

# Non-Linear effects

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## Lecture 3



# Outline

## 1 Introduction

## 2 The Nonlinear Optical Susceptibility

- Introduction
- Descriptions of Nonlinear Optical Processes
- Nonlinear Susceptibility of a Classical Anharmonic Oscillator

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# Nonlinear Optics

- **Linear optics** - 'Optics of weak light':  
Light is deflected or delayed but its frequency is unchanged.  
**The principle of superposition holds**
- **Nonlinear optics** - 'Optics of intense light':  
We are concerned with the effects that light itself induces as it propagates through the medium.  
**Superposition principle is not valid**
  - Fundamental physics.
  - It leads to important applications.

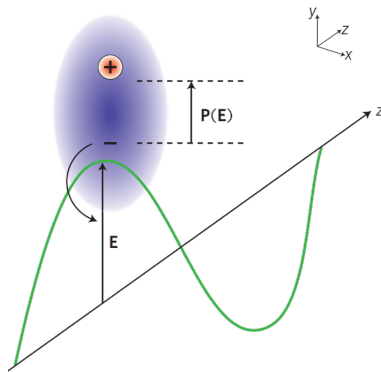
# Why Silicon?



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## Linear and Nonlinear Polarization



**Figure 1:** An electromagnetic wave with electric field  $\mathbf{E}$  passing through an atom and thereby inducing a dipole oscillation  $\mathbf{P}(\mathbf{E})$ .

# Simple Formulation of the Theory of Nonlinear Optics

- Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light.
- Typically, only laser light is sufficiently intense to modify the optical properties of a material system.
- Nonlinear optical phenomena are "nonlinear" in the sense that they occur when the response of a material system to an applied optical field depends in a nonlinear manner on the strength of the optical field.

In order to describe more precisely what we mean by an optical nonlinearity, let us consider how the dipole moment per unit volume, or polarization  $\tilde{P}(t)$ , of a material system depends on the strength  $\tilde{E}(t)$  of an applied optical field.

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

where the constant of proportionality  $\chi^{(1)}$  is known as the linear susceptibility and  $\varepsilon_0$  is the permittivity of free space.

# Simple Formulation of the Theory of Nonlinear Optics

Assume that instantaneous dielectric response in an isotropic material, the relation between an induced polarization and an electric field is expressed by a power series in the electric field:

$$\begin{aligned}\tilde{P}(t) &= \varepsilon_0[\chi^{(1)}\tilde{E}(t) + \chi^{(2)}\tilde{E}^2(t) + \chi^{(3)}\tilde{E}^3(t) + \dots] \\ &\equiv \tilde{P}^{(1)}(t) + \tilde{P}^{(2)}(t) + \tilde{P}^{(3)}(t) + \dots\end{aligned}\tag{2}$$

The quantities  $\chi^{(2)}$  and  $\chi^{(3)}$  are known as the second- and third-order nonlinear optical susceptibilities, respectively.

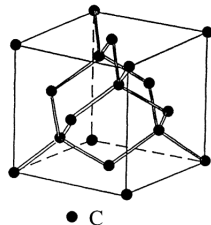
Physical processes that occur as a result of the second-order polarization  $\tilde{P}^{(2)}$  tend to be distinct from those that occur as a result of the third-order polarization  $\tilde{P}^{(3)}$ .

**Note:** Throughout the lecture, we use the tilde to denote a quantity that varies rapidly in time.

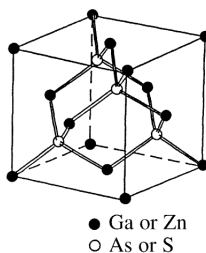
# Simple Formulation of the Theory of Nonlinear Optics

- Second-order nonlinear optical interactions can occur only in noncentrosymmetric crystals, in crystals that do not display inversion symmetry.
- Third-order nonlinear optical interactions (i.e., those described by a  $\chi^{(3)}$  susceptibility) can occur for both centrosymmetric and noncentrosymmetric media.

(a) diamond structure



(b) zincblende structure



**Figure 2:** Illustration of (a) the diamond structure and (b) the zincblende structure. Both possess a cubic lattice and thus cannot display birefringence, but the carbon structure is centrosymmetric, whereas the zincblende structure is noncentrosymmetric.



# Nonlinear Interactions Summary

We shall make a simple order-of-magnitude estimate of the size of these quantities for the common case in which the nonlinearity is electronic in origin.

One might expect that the lowest-order correction term  $\tilde{P}^{(2)}$  would be comparable to the linear response  $\tilde{P}^{(1)}$  when the amplitude of the applied field  $\tilde{E}$  is of the order of the characteristic atomic electric field strength

$$E_{at} = \frac{e}{4\pi\epsilon_0 a_0^2}$$

where  $a_0 = \frac{4\pi\epsilon_0\hbar}{me^2}$  is the Bohr radius of the hydrogen atom and  $\hbar$  is Planck's constant divided by  $2\pi$ .

Numerically,  $E_{at} = 5.14 \times 10^{11} \left[ \frac{\text{V}}{\text{m}} \right]$ .

# Nonlinear Interactions Summary

We thus expect that under conditions of nonresonant excitation the second order susceptibility  $\chi^{(2)}$  will be of the order of  $\chi^{(1)}/E_{at}$ . For condensed matter  $\chi^{(1)}$  is of the order of unity, and we hence expect that  $\chi^{(2)}$  will be of the order of  $1/E_{at}$ , or that

$$\chi^{(2)} \simeq \frac{\chi^{(1)}}{E_{at}} \Rightarrow \chi^{(2)} \simeq 1.94 \times 10^{-12} \left[ \frac{m}{v} \right]$$

Similarly, we expect  $\chi^{(3)}$  to be of the order of  $\chi^{(1)}/E_{at}^2$  at, which for condensed matter is of the order of

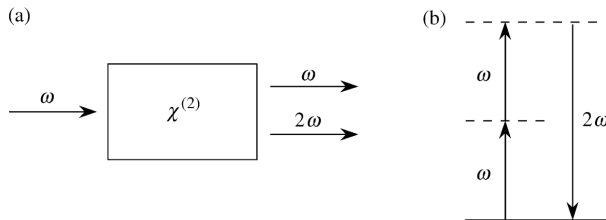
$$\chi^{(3)} \simeq \frac{\chi^{(1)}}{E_{at}^2} \Rightarrow \chi^{(3)} \simeq 3.78 \times 10^{-24} \left[ \frac{m^2}{v^2} \right]$$

## Second Harmonic Generation (SHG)

As an example of a nonlinear optical interaction, let us consider the process of second-harmonic generation. Here a laser beam whose electric field strength is represented as

$$\tilde{E}(t) = Ee^{-i\omega t} + c.c. \quad (3)$$

is incident upon a crystal for which the second-order susceptibility  $\chi^{(2)}$  is nonzero.



**Figure 3:** (a) Geometry of second-harmonic generation. (b) Energy-level diagram describing second-harmonic generation.

## Second-Harmonic Generation (SHG)

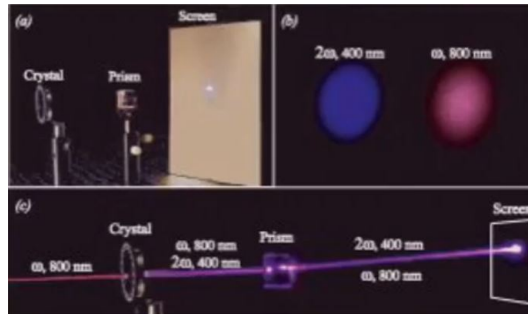
The nonlinear polarization that is created in such a crystal is:

$$\tilde{P}^{(2)}(t) = 2\varepsilon_0\chi^{(2)}EE^* + (\varepsilon_0\chi^{(2)}E^2e^{-i2\omega t} + c.c.) \quad (4)$$

- The second-order polarization consists of a contribution at zero frequency (the first term) and a contribution at frequency  $2\omega$  (the second term).
- This latter contribution can lead to the generation of radiation at the second-harmonic frequency.
- Under proper experimental conditions, nearly all of the power in the incident beam at frequency  $\omega$  is converted to radiation at the second-harmonic frequency  $2\omega$ .

## Second-Harmonic Generation (SHG)

- One common use of second-harmonic generation is to convert the output of a fixed-frequency laser to a different spectral region.
- The Nd:YAG laser operates in the near infrared at a wavelength of  $1.06\text{ }\mu\text{m}$ . Second-harmonic generation is routinely used to convert the wavelength of the radiation to  $0.53\text{ }\mu\text{m}$ , in the middle of the visible spectrum.



## Sum and Difference Frequency Generation

Lets assume that the optical field incident upon a second-order nonlinear optical medium consists of two distinct frequency components

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c. \quad (5)$$

Then, assuming that the second-order contribution to the nonlinear polarization is of the form:

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

the nonlinear polarization is given by

$$\begin{aligned} \tilde{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} [ & E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} \\ & + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + c.c.] + 2\varepsilon_0 \chi^{(2)} [E_1 E_1^* + E_2 E_2^*] \end{aligned} \quad (6)$$

It is convenient to express this result using the notation

$$\tilde{P}^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}$$

where the summation extends over positive and negative frequencies  $\omega_n$ .

## Sum and Difference Frequency Generation

$$P(2\omega_1) = \varepsilon_0 \chi^{(2)} E_1^2 \quad (\text{SHG})$$

$$P(2\omega_2) = \varepsilon_0 \chi^{(2)} E_2^2 \quad (\text{SHG})$$

$$P(\omega_1 + \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1 E_2 \quad (\text{SFG})$$

$$P(\omega_1 - \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1 E_2^* \quad (\text{DFG})$$

$$P(\omega = 0) = 2\varepsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*) \quad (\text{OR})$$

where (SHG) - second-harmonic generation, (SFG) - sum-frequency generation, (DFG) - difference-frequency generation and (OR) - optical rectification.

There is also a response at the negative of each of the nonzero frequencies. However, each of these quantities is simply the complex conjugate of one of the quantities, it is not necessary to take explicit account of both the positive and negative frequency components.

## Sum and Difference Frequency Generation

- Typically no more than one of these frequency components will be present with any appreciable intensity in the radiation generated by the nonlinear optical interaction.
- The reason for this behavior is that the nonlinear polarization can efficiently produce an output signal only if a certain phase-matching condition is satisfied, and usually this condition cannot be satisfied for more than one frequency component of the nonlinear polarization.
- Operationally, one often chooses which frequency component will be radiated by properly selecting the polarization of the input radiation and the orientation of the nonlinear crystal.

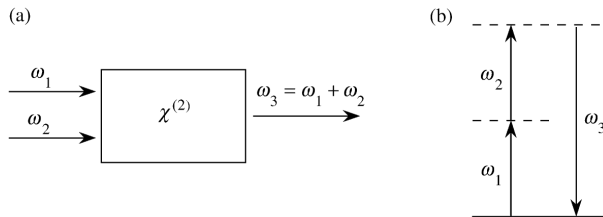


## Sum Frequency Generation (SFG)

consider the process of sum-frequency generation

$$P(\omega_1 + \omega_2) = 2\varepsilon_0\chi^{(2)}E_1E_2 \quad (7)$$

In many ways the process of sum-frequency generation is analogous to that of second-harmonic generation, except that in sum-frequency generation the two input waves are at different frequencies.



**Figure 4:** Sum-frequency generation. (a) Geometry of the interaction. (b) Energy-level description.

## Sum Frequency Generation (SFG)

One application of sum-frequency generation is to produce tunable radiation in the ultraviolet spectral region by choosing one of the input waves to be the output of a fixed-frequency visible laser and the other to be the output of a frequency-tunable visible laser.

Examples for Sum frequency generation are:

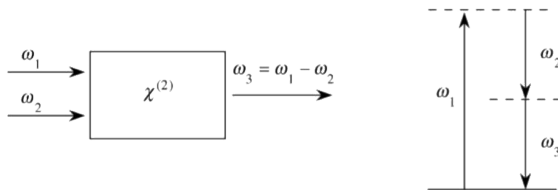
- Generation of ultraviolet light, e.g. by mixing the output of a 1064 nm Nd:YAG laser with frequency-doubled light at 532 nm, resulting in 355 nm UV light.
- Generation of red light, e.g. by mixing the outputs of a 1064 nm Nd:YAG laser and a 1535 nm fiber laser, resulting in an output at 628 nm.

## Difference Frequency Generation (DFG)

The process of difference-frequency generation is described by a nonlinear polarization of the form

$$P(\omega_1 - \omega_2) = 2\varepsilon_0\chi^{(2)}E_1E_2^* \quad (8)$$

Here the frequency of the generated wave is the difference of those of the applied fields. Difference-frequency generation can be used to produce tunable infrared radiation by mixing the output of a frequency-tunable visible laser with that of a fixed-frequency visible laser.



**Figure 5:** Difference-frequency generation. (a) Geometry of the interaction. (b) Energy-level description.

## Difference Frequency Generation (DFG)

Difference-frequency generation can be used to produce tunable infrared radiation by mixing the output of a frequency-tunable visible laser with that of a fixed-frequency visible laser.

Examples for Sum frequency generation are:

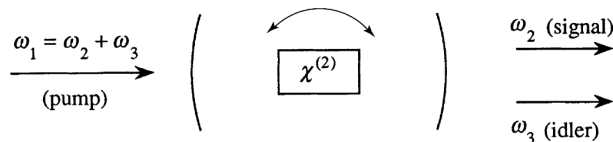
- Generation of light around  $3.3 \mu\text{m}$  by mixing  $1570 \text{ nm}$  from a fiber laser and  $1064 \text{ nm}$ .
- Generation of light around  $4.5 \mu\text{m}$  by mixing  $860 \text{ nm}$  from a laser diode and  $1064 \text{ nm}$ .

## Difference Frequency Generation (DFG)

- An important difference between the two processes can be deduced from the description of difference-frequency generation in terms of a photon energy-level diagram.
- We see that conservation of energy requires that for every photon that is created at the difference frequency  $\omega_3 = \omega_1 - \omega_2$ , a photon at the higher input frequency ( $\omega_1$ ) must be destroyed and a photon at the lower input frequency ( $\omega_2$ ) must be created.
- The lower frequency input field is amplified by the process of difference-frequency generation. The process of difference-frequency generation is also known as optical parametric amplification.
- According to the photon energy-level description of difference-frequency generation, the atom first absorbs a photon of frequency  $\omega_1$  and jumps to the highest virtual level.
- This level decays by a two-photon emission process that is stimulated by the presence of the  $\omega_2$  field, which is already present.
- Two-photon emission can occur even if the  $\omega_2$  field is not applied. The generated fields in such a case are very much weaker, since they are created by spontaneous two photon emission from a virtual level. This process is known as parametric fluorescence.

# Optical Parametric Oscillation

- The process of difference-frequency generation the presence of radiation at frequency  $\omega_2$  or  $\omega_3$  can stimulate the emission of additional photons at these frequencies.
- If the nonlinear crystal used in this process is placed inside an optical resonator, the  $\omega_2$  and/or  $\omega_3$  fields can build up to large values.
- One controls the output frequency of an optical parametric oscillator by adjusting the phase-matching condition.
- Optical parametric oscillators are frequently used at infrared wavelengths



**Figure 6:** The optical parametric oscillator. The cavity end mirrors have high reflectivities at frequencies  $\omega_2$  and/or  $\omega_3$ . The output frequencies can be tuned by means of the orientation of the crystal.

## Third-Order Nonlinear Optical Processes

Consider the third-order contribution to the nonlinear polarization

$$\tilde{P}^{(3)}(t) = \varepsilon_0 \chi^{(3)} \tilde{E}^3(t) \quad (9)$$

For simplicity, we first consider the simple case in which the applied field is monochromatic and is given by

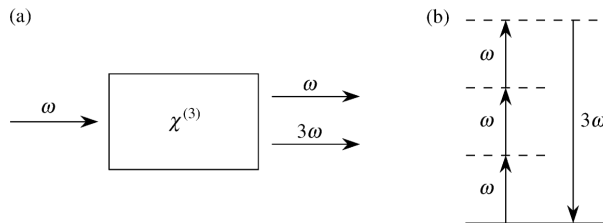
$$\tilde{E}(t) = \mathcal{E} \cos(\omega t) \quad (10)$$

Using the identity  $\cos^3(\omega t) = \frac{1}{4} \cos(3\omega t) + \frac{3}{4} \cos(\omega t)$ , we get

$$\tilde{P}^{(3)}(t) = \frac{1}{4} \varepsilon_0 \chi^{(3)} \mathcal{E}^3 \cos(3\omega t) + \frac{3}{4} \varepsilon_0 \chi^{(3)} \mathcal{E}^3 \cos(\omega t) \quad (11)$$

## Third-Harmonic Generation (THG)

- The first term in Eq. (11) describes a response at frequency  $3\omega$  that is created by an applied field at frequency  $\omega$ .
- This term leads to the process of third-harmonic generation.



**Figure 7:** Third-harmonic generation. (a) Geometry of the interaction. (b) Energy-level description.



## Intensity-Dependent Refractive Index

The second term in Eq. (11) describes a nonlinear contribution to the polarization at the frequency of the incident field. This term hence leads to a nonlinear contribution to the refractive index experienced by a wave at frequency  $\omega$ .

$$n = n_0 + n_2 I$$

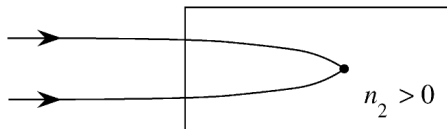
where  $n_0$  is the usual refractive index,  $I = \frac{1}{2} n_0 \epsilon_0 c \mathcal{E}^2$  is the intensity of the incident wave and  $n_2$  is defined as

$$n_2 = \frac{3}{2 n_0^2 \epsilon_0 c} \chi^{(3)}$$

which is an optical constant that characterizes the strength of the optical nonlinearity.

## Self-Focusing

- One of the processes that can occur as a result of the intensity dependent refractive index is self-focusing.
- This process can occur when a beam of light having a nonuniform transverse intensity distribution propagates through a material for which  $n_2$  is positive.
- The material effectively acts as a positive lens, which causes the rays to curve toward each other.
- This process is of great practical importance because the intensity at the focal spot of the self-focused beam is usually sufficiently large to lead to optical damage of the material.



**Figure 8:** Self-focusing of light.

## Third-Order Interactions (General Case)

Let us next examine the form of the nonlinear polarization

$$\tilde{P}^{(3)}(t) = \varepsilon_0 \chi^{(3)} \tilde{E}^3(t) \quad (12)$$

when the applied field consists of three frequency components:

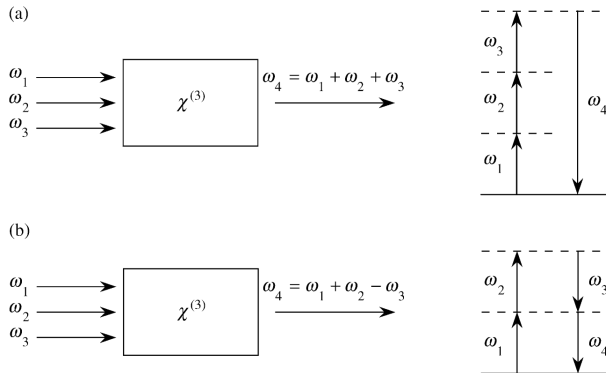
$$\tilde{E}(t) = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + E_3 e^{i\omega_3 t} + \text{c.c.} \quad (13)$$

When we calculate  $\tilde{E}^3(t)$ , we find that the resulting expression contains 44 different frequency components, if we consider positive and negative frequencies to be distinct.

In each case the frequency argument of  $P$  is equal to the sum of the frequencies associated with the field amplitudes appearing on the right-hand side of the equation. For example:

$$P(\omega_1 + \omega_2 + \omega_3) = 6\varepsilon_0 \chi^{(3)} E_1 E_2 E_3 \quad P(\omega_1 + \omega_2 - \omega_3) = 6\varepsilon_0 \chi^{(3)} E_1 E_2 E_3^*$$

## Third-Order Interactions



**Figure 9:** Two of the possible mixing processes, that can occur when three input waves interact in a medium characterized by a  $\chi^{(3)}$  susceptibility.

## Third-Order Interactions

Third-order nonlinearities are especially important in silicon as they exhibit a wide variety of phenomena.

$$\tilde{P}^{(3)}(t) = \frac{3}{4}\varepsilon_0\chi^3[|\mathbf{E}_{\omega_1}|^2\mathbf{E}_1 + \text{.}.] \quad (\text{SPM})$$

$$+ \frac{6}{4}\varepsilon_0\chi^3[(|\mathbf{E}_{\omega_2}|^2 + |\mathbf{E}_{\omega_3}|^2)\mathbf{E}_1 + \text{.}.] \quad (\text{XPM})$$

$$+ \frac{1}{4}\varepsilon_0\chi^3[(\mathbf{E}_{\omega_1}^3)e^{i3\omega_1 t} + c.c.] + \text{.}.] \quad (\text{THG})$$

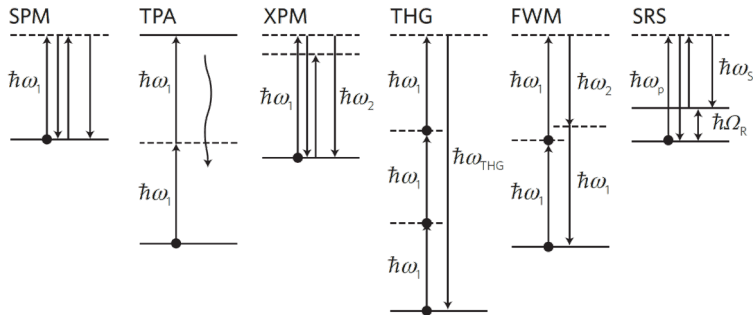
$$+ \frac{3}{4}\varepsilon_0\chi^3\left[\frac{1}{2}(\mathbf{E}_{\omega_1}^2\mathbf{E}_{\omega_2}e^{i(2\omega_1+\omega_2)t} + c.c.) + \text{.}.\right] \quad (\text{FWM})$$

$$+ \frac{3}{4}\varepsilon_0\chi^3\left[\frac{1}{2}(\mathbf{E}_{\omega_1}^2\mathbf{E}_{\omega_2}^*e^{i(2\omega_1-\omega_2)t} + c.c.) + \text{.}.\right] \quad (\text{FWM})$$

$$+ \frac{6}{4}\varepsilon_0\chi^3\left[\frac{1}{2}(\mathbf{E}_{\omega_1}\mathbf{E}_{\omega_2}\mathbf{E}_{\omega_3}^*e^{i(\omega_1+\omega_2-\omega_3)t} + c.c.) + \text{.}.\right] \quad (\text{FWM})$$

$$+ \frac{6}{4}\varepsilon_0\chi^3\left[\frac{1}{2}(\mathbf{E}_{\omega_1}\mathbf{E}_{\omega_2}\mathbf{E}_{\omega_3}e^{i(\omega_1+\omega_2+\omega_3)t} + c.c.) + \text{.}.\right] \quad (\text{FWM})$$

## Third-Order Interactions



**Figure 10:** Third-order nonlinear dipole transitions, showing self-phase modulation (SPM), two-photon absorption (TPA), cross-phase modulation (XPM), third-harmonic generation (THG), partially degenerate and non-degenerate four-wave mixing (FWM) and stimulated Raman scattering (SRS).

## Influence of Inversion Symmetry on the Second-Order Nonlinear Response

One of the symmetry properties that some but not all crystals possess is centrosymmetry, also known as inversion symmetry. For a material system that is centrosymmetric (i.e., possesses a center of inversion) the  $\chi^{(2)}$  nonlinear susceptibility must vanish identically.

This rule is very powerful since 11 of the 32 crystal classes possess inversion symmetry. We shall demonstrate this fact only for the special case of second-harmonic generation in a medium that responds instantaneously to the applied optical field.

$$\tilde{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} \tilde{E}^2(t) \quad (14)$$

where

$$\tilde{E}(t) = \mathcal{E} \cos(\omega t)$$

## Influence of Inversion Symmetry on the Second-Order Nonlinear Response

If we now change the sign of the applied electric field  $\tilde{E}(t)$ , the sign of the induced polarization  $\tilde{P}(t)$  must also change, because we have assumed that the medium possesses inversion symmetry.

$$-\tilde{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} [-\tilde{E}(t)]^2 \quad (15)$$

and

$$-\tilde{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} \tilde{E}^2(t) \quad (16)$$

It shows that

$$\chi^{(2)} = 0$$

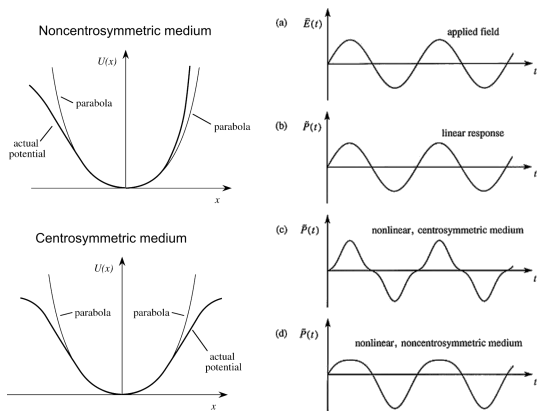
This result can be understood intuitively by considering the motion of an electron in a nonparabolic potential well. The nonlinearity of the associated restoring force, the atomic response will show significant harmonic distortion.



# Influence of Inversion Symmetry on the Second-Order Nonlinear Response

Part (c) shows the induced polarization for the case of a non-linear medium that possesses a center of symmetry. Although significant waveform distortion is evident, only odd harmonics of the fundamental frequency are present.

For the case (d) of a nonlinear, noncentrosymmetric medium, both even and odd harmonics are present in the waveform associated with the atomic response.



**Figure 11:** Potential energy function for a noncentrosymmetric and centrosymmetric medium and waveforms associated with the atomic response.

## Maxwell's equations

In non-conducting and non-magnetic media:  $\rho = 0$  and  $\mathbf{J} = 0$

$$\nabla \times \tilde{\mathbf{E}} = -\frac{\partial \tilde{\mathbf{B}}}{\partial t} \quad (17)$$

$$\nabla \times \tilde{\mathbf{H}} = \frac{\partial \tilde{\mathbf{D}}}{\partial t} \quad (18)$$

$$\nabla \cdot \tilde{\mathbf{D}} = 0 \quad (19)$$

$$\nabla \cdot \tilde{\mathbf{B}} = 0 \quad (20)$$

where

$$\tilde{\mathbf{B}} = \mu_0 \tilde{\mathbf{H}}$$

$$\tilde{\mathbf{D}} = \varepsilon_0 \tilde{\mathbf{E}} + \tilde{\mathbf{P}}$$

# The Wave Equation for Nonlinear Optical Media

$$\begin{aligned}\nabla \times \nabla \times \tilde{\mathbf{E}} + \mu_0 \frac{\partial^2 \tilde{\mathbf{D}}}{\partial t^2} &= 0 \\ \nabla \times \nabla \times \tilde{\mathbf{E}} + \frac{1}{c^2} \frac{\partial^2 \tilde{\mathbf{E}}}{\partial t^2} &= -\frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \tilde{\mathbf{P}}}{\partial t^2}\end{aligned}$$

with

$$\begin{aligned}\nabla \times \nabla \times \tilde{\mathbf{E}} &= \nabla(\nabla \cdot \tilde{\mathbf{E}}) - \nabla^2 \tilde{\mathbf{E}} \\ \nabla^2 \tilde{\mathbf{E}} - \frac{1}{c^2} \frac{\partial^2 \tilde{\mathbf{E}}}{\partial t^2} &= \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \tilde{\mathbf{P}}}{\partial t^2}\end{aligned}$$

Alternatively, the wave equation can be expressed as

$$\nabla^2 \tilde{\mathbf{E}} - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \tilde{\mathbf{D}}}{\partial t^2} = 0$$

where  $\tilde{\mathbf{D}} = \varepsilon_0 \tilde{\mathbf{E}} + \tilde{\mathbf{P}}$ .

# The Wave Equation for Nonlinear Optical Media

It is often convenient to split  $\tilde{\mathbf{P}}$  into its linear and nonlinear parts as

$$\tilde{\mathbf{P}} = \tilde{\mathbf{P}}^{(1)} + \tilde{\mathbf{P}}^{\text{NL}}$$

the displacement field  $\tilde{\mathbf{D}}$  into its linear and nonlinear parts as

$$\tilde{\mathbf{D}} = \tilde{\mathbf{D}}^{(1)} + \tilde{\mathbf{P}}^{\text{NL}} \qquad \tilde{\mathbf{D}}^{(1)} = \varepsilon_0 \tilde{\mathbf{E}} + \tilde{\mathbf{P}}^{(1)}$$

the wave equation can be written as

$$\nabla^2 \tilde{\mathbf{E}} - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \tilde{\mathbf{D}}^{(1)}}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \tilde{\mathbf{P}}^{\text{NL}}}{\partial t^2}$$

For the case of dispersive medium, we must consider each frequency component of the field separately.  $\tilde{\mathbf{E}}$ ,  $\tilde{\mathbf{D}}$ ,  $\tilde{\mathbf{P}}$  fields represented as the sums of their various frequency components:

$$\begin{aligned}\tilde{\mathbf{E}}(\mathbf{r}, t) &= \sum_n^I \tilde{\mathbf{E}}_n(\mathbf{r}, t) & \tilde{\mathbf{E}}_n(\mathbf{r}, t) &= \mathbf{E}_n(\mathbf{r})e^{-i\omega_n t} + c.c. \\ \tilde{\mathbf{D}}^{(1)}(\mathbf{r}, t) &= \sum_n^I \tilde{\mathbf{D}}_n^{(1)}(\mathbf{r}, t) & \tilde{\mathbf{D}}_n^{(1)}(\mathbf{r}, t) &= \mathbf{D}_n^{(1)}(\mathbf{r})e^{-i\omega_n t} + c.c. \\ \tilde{\mathbf{P}}^{\text{NL}}(\mathbf{r}, t) &= \sum_n^I \tilde{\mathbf{P}}_n^{\text{NL}}(\mathbf{r}, t) & \tilde{\mathbf{P}}_n^{\text{NL}}(\mathbf{r}, t) &= \mathbf{P}_n^{\text{NL}}(\mathbf{r})e^{-i\omega_n t} + c.c.\end{aligned}$$

where where the summation is of the various frequency components. The general case of a dissipative medium is treated by allowing the dielectric tensor to be a complex quantity that relates the complex field amplitudes according to

$$\begin{aligned}\mathbf{D}_n^{(1)}(\mathbf{r}) &= \varepsilon_0 \varepsilon^{(1)}(\omega_n) \mathbf{E}_n(\mathbf{r}) \\ \nabla^2 \mathbf{E}_n(\mathbf{r}) + \frac{\omega_n^2}{c^2} \varepsilon^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) &= -\frac{\omega_n^2}{\varepsilon_0 c^2} \mathbf{P}_n^{\text{NL}}(\mathbf{r})\end{aligned}$$

The nonlinear response of the medium acts as a source term which appears on the right-hand side of this equation.

## $\chi^{(1)}$ Processes - Linear Optics

The complex term  $\chi^{(1)}$  concerns dipole excitations with bound and free electrons induced by a single photon. The real part of  $\chi^{(1)}$  is associated with the real part of the refractive index, whereas the imaginary part describes loss or gain.

The contribution of bound electrons to the susceptibility:

$$\chi_{\text{Lorentz}}^{(1)} = \frac{\omega_p^2}{\omega_0^2 - \omega^2 + i\gamma_L\omega} \quad (21)$$

where  $\omega_0$  is the resonance frequency,  $\gamma_L$  is the associated damping constant and  $\omega_p$  is the plasma frequency defined as

$$\omega_p^2 = \frac{Nq^2}{\epsilon_0 m_e}$$

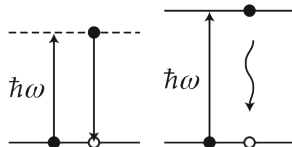
## $\chi^{(1)}$ Processes - Linear Optics

The contribution of free-carrier absorption to the susceptibility:

$$\chi_{\text{Drude}}^{(1)} = \frac{\omega_p^2}{-\omega^2 + i\gamma_D\omega}$$

For non-magnetic materials:

$$n^2 = 1 + \chi_{\text{Lorentz}}^{(1)} + \chi_{\text{drude}}^{(1)}$$



**Figure 12:** Energy level diagrams showing possible single-photon dipole transitions with contributions to refractive index changes (left) or to free-carrier absorption (right).

## $\chi^{(1)}$ Processes - Linear Optics

The refractive index changes with both wavelength and carrier concentration  $N$ .

$$n^2 = 1 + \chi_{\text{Lorentz}}^{(1)} + \chi_{\text{drude}}^{(1)}$$

When performing simulations, in order to fit the models with directly measured wavelength dependences and free-carrier contributions from electrons (concentration  $N_e$ ) and holes (concentration  $N_h$ ) to the refractive index, a useful empirical function is given by:

$$n(\lambda, N_e, N_h) = n_0(\lambda) + \Delta n_f(N_e, N_h) - i \frac{\lambda}{4\pi} \Delta \alpha_f(N_e, N_h)$$

Sellmeier relation:

$$n_0(\lambda)^2 = \varepsilon + \frac{A}{\lambda^2} + \frac{B\lambda^2}{\lambda^2 - \lambda_g^2}$$

In case of silicon:

$$\varepsilon = 11.686, A = 0.9398 \mu\text{m}^2, B = 8/1046 \times 10^{-3}, \text{ and } \lambda_g = 1.1071 \mu\text{m}$$



# Constitutive Relation Relative to the Electric Field

When light intensity  $\geq 1\text{MW}/\text{cm}^2$  (laser)

$$\tilde{P}(t) = \tilde{P}_0 + \tilde{P}^{(1)}(t) + \tilde{P}^{(2)}(t) + \tilde{P}^{(3)}(t) + \dots$$

with

$$\tilde{P}^{(1)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} d\tau R^{(1)}(t, \tau) \cdot \tilde{E}(\tau)$$

$$\tilde{P}^{(2)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2 R^{(2)}(t, \tau_1, \tau_2) : \tilde{E}(\tau_1) \otimes \tilde{E}(\tau_2)$$

$$\tilde{P}^{(3)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2 \int_{-\infty}^{\infty} d\tau_3 R^{(3)}(t, \tau_1, \tau_2, \tau_3) \therefore \tilde{E}(\tau_1) \otimes \tilde{E}(\tau_2) \otimes \tilde{E}(\tau_3)$$

$\otimes$  Tensorial product

$\therefore$  Contracted products

# Parametric Optics

**Parametric Optics** - interaction between the Fourier components of the electro-magnetic field.

$$\vec{F}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \tilde{F}(\omega) \exp(i\omega t)$$

$$\vec{F} \equiv \vec{E}, \vec{H}$$

$$\|\vec{F}(t)\| \in R \rightarrow \tilde{F}(\omega)^* = \tilde{F}(-\omega)$$

$$\tilde{\chi}^{(1)}(\omega) = \int_{-\infty}^{\infty} d\tau R^{(1)}(\tau) \exp(i\omega\tau)$$

$$\tilde{\chi}^{(2)}(\omega = \omega_1 \pm \omega_2) = \int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2 R^{(2)}(\tau_1, \tau_2) \exp(i\omega_1\tau_1 \pm i\omega_2\tau_2)$$

## The orders of the Fourier components of the polarization

$$\tilde{P}(\omega) = \tilde{P}^{(1)}(\omega) + \tilde{P}^{(2)}(\omega) + \tilde{P}^{(3)}(\omega) + \dots$$

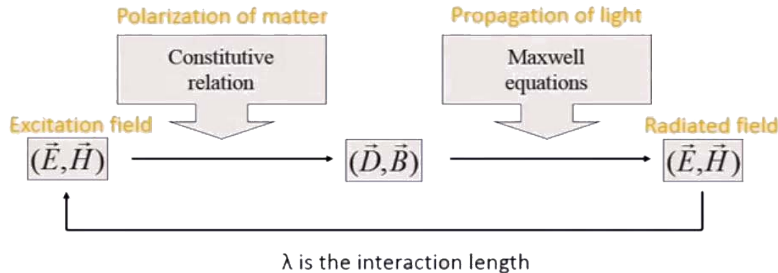
with

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

$$\tilde{P}^{(2)}(\omega) = \varepsilon_0 \chi^{(2)}(\omega = \omega_1 \pm \omega_2) : \tilde{E}(\omega_1) \otimes \tilde{E}(\pm\omega_2)$$

$$\tilde{P}^{(3)}(\omega) = \varepsilon_0 \chi^{(3)}(\omega = \omega_3 \pm \omega_4 \pm \omega_5) \therefore \tilde{E}(\omega_1) \otimes \tilde{E}(\omega_2) \otimes \tilde{E}(\omega_3)$$

# The Algorithm of Light/Matter Interaction



$$\vec{D}(\omega) = \epsilon_0 \vec{E}(\omega) + \vec{P}^{(1)}(\omega) + \vec{P}^{(2)}(\omega) + \vec{P}^{(3)}(\omega) \dots$$

**Linear optics**

Only one Fourier

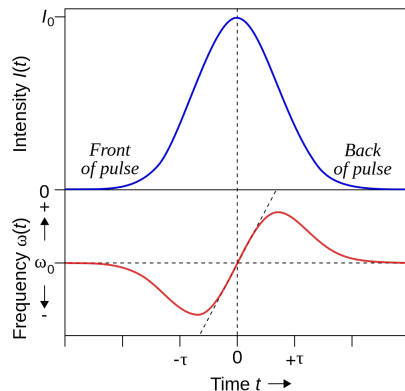
Component is involved

**Nonlinear optics**

Possibility to have interactions

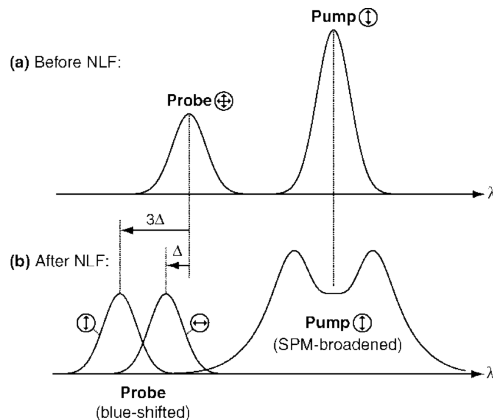
between several Fourier components

A pulse (top curve) propagating through a nonlinear medium undergoes a self-frequency shift (bottom curve) due to self-phase modulation. The front of the pulse is shifted to lower frequencies, the back to higher frequencies. In the centre of the pulse the frequency shift is approximately linear. <sup>2</sup>



<sup>2</sup>wikipedia

Illustration of how the spectral shift produced through cross-phase modulation depends upon the relative polarization state of the pump and probe. The greatest shift is produced when the pump and probe are copolarized, while the spectral shift is x3 smaller when the pump and probe are cross-polarized.<sup>3</sup>



<sup>3</sup>www.semanticscholar.org

# The Intensity-Dependent Refractive Index

The intensity-dependent refractive index and absorption changes associated with SPM and TPA affect the complex refractive index:

$$n = n_0 + n_2 I - i \frac{\lambda}{4\pi} (\alpha_0 + \alpha_2 I)$$

where  $I$  is the intensity,  $n_0$  represents the usual, weak-field refractive index, and  $\alpha_0$  is the linear absorption coefficient.

The Kerr coefficient:

$$n_2 = \frac{1}{n_0^2 \epsilon_0 c} \frac{3}{4} \Re(\chi^{(3)})$$

The TPA coefficient:

$$\alpha_2 = \frac{-\omega}{n_0^2 \epsilon_0 c} \frac{3}{2} \Im(\chi^{(3)})$$

# The Intensity-Dependent Refractive Index

A figure of merit (FOM) is often used to compare the magnitude of the Kerr coefficient  $n_2$  with the strength of the TPA coefficient  $\alpha_2$ :

$$\text{FOM} = \frac{1}{\lambda} \frac{n_2}{\alpha_2}$$

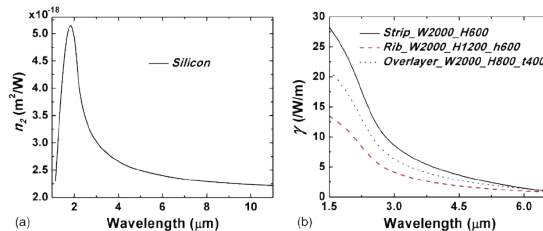
The nonlinear waveguide parameter defined by:

$$\gamma = \frac{2\pi}{\lambda} \frac{n_2}{A_{\text{eff}}^{(3)}}$$

where  $A_{\text{eff}}^{(3)}$  third-order nonlinear effective area



# The Intensity-Dependent Refractive Index



**Figure 13:** (a) Nonlinear refractive index of silicon and (b) the nonlinear coefficients in strip, rib, and conformal overlayer waveguides.

4

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<sup>4</sup>Silicon-on-Nitride Waveguide With Ultra low Dispersion Over an Octave-Spanning Mid-Infrared Wavelength Range

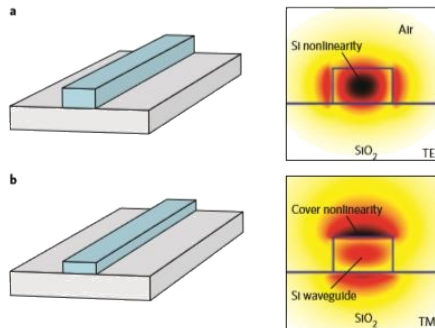
TABLE 4.1.1 Typical values of the nonlinear refractive index <sup>a</sup>

Mechanism	$n_2$ (cm <sup>2</sup> /W)	$\chi_{1111}^{(3)}$ (m <sup>2</sup> /V <sup>2</sup> )	Response Time (sec)
Electronic polarization	10 <sup>-16</sup>	10 <sup>-22</sup>	10 <sup>-15</sup>
Molecular orientation	10 <sup>-14</sup>	10 <sup>-20</sup>	10 <sup>-12</sup>
Electrostriction	10 <sup>-14</sup>	10 <sup>-20</sup>	10 <sup>-9</sup>
Saturated atomic absorption	10 <sup>-10</sup>	10 <sup>-16</sup>	10 <sup>-8</sup>
Thermal effects	10 <sup>-6</sup>	10 <sup>-12</sup>	10 <sup>-3</sup>
Photorefractive effect <sup>b</sup>	(large)	(large)	(intensity-dependent)

<sup>a</sup> For linearly polarized light.

<sup>b</sup> The photorefractive effect often leads to a very strong nonlinear response. This response usually cannot be described in terms of a  $\chi^{(3)}$  (or an  $n_2$ ) nonlinear susceptibility, because the nonlinear polarization does not depend on the applied field strength in the same manner as the other mechanisms listed.

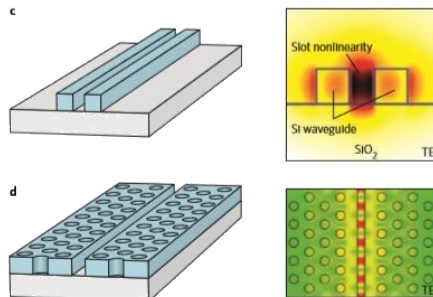
# Silicon Waveguide



**Figure 14:** (a) Strip waveguide using silicon nonlinearity in core. (a) Strip waveguide using cover nonlinearities.

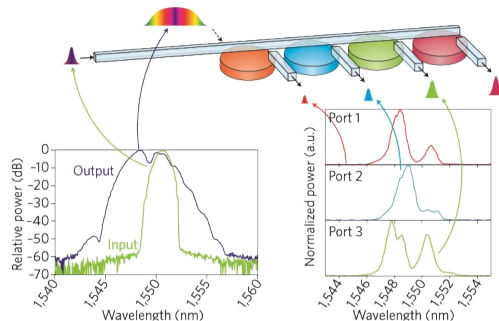
## Silicon Waveguide

The transverse electric (TE) mode has its dominant electric field component parallel to the substrate plane, whereas the transverse magnetic (TM) dominant field contribution is perpendicular to the substrate [2].



**Figure 15:** (c) Slot waveguide using nonlinearities in the slot. (d) Slot slow-light waveguide.

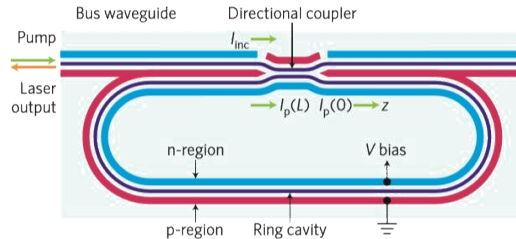
# Nonlinear Silicon Light Sources



A short input pulse is spectrally broadened in a nonlinear silicon wire waveguide using SPM and subsequently distributed to several outputs, each at different wavelengths <sup>6</sup>.

<sup>6</sup>[2]

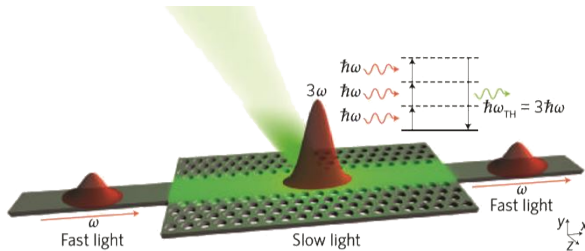
# Nonlinear Silicon Light Sources



A silicon Raman ring laser within a ring laser cavity. A reverse-bias p-i-n structure is placed across the silicon rib waveguide to ‘sweep out’ TPA-induced carriers.<sup>7</sup>

<sup>7</sup>[2]

# Nonlinear Silicon Light Sources



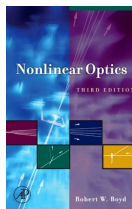
Using THG to generate green light in a slow-light silicon photonic crystal waveguide. <sup>8</sup>.

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<sup>8</sup>[2]

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