Physical Optics

Lecture 9: Nonlinear Optics

2018-12-20

Michael Kempe
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K = Kempe  
G = Gross
**Linear Photon-Matter Interaction**

- **Refractive index**
  \[ n = \pm \sqrt{\varepsilon_r \mu_r} \]

- **Relative electric permittivity**
  \[ \varepsilon_r = 1 + \chi \]
  \( \chi \): electric susceptibility

- **Displacement field**
  \[ \vec{D} = \varepsilon_0 \varepsilon_r \vec{E} \]
  \[ \vec{D} = \varepsilon_0 \vec{E} + \vec{P} \]

- **Polarisation density**
  \[ \vec{P} = \varepsilon_0 \chi \vec{E} \]

\[ [E] = \frac{V}{m} \quad [D] = \frac{C}{m^2} \]
Nonlinear Photon-Matter Interaction

Linear regime

Nonlinear regime

Nonlinearity scales with atomic field $E_a \approx 10^{10} \text{ V/m}$ corresponding to $P = 10^{14} \text{ W/cm}^2$

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

$P^{(1)}(t) = \varepsilon_0 \chi^{(1)} E(t)$

linear polarisation

$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E^2(t)$

$2^{\text{nd}}$ order nonlinear polarisation

$P^{(3)}(t) = \varepsilon_0 \chi^{(3)} E^3(t)$

$3^{\text{rd}}$ order nonlinear polarisation

$\chi^{(1)} \approx 1$

$\chi^{(2)} \approx 2 \times 10^{-12} \frac{m}{V}$

$\chi^{(3)} \approx 4 \times 10^{-24} \frac{m^2}{V^2}$
Nonlinear Effects

\[ \chi^{(2)} = 0 \quad \text{inversion-symmetric materials (gases, glasses, fluids, \ldots)} \]

\[ \chi^{(3)} \neq 0 \quad \text{materials with and without inversion symmetry (universal)} \]

- second-harmonic generation
- sum-frequency generation
- difference-frequency generation
- optical parametric oscillation

- third-harmonic generation
- self-focussing
- saturable absorption
- two-photon absorption
- stimulated Raman scattering
- 4-wave mixing (CARS)
Nonlinear Wave Equation

\[ \nabla^2 E - \frac{1}{c^2} \frac{\partial E}{\partial t^2} = 0 \]

wave equation in linear optical media

\[ \nabla^2 E - \frac{n_0^2}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P^{NL}}{\partial t^2} \]

wave equation in nonlinear optical media

Linear response

Nonlinear response: source term

Often assumption of weak depletion of field that creates nonlinear polarization is made (first Born approximation)

Source: Saleh/Teich
Second-Order Polarization

2\textsuperscript{nd} order polarisation

Electric field

\[ P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E^2(t) \]

\[ E(t) = E \cdot e^{-i\omega t} + E^* \cdot e^{i\omega t} \]

\[ P^{(2)}(t) = 2\varepsilon_0 \chi^{(2)} E E^* + \left( \varepsilon_0 \chi^{(2)} E^2 e^{-i2\omega t} + \varepsilon_0 \chi^{(2)} E^* E e^{i2\omega t} \right) \text{c.c.} \]

optical rectification

2\textsuperscript{nd} harmonic frequency radiation

Source: Saleh/Teich
Second-Order Polarization

electric field

\[ E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c. \]

2\textsuperscript{nd} order polarisation

\[ P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E^2(t) \]

\[ P^{(2)}(t) = \varepsilon_0 \chi^{(2)} \left[ E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1+\omega_2)t} + 2E_1 E_2^* e^{-i(\omega_1-\omega_2)t} + c.c. \right] + 2\varepsilon_0 \chi^{(2)} \left[ E_1 E_1^* + E_2 E_2^* \right] \]

\[ P(2\omega_1) = \varepsilon_0 \chi^{(2)} E_1^2 e^{-i2\omega_1 t} \]

\[ P(2\omega_2) = \varepsilon_0 \chi^{(2)} E_2^2 e^{-i2\omega_2 t} \]

\[ P(\omega_1 + \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1 E_2 e^{-i(\omega_1+\omega_2)t} \]

\[ P(\omega_1 - \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1 E_2^* e^{-i(\omega_1-\omega_2)t} \]

\[ P(0) = 2\varepsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*) \]

\begin{align*}
\{ & \text{second harmonic generation (SHG)} \\
& \text{sum frequency generation (SFG)} \\
& \text{difference frequency generation (DFG)} \\
& \text{optical rectification (OR)} \}
\end{align*}
Third-Order Polarization

electric field

\[ E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + c.c. \]

3\textsuperscript{rd} order polarisation

\[ P^3(t) = \varepsilon_0 \chi^{(3)} E^3(t) \]

contains 22 different frequency components:

\[ \omega_1, \omega_2, \omega_3, 3\omega_1, 3\omega_2, 3\omega_3 \]

Third harmonic generation

Sum and difference frequency generation

\[ (\omega_1 + \omega_2 + \omega_3), (\omega_1 + \omega_2 - \omega_3), (\omega_1 + \omega_3 - \omega_2), (\omega_2 + \omega_3 - \omega_1) \]

Four-wave mixing

\[ (2\omega_1 \pm \omega_2), (2\omega_1 \pm \omega_3), (2\omega_2 \pm \omega_1), (2\omega_2 \pm \omega_3), (2\omega_3 \pm \omega_1), (2\omega_3 \pm \omega_2) \]
Phase Matching

Energy and momentum conservation:

(1) frequency matching condition

\[ \omega_2 = 2\omega_1 \quad \text{SHG} \]

\[ \omega_3 = \omega_1 + \omega_2 \quad \text{SFG} \]

(2) phase matching condition

\[ k_2 = 2k_1 \quad \text{SHG} \]

\[ k_3 = k_1 + k_2 \quad \text{SFG} \]

a) Conservation of energy

\[ \omega_2 \]

\[ \omega_3 \]

\[ \omega_1 \]

b) Conservation of momentum

\[ k(\omega_1) \quad k(\omega_2) \]

\[ k(\omega_3) \]
Phase Matching in SHG

\[ k_2 = 2k_1 \quad \omega_2 = 2\omega_1 \]

\[ \frac{n_2\omega_2}{c_0} = 2\frac{n_1\omega_1}{c_0} \]

\[ n_2(\omega_2)\omega_2 = 2n_1(\omega_1)\omega_1 \]

\[ n(2\omega_1)2\omega_1 = 2n(\omega_1)\omega_1 \]

same material

different frequency

normal dispersive material

birefringent uniaxial crystal

\[ k = \frac{n\omega}{c_0} \]
Phase Matching by Birefringence

Type I oo-e

\[ 2n_o(\omega) = 2n_e(2\omega) \quad \text{or} \quad 2n_o(\lambda) = 2n_e(\lambda / 2) \]

Type I ee-o

\[ 2n_e(\omega) = 2n_o(2\omega) \]

Type II oe-e

\[ n_o(\omega) + n_e(\omega) = 2n_e(2\omega) \quad \text{or} \quad n_o(\lambda) + n_e(\lambda) = 2n_e(\lambda / 2) \]
Phase Mismatch in SHG

\[
I(2\omega) = \frac{2^7 \pi^3 \omega^2 |\chi^{(2)}|^2}{(n \cdot c)^3} \cdot I(\omega)^2 L^2 \cdot \text{sinc}^2 \left( \frac{\Delta k \cdot L}{2} \right)
\]

\[
\Delta k = \frac{4\pi}{\lambda_1} (n_2 - n_1)
\]

Intracavity SHG

- Pump beam 808 nm
- Mirror $M_1$
- Nd:YAG crystal
- Resonator beam 1064 nm
- KTP crystal
- Mirror $M_2$
- Doubled output beam 532 nm
Quasi-Phase Matching in SHG

- Change of index together with phase sign after length
- Achieved by poling of crystals, e.g. lithium niobate (LiNbO$_3$)

$$\Delta z = \frac{\pi}{\Delta k}$$
Parametric and Non-Parametric Processes

**Parametric Process**
No transfer of energy

\[ n = \sqrt{\mu_r (1 + \chi)} \]

- Virtual state
- Ground state

**Non-Parametric Process**
Transfer of energy

\[ \tilde{n} = n + i\kappa \]

- Second harmonic generation (SHG)
- Sum frequency generation (SFG)
- Third harmonic generation (THG)
- Coherent anti-Stokes Raman scattering (CARS)

- Two photon excitation fluorescence (TPEF)
- Stimulated Raman scattering (SRS)
Second-Order Parametric Processes

a) OFC

![Diagram](image)

- Signal $\omega_1$
- Pump $\omega_2$

Frequency up-conversion

b) OPA

![Diagram](image)

- Signal $\omega_1$
- Pump $\omega_3$
- Idler $\omega_2$

Parametric amplification

c) OPO

![Diagram](image)

- Pump $\omega_3$
- Mirror

Parametric oscillation
Index of refraction depends on intensity

\[ n = n_o + n_2 \cdot I \]

\[ n_2 \cdot I = \frac{6 \cdot \text{Re}\left(\chi^{(3)}\right) \cdot |E|^2}{8 \cdot c \cdot \varepsilon_o n_o^2} \]

### Material properties

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<th>( n_2 ) (cm(^2)/W)</th>
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<tr>
<td>Glas BK7</td>
<td>( 5 \cdot 10^{-15} )</td>
</tr>
<tr>
<td>Water</td>
<td>( 10^{-16} )</td>
</tr>
<tr>
<td>Doped fiber</td>
<td>( 10^{-10} )</td>
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### Propagation effects:

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<th>Effect</th>
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<tr>
<td>focussed onto medium surface</td>
<td>channelling, self trapping</td>
</tr>
<tr>
<td>Collimated</td>
<td>self focussing, then trapping</td>
</tr>
<tr>
<td>Focussed</td>
<td>faster focussing, then trapping</td>
</tr>
<tr>
<td>weak divergent</td>
<td>self focussing, then trapping</td>
</tr>
<tr>
<td>strong divergent</td>
<td>lowering of the divergence</td>
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Third-Order: Two-Photon Absorption

- Simultaneous absorption of two photons through a virtual state → absorption with photons with half the energy compared to single photon absorption
- As all non-parametric processes it cannot be described well by a susceptibility
- In a fluorophore it can result in fluorescence

\[ \sigma(\nu) : \text{transition cross section} \]

\[ P_{abs} = \phi \cdot \sigma(\nu) \]

\[ P_{tpa} = \phi^2 \cdot \sigma_2 (\nu) \]

\[ \sigma \approx 10^{-16} \text{ cm}^2 \]

\[ \sigma_2 \approx 10^{-50} \text{ cm}^4 \text{ s} = 1 \text{ GM} \]

Goeppert-Mayer (GM)
Two-Photon Excited Fluorescence

- Short pulses at high peak power and strong focusing are required to achieve significant two-photon fluorescence (TPF)

\[
\frac{P_{tpa}(\nu)}{P_{abs}(2\nu)} = \phi \cdot \frac{\sigma_{2}(\nu)}{\sigma(2\nu)} \approx 10^{-34} \text{cm}^2 \text{s} \cdot \phi
\]

- Example: Short pulse laser @ 800nm: 100 mW, 100 MHz, 100 fs
  Focused with NA = 1.0

\[
\Phi_{pulse} \approx \frac{0.1 \text{ W}}{10^{-19} \text{ Ws}} \frac{1}{10^{-13} \text{s} \cdot 10^8 \text{s}^{-1}} = 10^{23} \text{s}^{-1} \quad A \approx \left(\frac{\lambda}{2NA}\right)^2 \approx 0.5 \cdot 10^{-7} \text{cm}^2
\]

\[
\phi = \frac{\Phi_{pulse}}{A} \approx 2 \cdot 10^{30} \text{cm}^{-2} \text{s}^{-1}
\]

Excitation limited to focal region \(\to\) reduces photobleaching
Two-Photon Fluorescence Microscopy

Single photon scanning confocal

Multi photon scanning

\[ I_i(x_i) = \int |h_{obs}(x_i, x_o; \omega)|^4 |O(x_o)|^2 dx_o \]

\[ I_i(x_i) = \int |h_{obs}(x_i, x_o; \omega/2)|^4 |O(x_o)|^2 dx_o \]
no molecular asymmetry required

Microscopy: imaging of interfaces -> cell surface, intercellular lipid droplets
Inherent sectioning and use of NIR excitation
-> deep imaging: 400μm penetration depth
observe development in real time
-> minute-temporal resolution
high resolution microscopy
-> 1μm spatial resolution

TPEF + SHG + THG

\[\text{N Olivier et al., Science 329, 967 (2010)}\]
Third-Order: Nonlinear Raman Scattering

non-parametric process

parametric process

\[ \omega_{SRS} = \omega_L - \omega_v \]

\[ \omega_{CARS} = \omega_{pump} + \omega_{probe} - \omega_{Stokes} \]

Stimulated Raman scattering (SRS)

Coherent Anti-Stokes Raman scattering (CARS)

Raman signals in cells (HeLa)

CARS scanning microscopy with detection in forward (F-CARS) and epi direction (E-CARS)

- In contrast to TPE, CARS is a coherent processes (phase matching in tight focus)
- Strong forward emission (but background by non-resonant water signal)
- backward emission can occur at interfaces

Imaging myelin sheath with CARS

- healthy spinal cords
- injured spinal cords
- treated spinal cords

**Intra-axonal free Ca^{2+}**

- myelin sheath: CARS (coherent anti-Stokes Raman scattering)
- calcium indicator Oregon Green 488: TPEF (two photon excited fluorescence)

*Y Shi et al., Nat. Nanotechnol. 5, 80 (2010)*
Same signal as linear Raman scattering but with enhanced sensitivity → suitable for scanning microscopy.
Stimulated Raman Scattering Microscopy

**Fig. 3.** SRL imaging of fresh mouse tissue. (A) Neuron bundles in corpus callosum of mouse brain imaged at 2845 cm$^{-1}$ highlighting myelin sheaths rich in CH$_2$. See movie in (18). (B) Epi-detected SRL CH$_2$ image acquired from thick brain tissue. (C) SRL CH$_2$ images of mouse ear skin in the same area at the indicated depths. From left to right: stratum corneum (4 μm), sebaceous gland (42 μm), and subcutaneous fat layer (105 μm). See movie in (18). (D) Comparison of SRL and CARS images of stratum corneum on (2845 cm$^{-1}$) and off (2780 cm$^{-1}$) the CH$_2$ resonance. Unlike CARS, SRL has no nonresonant background.