + \cdots)$$

- Single photon resonance
  - **Saturation of absorption** $(\chi''^{(3)} > 0)$
  - **Gain saturation (Lasers)** $(\chi''^{(3)} < 0)$



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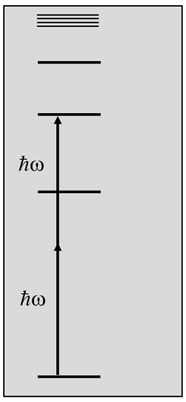
Two photon absorption

$\chi''^{(3)}$ **process** $\Delta\alpha = \beta_{TPA} I$

TPA coefficient

$$\beta_{TPA} = -\frac{3\omega\chi''^{(3)}}{2\varepsilon_0 n^2 c^2}$$

Typically, $\beta_{TPA} = 10^{-10}$ **cm/W**



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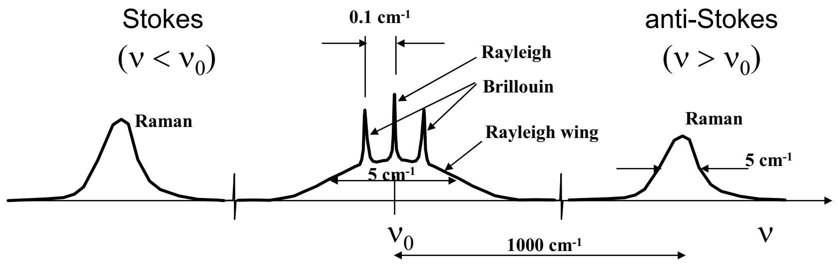
Stimulated Scattering

- (Spontaneous) Scattering Spectrum



Raman: Vibrations/Optical phonons
Brilluoin: Sound waves/Acoustic phonons
Rayleigh wing: Orientational fluctuations (liquids)
Rayleigh: Density (entropy) fluctuations

Lasers can excite strong material oscillations. Scattered light wave interfere with the laser possibly reinforcing the material excitation, thus leading to positive feedback: stimulated scattering.

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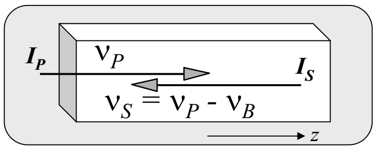
Stimulated Brillouin Scattering (SBS)

- Backward scattering
  - **Coupling to acoustic waves, acoustic phonons**
  - **Acoustic resonance**



$$\frac{dI_S}{dz} = -(g_B I_P - \alpha) I_S - seed$$

(α = loss coefficient)

Seed to initiate process comes from spontaneous scattering

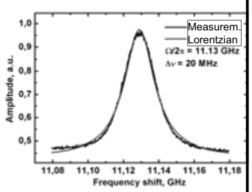
Typical Frequency shift:

$\nu_B \sim 50$ GHz **(glasses)**

$\nu_B = 2\nu_P$ **(sound velocity/light velocity)**

Bandwidth $\Delta\nu_B \sim$ **20 - 100 MHz**

- **Gain intensity factor** $g_B \sim 10^{-9} - 10^{-8}$ **cm/W (glasses)**



Threshold: $I_{th} \cong \frac{21}{g_B L}\left(1+\frac{\Delta\nu_{laser}}{\Delta\nu_B}\right) \sim 10$ MW/cm^2 ($L \sim 1$ m)

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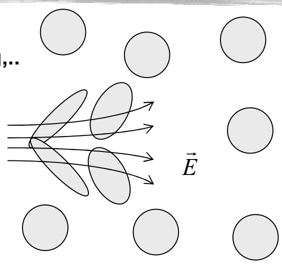
Electrostriction

Electric field gradient attracts polarizable molecules.
Driving force for acoustic waves (SBS), self focusing,..



Force/Volume: $\vec{f} = -\nabla u = \frac{1}{2}\varepsilon_0 N\alpha\nabla E^2$

(take time average over 1 optical cycle) :

Pressure: $p = -\rho\frac{\partial U}{\partial\rho} = -\frac{1}{2}\rho\frac{\partial\varepsilon}{\partial\rho}E^2$

Density change: $\delta\rho = \frac{1}{2}\rho C\gamma_e E^2$

Compressibility — $C = \frac{1}{\rho}\frac{\partial\rho}{\partial p} \approx 10^{-9}\,\mathrm{m}^2/\mathrm{N}$

Electrostriction coefficient — $\gamma_e = \rho\frac{\partial\varepsilon}{\partial\rho} \cong \frac{1}{3}\varepsilon_0(n_0^2-1)(n_0^2+2)$

Refractive index change:

$$\delta n = \frac{\partial n}{\partial\rho}\delta\rho = \frac{\varepsilon_0\left[(n_0^2+2)(n_0^2-1)\right]^2}{72\rho C}E_0^2$$

In glasses, shear acoustic waves may play a role (specially in waveguides)

$$\boldsymbol{f}_{el} = \gamma_{12}\nabla E^2 + 2\gamma_{44}\nabla\cdot\left(\boldsymbol{E}\otimes\boldsymbol{E}\right),$$

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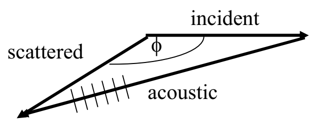
Brillouin frequency

In bulk

$$\omega_{ac}(\phi) = V_S k_{ac} = 2k_{opt}V_S\sin(\phi/2) = \omega_{ac}^{\max}\sin(\phi/2)$$

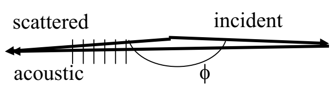


Max. freq. ($\phi = \pi$) ν_B = 12 GHz in silica

$$\omega_{ac}^{\max} = V_S k_{ac} = 2k_{opt}V_S = 2\omega n V_S/c$$

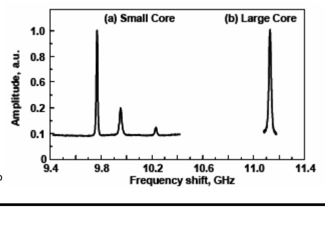
In fibers

Transverse k: $k_T = \sqrt{(\omega n_1/c)^2 - \beta^2} \approx \pi/a$



Max. freq. ν_B = 11 GHz in silica

$$\nu_B = V_S k_{ac} = 2\beta_{opt}V_S = 2\omega n_{eff}V_S/c$$



Dainese et.al., Nature Phys. 2006

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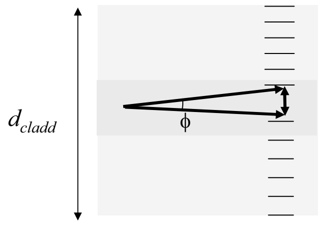
Forward Brillouin scattering

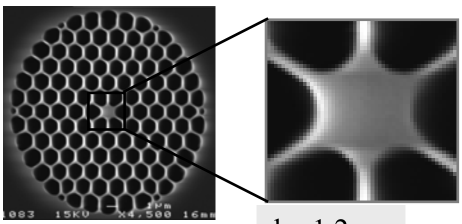
In waveguides ~ transverse acoustic waves can "resonate" with cladding reflections



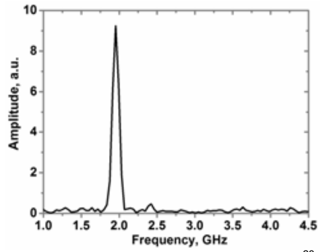
Min. freq. ν_B = 20 MHz in 125 um cladding fibers.
Max freq ~ 400 MHz.

Min. freq. ν_B = 2 GHz in 1 um PCF

$$\omega_{ac} = V_S k_{ac} \approx V_S \pi / d_{clad}$$



d = 1.2 μm



Dainese et.al., Opt. Express 2006

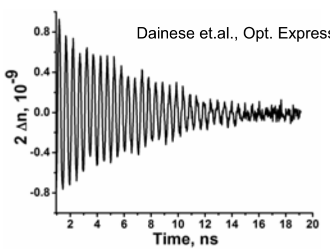
Impulsive Brilouin Scattering

Conventional 125 um cladding fiber

Mouza et.al., PTL 1998

- **Short pulse excites transverse acoustic wave packets that reflects at cladding interface**
- **A weak probe senses refractive index changes each time the wave packet crosses the fiber core (round trip time ~10 ns)**
- **In solid core PCFs (1 um diam) the round trip time is much shorter**

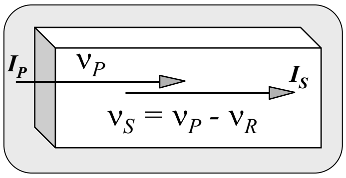
Dainese et.al., Opt. Express 2006



PCF 1.2 um core diam.

Stimulated Raman Scattering (SRS)

- Forward Scattering
- Coupling to molecular vibrations, optical phonons



$$\frac{dI_S}{dz} = (g_R I_P - \alpha) I_S + seed$$

- **Frequency shift**

  ν_R **~ 200 – 500 cm^{-1}**

- **Bandwidth**

  $\Delta\nu$ **~ 1 GHz (gases) – 5 THz (glasses)**

- **Gain intensity factor** $g_R \sim 10^{-12}$ - 10^{-8} cm/W

Threshold: $I_{th} \cong \frac{16}{g_R L}$ ~ 1 GW/cm^2 (L ~ 1 m)

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Formal Theory of Nonlinear Optics

Goals:

- **Understand the general properties of $\chi^{(3)}$**
  - **Symmetries**
  - **Frequency dependencies**
  - **What $\chi^{(3)}$ describes a given NLO effect**
- **Conventions to facilitate reading of literature on NLO**
  - **Different systems of units**

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Theory of nonlinear susceptibilities

Let us start with the linear susceptibility:
What is the most general linear relation between two vector fields (*P* and *E*)?

Then:
What is the most general nonlinear relation between two vector fields (*P* and *E^n^*)?

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Linear constitutive relation

Most general linear relation between vector functions:

- **Time domain**

$$\vec{P}^{(1)}(\vec{r},t)=\varepsilon_0\int_{-\infty}^{\infty}dt'\int_V d^3r'\,\vec{\vec{\chi}}^{(1)}(\vec{r},t,\vec{r}',t')\cdot\vec{E}(\vec{r}',t')$$

tensor (3x3 elements)

Time and space invariance $\vec{\vec{\chi}}^{(1)}(\vec{r},\vec{r}',t,t')\rightarrow\vec{\vec{\chi}}^{(1)}(\vec{r}-\vec{r}',t-t')$

Locality **(good approximation)**

$$\vec{P}^{(1)}(\vec{r},t)=\varepsilon_0\int_{-\infty}^{\infty}dt'\,\vec{\vec{\chi}}^{(1)}(\vec{r},t-t')\cdot\vec{E}(\vec{r},t')$$

Homogeneous media

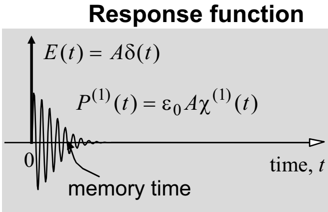
$$\boxed{\vec{P}^{(1)}(\vec{r},t)=\varepsilon_0\int_{-\infty}^{\infty}dt'\,\vec{\vec{\chi}}^{(1)}(t-t')\cdot\vec{E}(\vec{r},t')}$$ ☜

Response function

Properties of the response function

Reality $\vec{P}(t)$ and $\vec{E}(t)$ are real $\Rightarrow\chi(t)$ is real

Causality $\vec{\vec{\chi}}^{(1)}(t-t')=0$ for $t<t'$



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Linear susceptibility

- **Things are more simple in the frequency domain:**
- **Constitutive relation in the frequency domain**

$$\vec{P}^{(1)}(\omega) = \varepsilon_0 \overleftrightarrow{\chi}^{(1)}(\omega) \cdot \vec{E}(\omega)$$

Susceptibility is a frequency domain concept

$$\overleftrightarrow{\chi}^{(1)}(\omega) = \int_{-\infty}^{+\infty} e^{-i\omega t} \overleftrightarrow{\chi}^{(1)}(t) dt$$

Fourier transform pair (convention)

$$\vec{E}(\omega) = \int_{-\infty}^{+\infty} e^{-i\omega t} \vec{E}(t) dt \qquad \vec{E}(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} e^{i\omega t} \vec{E}(\omega) d\omega$$

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Properties of susceptibility

- **Complex function** $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$

Reality of response function ($\chi(t)$) $\chi(-\omega) = \chi^*(\omega)$

Causality of $\chi(t)$
Kramers-Krönig relations
(Hilbert transform pair)
(take Cauchy principal value of integrals)

$$\chi''(\omega) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \chi'(\omega') / (\omega' - \omega)$$

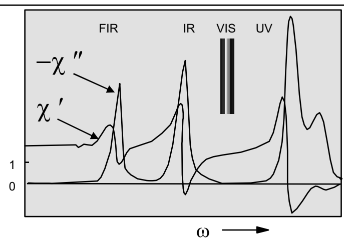
$$\chi'(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \chi''(\omega') / (\omega' - \omega)$$

Complex refractive index
- ***n* = refractive index**
- **κ = extinction coefficient**
- **(absorption coefficient $\alpha = 4\pi\kappa/\lambda$)**

$$(n - i\kappa)^2 = 1 + \chi^{(1)}$$

$$\chi' = n^2 - \kappa^2 - 1 \cong n^2 - 1$$

$$\chi'' = -2n\kappa$$



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Nonlinear susceptibilities

- **General definition**

$$\vec{P}^{(n)}(\omega) = \frac{\varepsilon_0}{(2\pi)^{n-1}} \int d\omega_1 d\omega_2 \cdots d\omega_{n-1} \overleftrightarrow{\chi}^{(n)}(\omega;\omega_1,\omega_2,\cdots\omega_n)\cdot\vec{E}(\omega_1)\vec{E}(\omega_2)\cdots\vec{E}(\omega_n)$$

tensor with 3^{n+1} elements $\qquad (\omega = \omega_1 + \omega_2 + \cdots + \omega_n)$

- **Properties**

Reality $\qquad \overleftrightarrow{\chi}^{(n)}(-\omega;-\omega_1,-\omega_2,\cdots-\omega_n) = \overleftrightarrow{\chi}^{(n)*}(\omega;\omega_1,\omega_2,\cdots\omega_n)$

Kramers-Krönig relations are valid for ω

Intrinsic permutation symmetry:

$$\chi^{(n)}_{ij_1j_2\cdots j_n}(\omega;\omega_1,\omega_2,\cdots\omega_n) = \chi^{(n)}_{ij_2j_1\cdots j_n}(\omega;\omega_2,\omega_1,\cdots\omega_n)$$

invariance under simultaneous exchange of pairs of indices and frequencies... but not (*i*, ω)

Spatial symmetry:

invariance under crystal symmetry operations reduce the number of independent tensor elements

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Global permutation symmetry

Far from resonances (high transparency region):

$\chi^{(n)}$ is real

$\chi^{(n)}$ is invariant when all frequencies are negated

$$\overleftrightarrow{\chi}^{(n)}(\omega;\omega_1,\omega_2,\cdots,\omega_n) = \overleftrightarrow{\chi}^{(n)}(-\omega;-\omega_1,-\omega_2,\cdots,-\omega_n)$$

- **Kleinman symmetry**

$$\chi^{(n)}_{ij_1j_2\cdots k\cdots j_n}(\omega;\omega_1,\omega_2,\cdots\omega_k,\cdots\omega_n) = \chi^{(n)}_{kj_1j_2\cdots i\cdots j_n}(-\omega_k;\omega_1,\omega_2,\cdots-\omega,\cdots\omega_n)$$

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Third order susceptibility $\chi^{(3)}$

$$\vec{P}^{(3)}(\omega) = \frac{\varepsilon_0}{(2\pi)^2} \int d\omega_1 d\omega_2 \vec{\chi}^{(3)}(\omega;\omega_1,\omega_2,\omega_3) \cdot \vec{E}(\omega_1)\vec{E}(\omega_2)\vec{E}(\omega_3)$$

Tensor, 81 elements

$$(\omega = \omega_1 + \omega_2 + \omega_3)$$

- **Properties**

Reality of E and P $\quad \vec{\chi}^{(3)}(-\omega;-\omega_1,-\omega_2,-\omega_3) = \vec{\chi}^{(3)}(\omega;\omega_1,\omega_2,\omega_3)$

Intrinsic Permutation Symmetry

$$\chi^{(3)}_{ijkl}(\omega;\omega_1,\omega_2,\omega_3) = \chi^{(3)}_{ikjl}(\omega;\omega_2,\omega_1,\omega_3) = \cdots$$

Global Permutation Symmetry (Kleinman symmetry)

Far from any resonance

$$\chi^{(3)}_{ijkl}(\omega;\omega_1,\omega_2,\omega_3) = \chi^{(3)}_{jkil}(-\omega_2;\omega_1,-\omega,\omega_3) = \cdots$$

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$\chi^{(3)}$ for isotropic media

General form

- Only 21 non vanishing elements and only 3 independent elements

xxxx = yyyy = zzzz
yyzz = zzyy = zzxx = xxzz = xxyy = yyxx
yzyz = zyzy = zxzx = xzxz = xyxy = yxyx
yzzy = zyyz = zxxz = xzzx = xyyx = yxxy
xxxx= xxyy + xyxy + xyyx

$$\chi^{(3)}_{ijkl} = \chi^{(3)}_{1122}\delta_{ij}\delta_{kl} + \chi^{(3)}_{1212}\delta_{ik}\delta_{jl} + \chi^{(3)}_{1221}\delta_{il}\delta_{jk}$$

- Far from any resonance (Kleinman symmetry valid): only one independent element

xxxx = 3xxyy = 3xyxy = 3xyyx

Third harmonic generation: $\chi^{(3)}_{1122} = \chi^{(3)}_{1212} = \chi^{(3)}_{1221}(3\omega;\omega,\omega,\omega)$

- Only one independent element

Nonlinear Refractive index: $\chi^{(3)}_{1122} = \chi^{(3)}_{1212}(\omega;\omega,\omega,-\omega)$

- Only two independent elements

$$\chi^{(3)}_{ijkl}(\omega;\omega,\omega,-\omega) = \chi^{(3)}_{1122}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + \chi^{(3)}_{1221}\delta_{il}\delta_{jk}$$

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Maker-Terhune Coefficients

General expression of nonlinear polarization for monochromatic field (frequency ω) in isotropic medium

$$\boxed{\vec{P}^{(3)}(t) = \varepsilon_0 A(\vec{E}\cdot\vec{E}^*)\vec{E} + \tfrac{1}{2}\varepsilon_0 B(\vec{E}\cdot\vec{E})\vec{E}^*} \qquad \text{Actual field is } \tfrac{1}{2}(\vec{E}e^{i\omega t} + c.c.)$$

$$A = \tfrac{3}{2}\chi^{(3)}_{1122}$$
$$B = \tfrac{3}{2}\chi^{(3)}_{1221}$$

- Effective "linear" tensor $\boxed{P = \varepsilon_0 \vec{\chi}\cdot\vec{E}}$

$$\chi_{ij} = (A - \tfrac{1}{2}B)|\vec{E}|^2\,\delta_{ij} + \tfrac{1}{2}B(E_i E_j^* + E_j E_i^*)$$

• Molecular orientations:	***B/A* = 3**
• Non-resonant electronic:	***B/A* = 1**
• Electrostriction:	***B/A* = 0**

P.D. Maker and R.W. Terhune, Phys. Rev. 137, A801 (1965).

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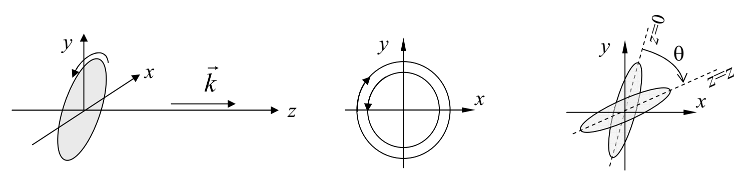
Ellipsoid rotation

Arbitrary polarized field as a sum of left (-) and right (+) circularly polarized waves

$$\vec{E} = E_+\hat{\sigma}_+ + E_-\hat{\sigma}_- \qquad \hat{\sigma}_\pm = (\hat{x} \pm i\hat{y})/\sqrt{2}$$



Then: $P_\pm = \varepsilon_0\chi_\pm E_\pm \qquad \chi_\pm = A|E_\pm|^2 + (A+B)|E_\mp|^2$

and the refractive index difference for the two circularly polarized waves is

$$\Delta n = \frac{B}{\varepsilon_0 n_0^2 c}(I_+ - I_-) \qquad \text{(depends on } B \text{ only)}$$

The polarization ellipse rotates by an angle $\theta = \tfrac{1}{2}\Delta n\omega z / c$

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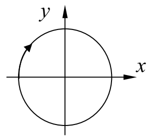
Circularly vs. linearly polarized waves

Circularly polarized waves

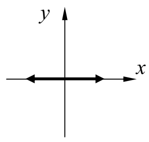
$\vec{E} = E_+\hat{\sigma}_+ \quad \text{or} \quad \vec{E} = E_-\hat{\sigma}_-$



$$\Delta n_{circular} = \frac{A}{\varepsilon_0 n_0^2 c} I_{circular}$$

Linearly polarized waves

$E_+ = E_-$



$$\Delta n_{linear} = \frac{A + B/2}{\varepsilon_0 n_0^2 c} I_{linear}$$

- Molecular orientations: $\Delta n_{linear} / \Delta n_{circular} = 4$
- Non-resonant electronic: $\Delta n_{linear} / \Delta n_{circular} = 3/2$
- Electrostriction: $\Delta n_{linear} / \Delta n_{circular} = 1$

Physical origin of n_2

Physical mechanism	n_2 (cm^2/W)	response time
Electronic (non-resonant)	10^{-16}	fs
Molecular orientation	10^{-14}	ps
Electrostriction	10^{-14}	ns
Thermal	10^{-6}	ms
Population (resonant)	10^{-10}	ns

Examples

Medium	n_2 (cm^2/W)	response time
Air	10^{-19}	fs
SiO_2 glass	3×10^{-16}	< 1 fs
CS_2	10^{-14}	2 ps
GaAs	10^{-6}	20 ns
CdSe doped glasses	10^{-13}	30 ps
Polydiacetylene (resonant)	10^{-8}	2 ps
Polydiacetylene (non-resonant)	10^{-12}	fs

Monochromatic fields

Laser fields are ~ monochromatic

$$\vec{E}(t) = \tfrac{1}{2}\vec{E}_0 e^{i\omega_0 t} + c.c. \qquad \vec{E}(\omega) = \pi\vec{E}_0\delta(\omega-\omega_0) + \pi\vec{E}_0^*\delta(\omega+\omega_0)$$

Mixing of monochromatic waves

$$P^{(3)}_{\omega_4}(t) = \tfrac{1}{2}\varepsilon_0\chi^{(3)}_{eff}E^{(\omega_1)}E^{(\omega_2)}E^{(\omega_3)}e^{i\omega_0 t} + c.c.$$

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

Examples:

generation of wave at $\omega_4 = 2\omega_2 - \omega_1$	$\chi^{(3)}_{eff}(\omega_4) = \frac{3}{4}\chi^{(3)}(\omega_4;-\omega_1,\omega_2,\omega_2)$
THG: $3\omega = \omega + \omega + \omega$	$\chi^{(3)}_{eff}(3\omega) = \frac{1}{4}\chi^{(3)}(3\omega;\omega,\omega,\omega)$
generation of wave at $\omega_4 = 2\omega_1 + \omega_2$	$\chi^{(3)}_{eff}(\omega_3) = \frac{3}{4}\chi^{(3)}(\omega_3;\omega_1,\omega_1,\omega_2)$
Self phase modulation $\omega_1 = \omega_1 + \omega_1 - \omega_1$	$\chi^{(3)}_{eff}(\omega_1) = \frac{3}{4}\chi^{(3)}(\omega_1;\omega_1,\omega_1,-\omega_1)$
Cross phase modulation $\omega_1 = \omega_1 + \omega_2 - \omega_2$	$\chi^{(3)}_{eff}(\omega_1) = \frac{3}{2}\chi^{(3)}(\omega_1;\omega_1,\omega_2,-\omega_2)$
DC Kerr effect $\omega_1 = \omega_1 + 0 - 0$	$\chi^{(3)}_{eff}(\omega_1) = 3\chi^{(3)}(\omega_1;\omega_1,0,0)$

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Systems of units

	Système Internationale, SI	**Gauss [esu(q)]**
electric charge	$1\ \text{C} = 3^*\times10^9\ \text{esu}$	
electric field	$E\ (\text{V/m}) = 3^*\times10^4\ E(\text{esu})$	
constitutive relations	$P = \varepsilon_0\sum\chi^{(n)}E^n$ $D = \varepsilon_0 E + P = \varepsilon E$	$P = \sum\chi^{(n)}E^n$ $D = E + 4\pi P = \varepsilon E$
refractive index	$n = \sqrt{1+\chi}$	$n = \sqrt{1+4\pi\chi}$
susceptibilities	$\chi^{(n)}[(m/V)^{n-1}] = 4\pi/(3^*\times10^4)^{n-1}\chi^{(n)}(esu)$	

$(3^* = 2.9979...)$

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Alternative definitions

- **Different definitions, conventions and units**

$E(t) = E_0 e^{i\omega t} + c.c.$	divide $\chi^{(n)}$ by 2^{n-1}
$E(\omega) = \int E(t) e^{i\omega t} dt$	take complex conjugate of $\chi^{(n)}$
$E(\omega) = \frac{1}{2\pi} \int E(t) e^{-i\omega t} dt$	divide $P^{(n)}(\omega)$ by $(2\pi)^{n-1}$
$E(t) = \int E(\nu) e^{i2\pi\nu t} d\nu$	do nothing
$P_i^{(2)} = d_{il}(EE)_l$	divide d by ε_0
$P^{(n)}(\omega_1 + \cdots \omega_n) = \varepsilon_0 \chi^{(n)} E^{(\omega_1)} \cdots E^{(\omega_n)}$	substitute χ by χ_{eff}
$\mu = \varepsilon_0(\alpha E_{loc} + \frac{1}{2}\gamma^{(2)} E_{loc}^2 + \frac{1}{6}\gamma^{(3)} E_{loc}^3 + \cdots)$	divide $\gamma^{(n)}$ by $n!$
$\vdots$	

Convert to SI units before designing experiments

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Hyperpolarizability

The (local) field acting on a molecule differs from the macroscopic field.

Induced dipole moment per molecule

Linear case $\mu = \varepsilon_0 \ \alpha \ E_{loc}$ $\qquad P = N \ \mu$

α = **Polarizability of molecule**

Nonlinear case

$$\mu = \varepsilon_0 \left[\alpha E_{loc} + \gamma^{(2)} E_{loc}^2 + \gamma^{(3)} E_{loc}^3 + \cdots \right]$$

$\gamma^{(2)}$ = **Second order hyperpolarizability**
$\gamma^{(3)}$ = **Third order hyperpolarizability**

In dilute media (gases), metals, and semiconductors $E_{loc} = E$ and $\chi^{(n)} = N\gamma^{(n)}$

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Local field

Relation between local field acting on the molecule and macroscopic field

Isotropic medium $\vec{E}_{loc} = \vec{E} + \vec{P} / 3\varepsilon_0$

Linear case $\vec{P} = N\varepsilon_0 \alpha \vec{E}_{loc}$

$$\tfrac{1}{3} N\alpha = \frac{\chi}{\chi + 3} = \frac{n^2 - 1}{n^2 + 2}$$ **Lorentz-Lorenz**

$$\vec{E}_{loc} = f\,\vec{E}$$

$$f = \frac{n^2+2}{3}$$ **Local field correction factor**

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Local field correction

- **Nonlinear case**

Total polarization at ω $P = N\varepsilon_0 \alpha E_{loc} + \Delta P = \varepsilon_0 \chi^{(1)} E + f\Delta P$

Nonlinear polarization at ω $P_{NL}(\omega) = f(\omega)\Delta P(\omega)$

Second order

$$\Delta P(\omega) = N\varepsilon_0 \gamma^{(2)}(\omega;\omega_1,\omega_2) E_{loc}(\omega_1) E_{loc}(\omega_2)$$

$$\therefore\ P^{(2)}(\omega) = f(\omega) f(\omega_1) f(\omega_2) N\varepsilon_0 \gamma^{(2)}(\omega;\omega_1,\omega_2) E(\omega_1) E(\omega_2)$$

General case

$$\chi^{(n)}(\omega;\omega_1,\cdots,\omega_n) = f(\omega) f(\omega_1) \cdots f(\omega_2) N\gamma^{(n)}(\omega;\omega_1,\cdots,\omega_n)$$

hyperpolarizability

Use local field factors to convert hyperpolarizability (microscopic) into susceptibility (macroscopic)

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Quantum mechanics theory of NLO

Goals:

- **Understand the connection of $\chi^{(n)}$ to microscopic properties of material systems**
- **NLO effects due to the dependence of populations on intensity**
- **Physical origin of $\chi^{(n)}$**
  - Insights on what makes one material to be more nonlinear than another
  - Importance of resonance and relaxation

Density matrix

- **Statistical quantum mechanics**
- **Expectation of any physical observable such as *P***

$$\vec{P} = N\,\langle\vec{\mu}\rangle = N\,Tr(\vec{\mu}\rho) = N\sum_{kj}\vec{\mu}_{kj}\rho_{jk}$$

ρ is the density operator, given by the Liouville Equation

$$i\hbar\left(\frac{\partial}{\partial t}-\frac{\delta}{\delta t}\right)\rho = [H,\rho]$$

Hamiltonian $H = H_0 - \vec{\mu}\cdot\vec{E}(t)$

Interaction potential

$$i\hbar\left(\tfrac{\partial}{\partial t}-\tfrac{\delta}{\delta t}+i\omega_{jk}\right)\rho_{jk} = \sum_l(\rho_{jl}\vec{\mu}_{lk}-\vec{\mu}_{jl}\rho_{lk})\cdot\vec{E}$$

$$\omega_{jk} = (H_{jj}-H_{kk})/\hbar$$

Relaxation (incoherent interactions, such as collisions)

Relaxation

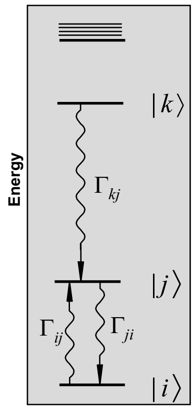
Boltzmann distribution $\rho^{e}_{nn} \propto e^{-E_n/kT}$

Form of relaxation term should restore thermal equilibrium:

Postulate of Random Phases $\rho^{e}_{jk} = 0 \quad (j \neq k)$

Principle of Detailed Balance $\rho^{e}_{kk}\Gamma_{kj} = \rho^{e}_{jj}\Gamma_{jk}$

Γ_{jk} **= Incoherent (spontaneous) transition rate** $j \to k$



Diagonal elements: populations

$$\frac{\delta\rho_{kk}}{\delta t} = \sum_j \Gamma_{jk}\rho_{jj} - \rho_{kk}\Gamma_{kj}$$

Non diagonal elements: coherences

$$\frac{\delta\rho_{jk}}{\delta t} = -\gamma_{jk}\rho_{jk}$$

$\gamma_{jk} = \gamma_{kj}$ **(since ρ must be hermitian)**

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Perturbation theory

- **Perturbative expansion**

$$\rho = \rho^{e} + \rho^{(1)} + \rho^{(2)} + \cdots$$

$$\rho^{(n)} \propto (\vec{\mu}\cdot\vec{E})^{n}$$

Solve by iterations

$$i\hbar\left(\frac{\partial}{\partial t} - \frac{\delta}{\delta t}\right)\rho^{(n)} - [H_0, \rho^{(n)}] = [-\vec{\mu}\cdot\vec{E}, \rho^{(n-1)}]$$

Nonlinear polarization $\boxed{\vec{P}^{(n)} = N\, Tr(\vec{\mu}\rho^{(n)})}$

Note: expressions must be corrected for

1) local field (dielectrics)
2) orientational averaging (amorphous & fluids)
3) inhomogeneous broadening (ex., Doppler effect)

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First order

- **Linear polarizability**

$$\alpha_{jk}(\omega) = \frac{1}{\hbar\varepsilon_0}\sum_{ab}\rho^e_{aa}\left(\frac{\mu^j_{ba}\mu^k_{ab}}{\omega_{ba}-\omega-i\gamma_{ba}} + \frac{\mu^j_{ab}\mu^k_{ba}}{\omega_{ba}+\omega+i\gamma_{ba}}\right)$$

$$\mu^{1,2,3}_{ba} = -e\langle b\,|\,x,y,z\,|\,a\rangle$$

Interpretation: assume isotropic medium. Then

Linear dipole moment: $\langle\vec{\mu}^{(1)}(\omega)\rangle = \varepsilon_0\sum_a \rho^e_{aa}\alpha_a(\omega)\vec{E}(\omega)$

Polarizability of atom in state $|a\rangle$: $\alpha_a(\omega) = \dfrac{e^2}{m\varepsilon_0}\sum_b \dfrac{f_{ba}}{\omega^2_{ba}-\omega^2+i\gamma_{ba}\omega}$

Oscillator strength: $\boxed{f_{ba} = \dfrac{2m}{3\hbar e^2}\omega_{ba}\,|\,\mu_{ba}\,|^2}$ $\quad\left(\langle x^2_{ba}\rangle_{\text{orient}} = \frac{1}{3}\langle r^2_{ba}\rangle_{\text{orient}}\right)$

If all atoms are in the ground state, α coincides with that given by the classical Lorentz oscillator.
f_{ba} is interpreted classically as the "fraction of electrons" with resonance frequency ω_{ba}

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Third order

- **Cubic hyperpolarizability**

$$\gamma^{(3)}_{jklm}(\omega;\omega_1,\omega_2,\omega_3) = \frac{1}{3!\hbar^3\varepsilon_0}\sum_{abcd}\rho^e_{aa}\times$$

$$\frac{\mu^i_{ab}\mu^j_{bc}\mu^k_{ca}\mu^l_{ca}}{(\omega_{ba}-\omega+i\gamma_{ba})(\omega_{ca}-\omega_2-\omega_3+i\gamma_{ca})(\omega_{da}-\omega_3+i\gamma_{da})}$$

$$+\text{ other 47 similar terms}$$

$(\omega = \omega_1+\omega_2+\omega_3)$

Experimentally, frequency and polarization of laser fields are chosen to pick a few terms

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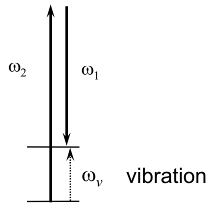
Two Photon Resonances

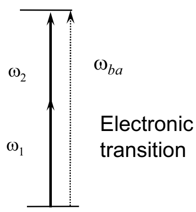
- **Types of resonances of $\chi^{(3)}$**

Raman resonance



TPA resonance

Two photon absorption



(There are many other types of resonances in the 48 terms of $\chi^{(3)}$)

NLO from populations

- **Intensity dependent populations** $\vec{P} = \varepsilon_0 N \sum_n \rho_{nn}(I)\alpha_n \vec{E}$
- **Nonlinear oscillator**

Perturbation theory fails

Example: Two level system: $\Delta N = \dfrac{\Delta N^e}{1 + I / I_{sat}}$

cannot be expanded in power series for $I > I_{sat}$

Populations must be calculated from rate equations

Quantum harmonic oscillator:

oscillator strengths $f_{nk} = n\delta_{n-1,k} - (n+1)\delta_{n+1,k} \quad \Rightarrow \quad \alpha_n = \alpha$

polarizabilities are equal for all levels!

$$\therefore \quad \vec{P} = \varepsilon_0 N \alpha \vec{E} \sum_n \rho_{nn}(I) = \varepsilon_0 N \alpha \vec{E}$$ **LINEAR!**

Nonlinearity is related to anharmonicity of potential

Optical Stark Shift

- **Energy shift due to difference in polarizabilities of two states**

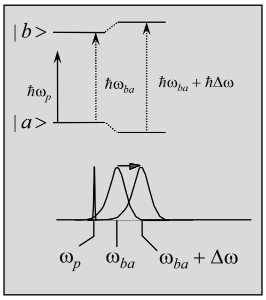
$$\hbar\Delta\omega = -\tfrac{1}{4}\varepsilon_0(\alpha_b - \alpha_a)E_p^2$$

polarizabilities **pump field**

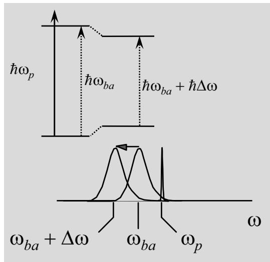
Two level system:

$$\alpha_b = -\alpha_a$$

$$\hbar\Delta\omega = \tfrac{1}{2}\varepsilon_0\alpha_a E_p^2 \approx \frac{|\mu_{ba}|^2(\omega_{ba}-\omega_p)E_p^2}{3\hbar\,[(\omega_{ba}-\omega_p)^2+\gamma^2]}$$



Transition is always pushed away from resonance



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Non-resonant electronic n_2

Result from quantum mechanics $\chi^{(3)}_{1111}(\omega;\omega,\omega,-\omega) = \chi^{(3)}_S + \chi^{(3)}_T$

$$\chi^{(3)}_S = -\frac{8N}{3\varepsilon_0\hbar^3}\sum_{pn}\frac{\mu^x_{gn}\mu^x_{ng}\mu^x_{gp}\mu^x_{pg}}{(\omega_{ng}-\omega)(\omega_{pg}-\omega)(\omega_{pg}-\omega)}$$

Single photon resonance
Optical Stark Shift

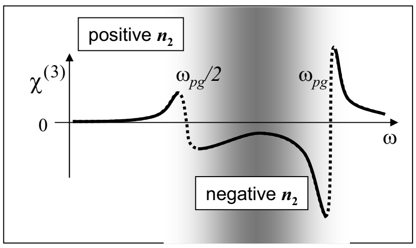
$$\chi^{(3)}_T = \frac{8N}{3\varepsilon_0\hbar^3}\sum_{mnp(\neq g)}\frac{\mu^x_{gn}\mu^x_{np}\mu^x_{pm}\mu^x_{mg}}{(\omega_{ng}-\omega)(\omega_{pg}-2\omega)(\omega_{mg}-\omega)}$$

Two photon resonance

$$\chi^{(3)}_S = -\frac{12N\varepsilon_0}{\hbar}\frac{\partial|\alpha_{xx}|^2}{\partial\omega}$$

$$\chi^{(3)}_T = \frac{24N\varepsilon_0}{\hbar}\sum_{p\neq g}\frac{|\alpha_{p,xx}|^2}{\omega_{pg}-2\omega}$$

$$\alpha_{p,jk} = \frac{1}{3\varepsilon_0\hbar}\sum_{n\neq g}\frac{\mu^j_{gn}\mu^k_{np}}{\omega_{ng}-\omega}$$



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Summary

- Susceptibility is a frequency domain concept
- Symmetries greatly simplify the form of tensors
- Be aware of the definitions, conventions and unit system used in consulted references
- Quantum mechanics gives a microscopic description of nonlinear optics
  - Use local field corrections to calculate susceptibilities from hyperpolarizabilities
- Resonance enhancement of nonlinearities
  - Single photon resonance: Optical Stark Shift
  - Two photon resonances: Two photon absorption, Raman resonance

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Nonlinear Pulse Propagation in Fibers

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Linear pulse propagation

Definitions
Wave equation in the frequency domain
Dispersion, Chirp
Example: gaussian pulse

Nonlinear pulse propagation in n_2 media

Nonlinear Schrödinger equation
Self Phase Modulation, spectral broadening and chirp
Solitons
Propagation regimes

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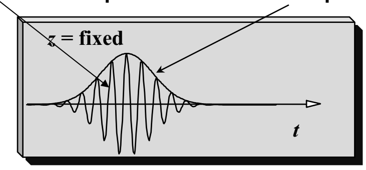
Light pulses

Complex envelope function $\tilde{E}(z,t) = |\tilde{E}(z,t)|\, e^{i\phi(z,t)}$

Real optical field

$$E(z,t) = \tfrac{1}{2}\tilde{E}(z,t)e^{i(\omega_0 t - \beta z)} + c.c. = |\tilde{E}(z,t)|\cos[\omega_0 t - \beta_0 z + \phi(z,t)]$$

envelope **amplitude** **phase**



$\beta_0 = \omega_0 n_0 / c$

$n_0 = n(\omega_0)$

$\omega_0/2\pi$: mean optical frequency
β_0: mean propagation constant

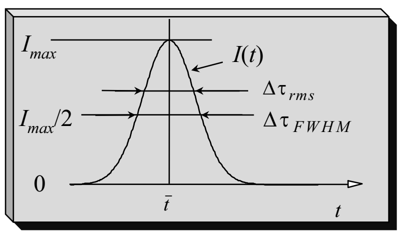
Intensity (instantaneous power/area) $I(z,t) = \dfrac{1}{2}\varepsilon_0 n_0 c |\tilde{E}(z,t)|^2$

Pulse duration

Definitions

rms **duration** $\sigma_t = \Delta\tau_{rms} = \sqrt{\dfrac{\int_{-\infty}^{\infty}(t-\bar{t})^2 I(t)dt}{\int_{-\infty}^{\infty} I(t)dt}}$ $\left(\bar{t} = \dfrac{\int_{-\infty}^{\infty} tI(t)dt}{\int_{-\infty}^{\infty} I(t)dt}\right)$

FWHM **(-3dB) duration:** $\Delta\tau_{rms}$



Wave propagation

General wave equation
Time domain

$$\left(\vec{\nabla}\times\vec{\nabla}\times+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E}(\vec{r},t)=-\mu_0\frac{\partial^2}{\partial t^2}\vec{P}(\vec{r},t)$$

Transverse waves
Isotropic media
Approximate in crystals

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E}(\vec{r},t)=\mu_0\frac{\partial^2}{\partial t^2}\vec{P}(\vec{r},t)$$

Problem: this equation is in the frequency domain, but the constitutive equation is easier in the frequency domain

Frequency domain

$$\left(\nabla^2+\frac{\omega^2}{c^2}\right)\vec{E}(\vec{r},\omega)=-\mu_0\omega^2\vec{P}(\vec{r},\omega)=-\frac{\omega^2}{c^2}\chi^{(1)}\vec{E}(\vec{r},\omega)-\mu_0\omega^2\vec{P}_{NL}(\vec{r},\omega)$$

$$\left[\nabla^2-(\omega n/c)^2\right]\vec{E}(\vec{r},\omega)=-\mu_0\omega^2\vec{P}_{NL}(\vec{r},\omega)$$

$$\left[\chi(\omega)=n^2(\omega)-1\right]$$

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Wave equation in waveguides

The optical field has the form of a mode, $\psi(x,y)$

$$E(\vec{r},\omega)=\psi(x,y)A(z,\omega)e^{-i\beta(\omega)z}$$

The mode amplitude can be normalized such that $\int|\psi(x,y,\omega)|^2dxdy=1$

(ψ also depends on frequency)

Substituting in the wave equation

$$A\left\{\nabla^2_{x,y}\psi+\left[\beta^2-(\omega n/c)^2\right]\psi\right\}+\psi\left(\frac{\partial^2A}{\partial z^2}-2i\beta\frac{\partial A}{\partial z}\right)=-\mu_0\omega^2P_{NL}(\vec{r},\omega)e^{i\beta z}$$

= 0 (since ψ is a mode)

≈ 0 (slowly varying function of z)

Multiply by ψ^*dxdy and integrate

$$2i\beta\frac{\partial A}{\partial z}=\mu_0\omega^2e^{i\beta z}\int P_{NL}(\vec{r},\omega)\psi^*(x,y)dxdy$$

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Most relevant $\chi^{(3)}$ term for fibers

Analytical representation $E(t)=\frac{1}{2}\left[\widehat{E}(t)+\widehat{E}^{*}(t)\right]$; $P(t)=\frac{1}{2}\left[\hat{P}(t)+\hat{P}^{*}(t)\right]$

$$\widehat{E}(\nu)=\begin{cases}2E(\nu) & \text{for } \nu\geq 0\\ 0 & \text{for } \nu<0\end{cases} \qquad (\nu=\omega/2\pi)$$

Only positive frequencies need be considered

$$\widehat{P}^{(3)}(\nu)=\tfrac{1}{4}\varepsilon_0\int\underbrace{d\nu_1 d\nu_2\chi^{(3)}_{SUM}\widehat{E}(\nu_1)\widehat{E}(\nu_2)\widehat{E}(\nu_3)}_{\nu=\nu_1+\nu_2+\nu_3}+\tfrac{3}{4}\varepsilon_0\int\underbrace{d\nu_1 d\nu_2\chi^{(3)}_{DIF}\widehat{E}(\nu_1)\widehat{E}(\nu_2)\widehat{E}^{*}(-\nu_3)}_{\nu=\nu_1+\nu_2-\nu_3}$$

3rd order sum term $\chi^{(3)}_{SUM}=\chi^{(3)}(\nu;\nu_1,\nu_2,\nu_3)$ **describes THG and high frequency sum generation**

3rd order difference term $\chi^{(3)}_{DIF}=\chi^{(3)}(\nu;\nu_1,\nu_2,-\nu_3)$ **describes FWM, n_2, SBS, SRS (everything that is most relevant in fibers).**

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No $\chi^{(3)}$ dispersion

If the input field spectrum is narrow-band we can ignore the frequency dependence in $\chi^{(3)}$ (valid far from resonances)

$$P^{(3)}(\nu)=\tfrac{3}{4}\varepsilon_0\chi^{(3)}\int\widehat{E}(\nu_1)\widehat{E}(\nu_2)\widehat{E}^{*}(\nu-\nu_1-\nu_2)d\nu_1 d\nu_2$$

(use convolution theorem)

$$\widehat{P}^{(3)}(t)=\tfrac{3}{4}\varepsilon_0\chi^{(3)}\,|\widehat{E}(t)|^2\,\widehat{E}(t)$$

The nonlinear polarization is remarkably simple in the time domain…

But, WARNING, this is true for nonresonant electronic $\chi^{(3)}$. (Not true for electrostriction, Raman, Brillouin,… if the field spectrum is broad – such as fs pulses or DWDM systems).

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SVEA

$$E(z,t)=\tfrac{1}{2}\tilde{E}(z,t)e^{i(\omega_0 t-\beta_0 z)}+c.c.$$

Slowly Varying Envelope Approximation

$$\left|\frac{\partial\tilde{E}}{\partial t}\right| << \left|\omega_0\tilde{E}\right| \text{ and } \left|\frac{\partial\tilde{E}}{\partial z}\right| << \left|\beta_0\tilde{E}\right|$$

Envelope varies very little in time over one optical period or in space over one wavelength

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Linear Propagation

Wave equation in frequency domain

$$\left(\frac{\partial^2}{\partial z^2}-\frac{\omega^2}{c^2}\right)E(z,\omega)=-\mu_0\omega^2 P(z,\omega)$$

Transparent, homogeneous and isotropic media

$$P(z,\omega)=\varepsilon_0[n^2(\omega)-1]\,E(z,\omega) \qquad \left[\chi(\omega)=n^2(\omega)-1\right]$$

Exact solution $$E(z,\omega)=E(0,\omega)e^{-i\beta(\omega)z}$$

Dispersion relation $\beta(\omega)=\omega\, n(\omega)/c$

Fourier transform

$$\vec{E}(\omega)=\int_{-\infty}^{+\infty}e^{-i\omega t}\vec{E}(t)dt \qquad \vec{E}(t)=\tfrac{1}{2\pi}\int_{-\infty}^{+\infty}e^{i\omega t}\vec{E}(\omega)d\omega$$

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Group velocity

Dispersion relation - Taylor series expansion

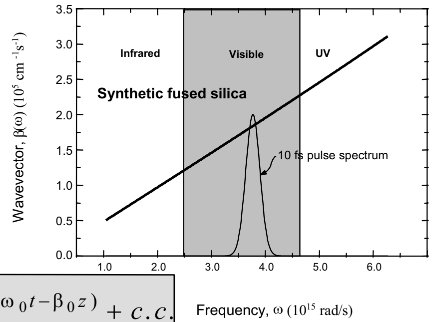
$$\beta(\omega)=\beta_0+\beta_1(\omega-\omega_0)+\tfrac{1}{2}\beta_2(\omega-\omega_0)^2+\tfrac{1}{3!}\beta_3(\omega-\omega_0)^3+\cdots$$

$$\beta_n=\left(\frac{d^n\beta}{d\omega^n}\right)_{\omega_0}$$

Phase and group velocities

$$v_{phase}=\omega_0/\beta_0=c/n(\omega_0)$$

$$v_{group}=v_g=1/\beta_1$$



If $\beta(\omega)=\beta_0+\beta_1(\omega-\omega_0)$
$(\beta_2=0,\ \beta_3=0,\ldots)\ \Rightarrow$

$$E(z,t)=\tfrac{1}{2}\tilde{E}(0,t-z/v_g)e^{i(\omega_0 t-\beta_0 z)}+c.c.$$

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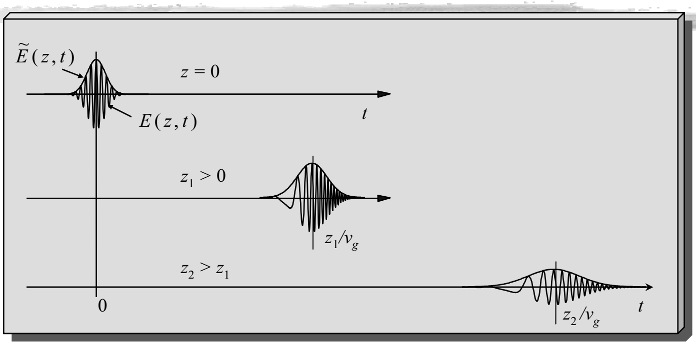
Pulse propagation in dispersive media



Equation for envelope function

Frequency domain $$\frac{\partial}{\partial z}\tilde{E}(z,\omega-\omega_0)=-i[\beta(\omega)-\beta_0]\tilde{E}(z,\omega-\omega_0)$$

Time domain $$\frac{\partial\tilde{E}(z,t)}{\partial z}=\left(\beta_1\frac{\partial}{\partial t}+i\frac{\beta_2}{2!}\frac{\partial^2}{\partial t^2}+\frac{\beta_3}{3!}\frac{\partial^3}{\partial t^3}+\cdots\right)\tilde{E}(z,t)$$

Equations in time domain are complicated!

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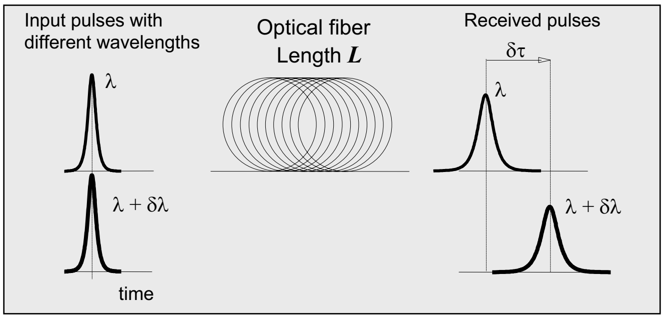
Dispersion parameter

Used in fiber optics

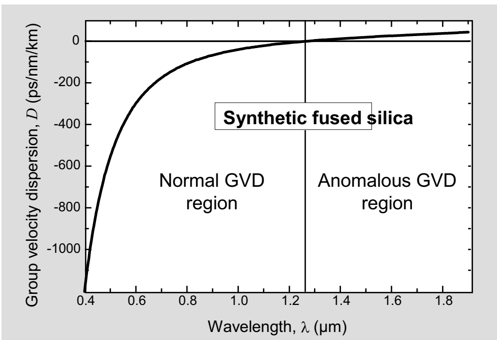


Dispersion parameter: $D = \frac{1}{L}\frac{\delta\tau}{\delta\lambda}$ [ps/nm/km]

Relation between D and β_2: $\beta_2 = -\frac{\lambda_0^2}{2\pi c} D$

GVD in silica

Optical fibers for communications are made of silica



Transparent materials exhibit a particular λ_{ZD} where $D(\lambda_{ZD}) = 0$.
In pure silica, λ_{ZD} = 1.27 μm (Zero Dispersion Wavelength).

Pulse broadening

Example: input Gaussian pulse $E(0,t) = \frac{1}{2} E_0 \, e^{-(t/2\sigma_0)^2} \, e^{i\omega_0 t} + c.c.$

(σ_0 = rms pulse duration; $\Delta\tau_{FWHM} = (8 \ln 2)^{1/2} \sigma_0$)

Go to the frequency domain, solve the wave equation, and return to time domain

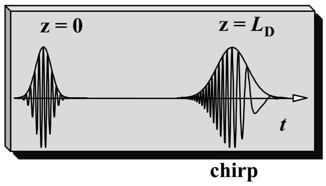
$$\tilde{E}(z,t) = \frac{E_0}{\sqrt{1 + iz/z_D}} \exp(-\tau^2/4\sigma^2) \exp(iz\tau^2/4z_D\sigma^2)$$

Pulse continues Gaussian but with different pulse duration and peak amplitude

$\tau = t - \beta_1 z$ — **τ : "local time"**

$\sigma = \sigma_0 \sqrt{1 + (z/L_D)^2}$ — **σ : pulse duration at z**

$z_D = \sigma_0^2 / \beta_2$ — **$L_D = |z_D|$: "dispersion length"**



Dispersion and Chirp

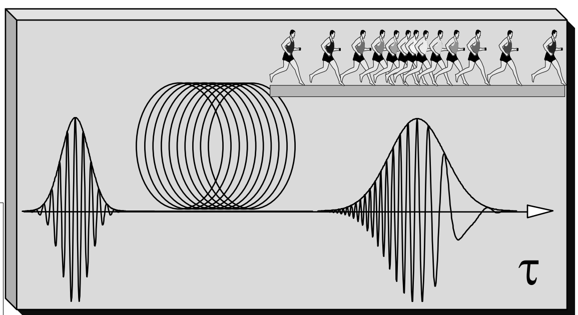
- **Because of GVD the pulse becomes chirped**

Instantaneous frequency

$$\omega(\tau) = \omega_0 + \frac{\partial\phi(z,\tau)}{\partial\tau}$$

Chirp parameter

$$\left(\frac{\partial\omega}{\partial\tau}\right)_{GVD} \cong -\frac{z\lambda^2 D}{2\pi c \sigma_0^4}$$



(can be very large for femtosecond pulses)

Pulse propagation in n_2 media

We know the linear polarization in the frequency domain

$$P(\omega,z) = \varepsilon_0[n^2(\omega)-1]\,E(\omega,z)$$

but P_{NL} in the time domain

$$P_{NL}(t,z) = \varepsilon_0\chi^{(3)}_{eff}\left|\tilde{E}(t,z)\right|^2\,E(t,z) \qquad \chi^{(3)}_{eff} = n_0^2 c n_2/\varepsilon_0$$

(far from any resonance $\chi^{(3)}$ is ~ independent of frequency: Δn responds instantaneously to the pulse envelope)

Propagation equation is solved in split steps: frequency domain for the linear part and time domain for the nonlinear part (Split Step Fourier method)

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Wave equation for short pulses

- **SVEA, GVD, and n_2 (nonresonant electronic)** $\left[\Gamma = \omega_0\chi^{(3)}_{eff}/2n_0 c\right]$

Wave equation with GVD

$$\frac{\partial}{\partial z}\tilde{E} = \left(\beta_1\frac{\partial}{\partial t} + i\frac{\beta_2\partial^2}{2!\partial t^2} + \cdots\right)\tilde{E} - i\Gamma\,|\tilde{E}|^2\,\tilde{E}(z,t)$$

In moving frame ($z' = z$, $\tau = t - z/v_g$)

$$\frac{\partial}{\partial z'}\tilde{E} = \underbrace{-i\left(\frac{\beta_2\partial^2}{2!\partial\tau^2} + i\frac{\beta_3\partial^3}{3!\partial\tau^3} + \frac{\beta_4\partial^4}{4!\partial\tau^4}\cdots\right)\tilde{E}}_{\text{Dispersion terms}}\ \underbrace{-\,i\Gamma\,|\tilde{E}|^2\,\tilde{E}(z',\tau)}_{\text{Nonlinear term}}$$

Usually β_2 is enough (Nonlinear Schrödinger Equation).
Loss, Raman, $\chi^{(3)}$ dispersion add terms to this equation.

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Self Phase Modulation and Chirp

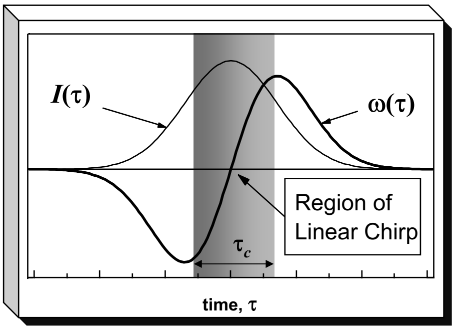
- **Instantaneous frequency of the pulse** $\omega(z', \tau) = \omega_0 - \frac{\omega_0 n_2 z'}{c} \frac{\partial I}{\partial \tau}$
- **Pulse shape is approximately parabolic near its peak**

$$I(\tau) \approx \frac{P_c}{A_{eff}}\left(1 - \frac{\tau^2}{2\tau_c^2}\right)$$

(P_c and τ_c: characteristic peak power and duration of light pulse)

- **This gives a ~linear chirp**

☞ $$\left(\frac{\partial \omega}{\partial \tau}\right)_{SPM} \cong \frac{\omega_0 n_2 z' P_c}{c A_{eff} \tau_c^2}$$



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Spectral broadening by SPM

Pulse shape is preserved

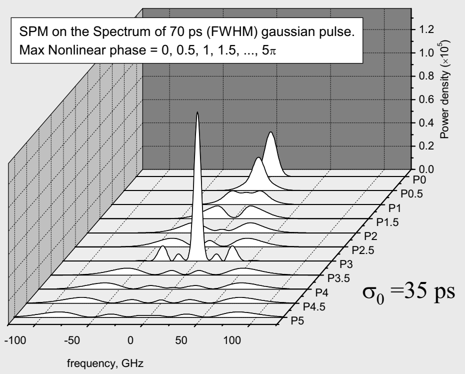
but spectrum changes (phase is a function of time and z):

$$\tilde{E}(z,\tau) = \tilde{E}(0,\tau) e^{i\Gamma|\tilde{E}(0,\tau)|^2 z'}$$

Maximum nonlinear phase

$$\phi_{max} = \Gamma E_0^2 z'$$

Initial pulse ($z = 0$): $\tilde{E}(0,t) = E_0 e^{-t^2/4\sigma_0^2}$



Spectra as a function of position (z) for fixed input power, or as a function of power for fixed z.

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Nonlinear Schrödinger equation

Wave equation for dispersive, nonlinear media

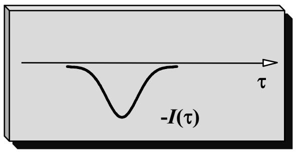
$$i\frac{\partial \tilde{E}}{\partial z'} = -\frac{\beta_2}{2}\frac{\partial^2 \tilde{E}}{\partial \tau^2} + \Gamma|\tilde{E}|^2\,\tilde{E}$$

Nonlinear Schrödinger Equation (NLSE)

Neglects third and higher order dispersion

Looks like Schrödinger equation for a particle of mass m in potential V:

$$i\frac{\partial \psi}{\partial t} = -\frac{\hbar}{2m}\frac{\partial^2 \psi}{\partial x^2} + \frac{V}{\hbar}\psi$$



Position and time exchanged. If $\beta_2 < 0$ and $n_2 > 0$ then the pulse forms a bounding potential. Pulse (particle) is trapped in time (space).

"Eigenstates" of NLSE are solitons.

If $\beta_2 < 0$ and $n_2 > 0$ (or $\beta_2 > 0$ and $n_2 < 0$) we have "bright solitons".
If $\beta_2 > 0$ and $n_2 > 0$ ($\beta_2 < 0$ and $n_2 < 0$) we can have "dark solitons".

Particular cases

Simple solutions of NLSE

$$i\frac{\partial \tilde{E}}{\partial z'} = -\frac{\beta_2}{2}\frac{\partial^2 \tilde{E}}{\partial \tau^2} + \Gamma|\tilde{E}|^2\,\tilde{E}(z',\tau)$$

1 - Linear case ($\Gamma = 0$)

Easily solved in frequency domain

$$\tilde{E}(z',\omega) = \tilde{E}(0,\omega)e^{\frac{i}{2}\beta_2\omega^2 z'}$$

2 - Purely nonlinear case (no dispersion)

Easily solved in time domain

$$\tilde{E}(z',\tau) = \tilde{E}(0,\tau)e^{-i\Gamma|\tilde{E}(0,\tau)|^2 z'}$$

3 - Soliton case

If $\tilde{E}(0,\tau) = \sqrt{-\beta_2/\Gamma\tau_c^2}\,\mathrm{sech}(\tau/\tau_c)$

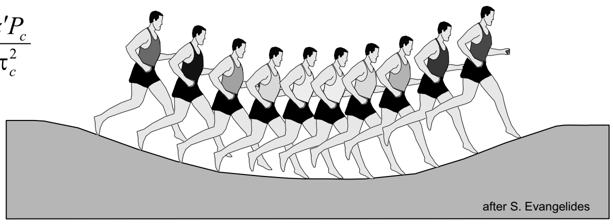
then $\tilde{E}(z',\tau) = \tilde{E}(0,\tau)e^{i(\beta_2/2\tau_c^2)z'}$

Solitons

- Exact cancellation of GVD and SPM chirps
- Good side of nonlinearities in communication systems
- Occur only in the anomalous dispersion region (*D* > 0)

$$\left(\frac{\partial\omega}{\partial\tau}\right)_{GVD} \cong -\frac{z\lambda^2 D}{2\pi c\tau_c^4}$$

$$\left(\frac{\partial\omega}{\partial\tau}\right)_{SPM} \cong \frac{\omega_0 n_2 z' P_c}{cA_{eff}\tau_c^2}$$



after S. Evangelides

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Soliton stability

- Assume an input pulse $E(0,\tau) = (1+\varepsilon)E_s \ \mathrm{sech}(\tau/\tau_c) \quad (|\varepsilon| < 0.5)$

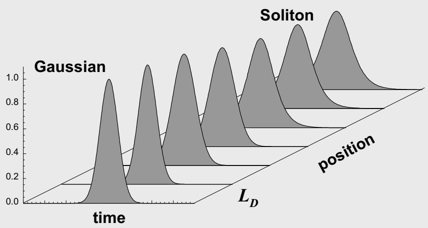
(which is not a soliton unless ε = 0)

$E_s = \sqrt{-\beta_2 / \Gamma\tau_c^2}$

THEN, if |ε| < 0.5, the pulse evolves into a soliton!

- **The soliton is stable against amplitude variations of ±50%**

- **Any reasonable pulse with enough amplitude becomes a soliton!!**



- **Soliton are (the only) exact, stationary solutions of NLSE**

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Propagation regimes

Characteristic lengths:

Dispersion length: $L_D = |z_D| = \tau_c^2/|\beta_2|$
depends on pulse duration (τ_c) and dispersion parameter

Nonlinear length: $L_{NL} = \lambda / 2\pi n_2 I$
depends on peak pulse intensity and nonlinear parameter

For a given fiber length, L, there are four possibilities:

1) $L \ll L_D$ and $L \ll L_{NL}$: **no pulse distortion**
2) $L \gg L_D$ and $L \ll L_{NL}$: **pulse broadens (Dispersive Regime)**
3) $L \ll L_D$ and $L \gg L_{NL}$: **spectral broadening (Nonlinear Regime)**
4) $L \geq L_D$ and $L \geq L_{NL}$: **Soliton Regime**

Typical values for optical fibers at λ = 1.5 µm:
β_2 = -20 ps^2/km, n_2 = 3x10^{-16} cm^2/W.
For τ_c = 10 ps and 50 mW peak power in 80 µm^2 effective area we have:
L_D = 5 km and L_{NL} = 12 km

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