Nonlinear optics

- Nonlinear optical media
- Second-order nonlinear optics
- Three-wave mixing phenomena
- Phase matching
- Third-order nonlinear optics
What is nonlinear optics?

It is the branch of optics that studies the behavior of light in *nonlinear media* where \( P \) and \( E \) have a *nonlinear relation*.

It is responsible for a number of interesting and exotic phenomena such as:

- Creating new frequencies
- Change the frequency of a light beam
- Changing its shape in space and time
- Mixing different laser pulses
- Create ultrashort laser pulses

…

In nonlinear optics the *superposition principle* is not valid anymore.
Nonlinear optics is not easy to observe

Nonlinear optical phenomena require very high intensities and/or electric fields, and several other conditions.

1. The light intensities of “normal” optics are too weak
2. Normal light sources are incoherent
3. Nonlinear media must possess certain symmetry conditions
4. **Phase-matching** is required
## Linear vs. nonlinear optics

<table>
<thead>
<tr>
<th>Linear optics</th>
<th>Nonlinear optics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Optical properties of materials are <strong>independent of intensity</strong></td>
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</tr>
<tr>
<td>The <strong>principle of superposition</strong> applies</td>
<td>The <strong>principle of superposition</strong> does not apply</td>
</tr>
<tr>
<td>The frequency of light going through a medium <strong>does not change</strong></td>
<td>The frequency of light going through a medium <strong>may change</strong></td>
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<tr>
<td>Two beams of light in the same region of a medium have no effect on each other: <strong>light can not control light</strong></td>
<td>Two beams of light in the same region of a medium affect each other: <strong>light can control light</strong></td>
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Where does nonlinear optics come from?

We can understand the basics of nonlinear optics in terms of waves or in terms of photons. Using a wave description, nonlinear behavior is associated with **distortion**.
Where does nonlinear optics come from?

**Linear optics**
- Molecular energy levels
  - Input light photon energy
  - Emitted light photon energy
- Input wave
- Emitted wave
- Molecule

**Nonlinear optics**
- Molecular energy levels
  - Input light photon energy
  - Emitted light photon energy
- Input wave
- Emitted wave: *new!*
- Molecule
Describing nonlinear optics

We are going to use a description based on the nonlinear wave equation, that can be derived from Maxwell's equations:

\[ \nabla \times \mathcal{H} = \frac{\partial \mathcal{D}}{\partial t} \quad \nabla \cdot \mathcal{B} = 0 \]
\[ \nabla \times \mathcal{E} = -\frac{\partial \mathcal{B}}{\partial t} \quad \nabla \cdot \mathcal{D} = 0 \]

- Electric field \( \mathcal{E} \)
- Magnetic field \( \mathcal{H} \)
- Electric flux density \( \mathcal{D} \)
- Magnetic flux density \( \mathcal{B} \)
- Polarization density \( \mathcal{P} \)
- Magnetization density \( \mathcal{M} \)
- El. permittivity (free sp.) \( \varepsilon_0 \)
- El. susceptibility \( \chi \)

\[ \mathcal{D} = \varepsilon_0 \mathcal{E} + \mathcal{P} \]
\[ \mathcal{B} = \mu_0 \mathcal{H} + \mu_0 \mathcal{M} \]

The specific shape of the wave equation depends on the relation between \( \mathcal{P} \) and \( \mathcal{E} \) for each type of medium (constitutive relation).
A *linear* medium is characterized by a *linear* constitutive relation

The relation at every \((r,t)\) is simply \((\chi \text{ is the electric susceptibility})\)

\[ \mathcal{P} = \varepsilon_0 \chi \mathcal{E} \]

You can think of this as similar to the restoring force of a spring \(F = -kx\)

Maxwell’s equations and wave equation for a linear medium:

\[
\nabla \times \mathcal{H} = \varepsilon \frac{\partial \mathcal{E}}{\partial t} \quad \nabla \cdot \mathcal{H} = 0
\]

\[
\nabla \times \mathcal{E} = -\mu \frac{\partial \mathcal{H}}{\partial t} \quad \nabla \cdot \mathcal{E} = 0
\]

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

\[
\quad c = \frac{1}{\sqrt{\varepsilon \mu}}
\]

\[
\quad n = \frac{c_0}{c} = \sqrt{\frac{\varepsilon \mu}{\varepsilon_0 \mu_0}}
\]

\[
\quad = \sqrt{1+\chi}
\]
A *nonlinear* medium is characterized by a *nonlinear* constitutive relation

\[ \mathcal{P} \propto a_1 \mathcal{E} + a_2 \mathcal{E}^2 + a_3 \mathcal{E}^3 + \cdots \]

The previous Maxwell’s equation in a linear medium and the wave equation are *not* valid.

Starting from the general Maxwell’s eqns. in a medium, and considering a homogeneous and isotropic medium:

\begin{align*}
\nabla \times \mathcal{E} &= -\mu \frac{\partial \mathcal{H}}{\partial t} \\
\nabla \times (\nabla \times \mathcal{E}) &= -\nabla \times \left( \mu \frac{\partial \mathcal{H}}{\partial t} \right)
\end{align*}

**general wave equation for a homogeneous, isotropic medium**

\[ \nabla^2 \mathcal{E} - \frac{1}{c_0^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}}{\partial t^2} \]
Linear vs. nonlinear dielectric media

Most dielectric media are approximately linear, except when using very high electric fields – e.g. powerful, focused laser beams. In that case, a Taylor expansion should be used. The nonlinearity becomes relevant for $E \sim 10^5 – 10^8$ V/m.

\[ P = \varepsilon_0 \chi E \]

\[ P \propto a_1 E + a_2 E^2 + a_3 E^3 + \cdots \]
The nonlinear polarization

It is more practical to write the nonlinear relation in the form

\[ P \propto \varepsilon_0 \chi \mathcal{E} + 2d \mathcal{E}^2 + 4\chi^{(3)} \mathcal{E}^3 + \ldots \]

the usual **linear susceptibility**

**quadratic nonlinearity**
\[ d \approx 10^{-24} - 10^{-21} \text{ C/V}^2 \]

**cubic nonlinearity**
\[ \chi^{(3)} \approx 10^{-34} - 10^{-29} \text{ Cm/V}^3 \]

This classifies the corresponding related phenomena in **second-order** and **third-order nonlinear optics**.
The nonlinear wave equation

Let’s look again at the general wave equation in a medium:

Now we separate \( P \) into its linear and nonlinear components and replace:

By using the well-known relations between \( c, c_0, n, \varepsilon_0, \mu_0 \) and \( \chi \):

This equation represents a wave propagating in a medium with a “source” term linked to \( \mathcal{E}^2 \) – it is a nonlinear PDE in \( \mathcal{E} \).
How to solve the nonlinear wave equation

A first approach is called the *Born Approximation*:

1. **1st Born approximation**
   - Given $\mathcal{E}_0$, calculate $S(\mathcal{E}_0)$
   - Solve the equation to obtain $\mathcal{E}_1$

2. **2nd Born approximation**
   - Given $\mathcal{E}_1$, calculate $S(\mathcal{E}_1)$
   - Solve the equation to obtain $\mathcal{E}_2$

For a small nonlinearity, the first approximation is enough – this is what we will use in the next derivations.
Harmonic field in a 2\textsuperscript{nd} order nonlinear medium

This is an example of a second-order process:

\[ \mathcal{P}_{NL} = 2d\varepsilon^2 \]

Let’s calculate the response of this nonlinear medium to a harmonic field:

Following the 1\textsuperscript{st} Born Approximation, we now calculate \( S(\varepsilon_0) \):

Replacing the expression for \( \varepsilon_0 \), we can write the nonlinear polarization as:

The polarization has the form of a sum of
- a DC component \( P_{NL}(0) \)
- a component \( P_{NL}(2\omega) \) at twice the frequency

\[ \varepsilon_0(t) = \text{Re}\left\{ E(\omega)e^{i\omega t} \right\} \]
\[ \frac{1}{2} \left[ E(\omega)e^{i\omega t} + E^*(\omega)e^{-i\omega t} \right] \]

\[ \mathcal{P}_{NL}(t) = 2d\varepsilon_0^2 \]

\[ S[\varepsilon_0(t)] = -\mu_0\varepsilon_0^2 \frac{\partial^2 P_{NL}}{\partial t^2} \]

\[ \mathcal{P}_{NL}(t) = P_{NL}(0) + \text{Re}\left\{ P_{NL}(2\omega)e^{i2\omega t} \right\} \]

\( dE(\omega)E^*(\omega) \quad dE^2(\omega) \)
Harmonic field in a 2\textsuperscript{nd} order nonlinear medium

Medium response

\[ P_{NL} = 2d\mathcal{E}^2 \]

Harmonic input field

\[ \mathcal{E}_0(t) \]

\[ P_{NL}(t) = P_{NL}(0) + \text{Re}\left\{ P_{NL}(2\omega)e^{i2\omega t} \right\} \]

DC component  Second harmonic
Second harmonic generation (SHG)

As we have seen, the nonlinear polarization $P_{NL}(t)$ has a component at a frequency $2\omega$.

So the source term $S(t)$ will also “radiate” at $2\omega$, with a complex amplitude $S(2\omega)$:

$$P_{NL}(t) = P_{NL}(0) + \text{Re}\left\{dE^2(\omega)e^{i2\omega t}\right\}$$

$$S(t) = -\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$$

$$S(2\omega) = 4\mu_0 \omega^2 dE^2$$

The field $E_1$ that is a solution of the wave equation will also have components at $2\omega$. This is a mechanism for generating light at twice the frequency.
SHG explained by photons

If the intensity is high enough, two photons may be absorbed at once in a transition to a virtual level, resulting in the emission of a single photon at twice the frequency.

Pantazis et al, PNAS 2010
The first demonstration of SHG


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 Å. The arrow at 3472 Å indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 Å is very large due to halation.
Efficiency of SHG

The amplitude of the emitted $2\omega$ light is proportional to $S(2\omega)$, so for the intensity we have:

$$I(2\omega) \propto |S(2\omega)|^2 \propto d^2 |E(\omega)|^4 \propto d^2 I^2(\omega)$$

It is also prop. to the $L^2$ (length) of the medium:

$$I(2\omega) \propto L^2 d^2 I^2(\omega)$$

So we can write for the **efficiency of SHG:**

$$\frac{I(2\omega)}{I(\omega)} \propto d^2 \frac{L^2}{A} P$$

For a given crystal length $L$ the power $P$ should be concentrated in the smallest possible area $A$
The electro-optic effect

In this case we consider an electric field $E(t)$ consisting of a steady state component and a harmonic component:

$$\mathcal{E}_0(t) = E(0) + \text{Re}\left\{E(\omega)e^{i\omega t}\right\}$$

steady-state

$$(\omega=0)$$

harmonic

$$(\omega)$$

The polarization has the form of a sum of:

- a DC component $P_{NL}(0)$
- a comp. $P_{NL}(\omega)$ at the original frequency
- a comp. $P_{NL}(2\omega)$ at twice the frequency

$$P_{NL}(t) = P_{NL}(0) + \text{Re}\left\{P_{NL}(\omega)e^{i\omega t}\right\} + \text{Re}\left\{P_{NL}(2\omega)e^{i2\omega t}\right\}$$

But if $E(\omega) \ll E(0)$ then $P_{NL}(2\omega)$ is much smaller than the other two, so the total polarization is essentially linear with $E(\omega)$.
The electro-optic effect

In the presence of a strong steady field $E(0)$, the nonlinear polarization becomes linear with the optical field $E(\omega)$:

$$P_{NL}(\omega) = \varepsilon_0 \Delta \chi E(\omega)$$

$$\Delta \chi = \left( \frac{4d}{\varepsilon_0} \right) E(0)$$

The constant of proportionality is governed by $E(0)$.

The medium is linear with a refractive index that can be controlled by the applied electric field = Pockels effect.
Pockels cells

= electrooptic devices used to *rotate* the polarization of light

One of the main applications of Pockels cells is as an electrooptic switch (together with a polarizer) in q-switched lasers ($E \sim 3 \text{ kV}$).
Three wave mixing

Now let's consider two harmonic waves at different frequencies:

It can be demonstrated that the polarization will have components at:

\[ 0, \quad 2\omega_1, 2\omega_2, \quad \omega_1 + \omega_2 \text{ and } \omega_1 - \omega_2, \]

\[
E_0(t) = \text{Re}\{E(\omega_1)e^{i\omega_1 t} + E(\omega_2)e^{i\omega_2 t}\}
\]

\[
P_{NL}(0) = d \left( |E(\omega_1)| + |E(\omega_2)| \right)
\]

\[
P_{NL}(2\omega_1) = dE(\omega_1)E(\omega_1)
\]

\[
P_{NL}(2\omega_2) = dE(\omega_2)E(\omega_2)
\]

\[
P_{NL}(\omega_1 + \omega_2) = 2dE(\omega_1)E(\omega_2)
\]

\[
P_{NL}(\omega_1 - \omega_2) = 2dE(\omega_1)E^*(\omega_2)
\]

A 2\textsuperscript{nd} order nonlinear medium can be used to mix two different frequencies and generate a third wave:

- at the sum frequency (\textit{frequency up-conversion})
- at the difference frequency (\textit{frequency down-conversion})
Sum frequency generation

\[ \hbar \omega_3 = \hbar \omega_1 + \hbar \omega_2 \]

\[ \frac{1}{\lambda_{03}} = \frac{1}{\lambda_{01}} + \frac{1}{\lambda_{02}} \]
Sum frequency generation vibrational spectroscopy

This technique allows the study of surfaces and interfaces by mixing an IR and a visible laser beams.
Frequency and phase matching

In a three-wave mixing process not necessarily all frequencies are generated: certain additional conditions are required

\[ P_{NL}(\omega_1 + \omega_2) = 2dA_1A_2 \exp[-i(k_1 + k_2) \cdot r] \]

\[ E(\omega_1) = A_1 \exp(-i k_1 \cdot r) \]
\[ E(\omega_2) = A_2 \exp(-i k_2 \cdot r) \]
\[ E(\omega_3) = A_3 \exp(-i k_3 \cdot r) \]

\[ \omega_1 + \omega_2 = \omega_3 \]  
\[ k_1 + k_2 = k_3 \]

These two conditions ensure that the phases of the three waves are matched in space and time, which is necessary for their mutual interaction.
Modalities of three-wave mixing

Optical frequency conversion

Optical parametric amplifier

Optical parametric oscillator

Spontaneous parametric downconversion
Three wave mixing in terms of photons

- two photons of frequency $\omega_1$ and $\omega_2$ are annihilated and a new photon of higher frequency $\omega_3$ is created, or
- one photon of higher frequency $\omega_3$ is annihilated and two new photons of frequency $\omega_1$ and $\omega_2$ are created

\[
\hbar \omega_1 + \hbar \omega_2 = \hbar \omega_3 \\
\hbar k_1 + \hbar k_2 = \hbar k_3
\]

conservation of energy

conservation of momentum
How to achieve phase matching?

Let’s look at the equations again (assuming that all beams are collinear):

- the first one is automatically met
- the second one requires the following:

\[ \omega_1 + \omega_2 = \omega_3 \]
\[ k_1 + k_2 = k_3 \]

\[ \omega_1 n_1 + \omega_2 n_2 = \omega_3 n_3 \]

\( (k = \omega n / c_0) \)

It would be easy to match the 2nd one if \( n_1 = n_2 = n_3 \), but dispersion does not allow. The higher the frequency, the higher the value of \( n \).

What can we do?
Phase matching is made possible by birefringence

Anisotropic materials exhibit birefringence i.e. the refractive index “seen” by light depends on its polarization and the angle it makes with special axes of the medium.

We can use these degrees of freedom to control the refractive index at the desired frequencies.

Example: phase matching for SHG in a uniaxial crystal

\[
\frac{1}{n^2(\theta, \omega)} = \frac{\cos^2 \theta}{n_o^2(\omega)} + \frac{\sin^2 \theta}{n_e^2(\omega)}
\]
Phase matching in SHG

**weak phase matching:**
Intensity is low and “spatial pulsation” is short

**strong phase matching:**
Intensity is high and “spatial pulsation” is long
Third order nonlinear optics

In some types of optical media the second-order term vanishes ($d = 0$) and we have a dominant cubic nonlinearity:

$$P_{NL} = 4\chi^{(3)}E^3$$

This is called a **Kerr medium**.

The nonlinear polarization has terms of the type

$$P_{NL}(\omega) = 3\chi^{(3)}|E(\omega)|^2 E(\omega)$$

$$P_{NL}(3\omega) = \chi^{(3)}E^3(\omega)$$
Optical Kerr effect

$P_{NL}(\omega)$ corresponds to an increase in the refractive index proportional to the intensity, where $n_2$ = optical Kerr coefficient.

This corresponds to a modulation in the phase of a laser pulse:

$$n(I) = n + n_2 I$$

$$E(z,t) = E_0 \exp\left[ ik_0 n(I) z \right]$$

$$= E_0 e^{iknz} \exp\left[ ik_0 n_2 \left( P / A \right) z \right]$$

Applications:

• generation of new frequencies in Kerr media (e.g. optical fibers)

• self-focusing (e.g. Kerr-lens mode locking)
Gaussian pulse in a dispersive and nonlinear medium

https://youtu.be/c5oud_h8R6I
Soliton in a positive dispersion and nonlinear medium

https://youtu.be/ua0GQHmlZaQ
Kerr lens mode locking

Remember that mode locking needs a mechanism to discriminate between low intensity and high intensity pulses. In this case, **self focusing** is that mechanism.

- **no pulses:** low intensity, no self focusing
- **short pulses:** high intensity, self focusing
Extreme nonlinear optics

High harmonic generation allows the creation of laser pulses down to only a few nm from a source at e.g. 800 nm
High harmonic generation

Three step model for HHG
Extreme nonlinear optics

Photonic crystal fibers can exhibit strong nonlinearity leading to intense self-phase modulation and generation of a very broad frequency range.
Extreme nonlinear optics

Supercontinuum generation in solid state media can also lead to a very broadband spectrum in only a few mm.
This is an alternative laser amplification technique, with no population inversion – energy is transferred from the pump to the signal pulse through three wave mixing.
OPCPA = Optical Parametric Chirped Pulse Amplification

(Optical Parametric Amplification + Chirped Pulse Amplification)
Research topics at the Laboratory for Intense Lasers

Laser development and laser applications
High intensity lasers
CPA and OPCPA amplifiers
Supercontinuum generation
Ultrashort pulse diagnostics
High harmonic generation
Coherent x-ray sources
X-ray imaging
Laser particle acceleration
Optical physics

http://xgolp.ist.utl.pt/