Nonlinear Optics

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This course is aimed to help students mastering the principles and technical materials of a graduate-level Nonlinear Optics. The following topics are covered: Macroscopic Theory of Optical Susceptibility Tensors, Second-Order Nonlinear Optical Effects, Third-Order Nonlinear Optical Effects, and Time-Resolved Nonlinear Optical Spectroscopies, *etc*.. It is required the audience of this course to possess a background of Applied Mathematics and Electromagnetism.

Lecture notes will be posted whenever possible. Not all lectures are available for distribution.

Textbook and References

1. **Y. R. Shen, "The Principles of Nonlinear Optics", Wiley-Interscience , 1984.**
2. **P. N. Bu tcher & D. Cotter, 'The Element of Nonlinear Optics, Cambridge University Press, 1990.**
3. **Robert Boyd, Nonlinear Optics, Academic Press, Inc., 1992.**

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Chapter 1. Introduction

1.1 Historical Background

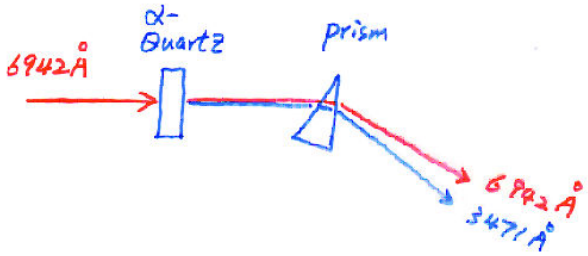
During the pre-laser age, some NLO phenomena and research had been reported. These include Pockels and Kerr EO effects, and some predictions by calculations of two-photon absorption published by Meyer *et al*.

1960 Invention of Ruby laser by Maiman

This invention provided a light source with high luminosity which had never been produced before.

1961 SHG by Franken (see PRL 7, 118 (1961))

But the conversion efficiency is as low as 10^{-10}%!



Nowadays typical SHG has achieved as large as 90%! This is because phase matching scheme was proposed by Giordmain and Maker using birefringence of a crystal.

Currently research direction of NLO can be separated into:

(1) **NLO Phenomena:** Investigating various NLO processes in media excited by ultrashort, ultrastrong laser pulses

(2) **Techniques:** Applying various NLO techniques for characterizing materials to yield spectroscopic or dynamical information of media.

Common features in each NLO process

(1) An intense light field $\vec{E}(\vec{r},t)$ induced a response in a medium $\vec{P}$

This part is usually governed by constitutive equations

$$P = P(E) \xrightarrow{\text{local response}}$$

$$P_\alpha(r,E) = P_\alpha^{(0)}(r,E=0) + \sum_\beta \left(\frac{\partial P_\alpha}{\partial E_\beta}\right)\Big|_0 \cdot E_\beta(r,t) + \ldots$$

(2) Medium in reacting modifies the optical fields in a nonlinear way. This step is governed by Maxwell's Equations for nonmagnetic materials:

$$\nabla \cdot D = \nabla \cdot (E + 4\pi P) = \nabla \cdot E + 4\pi \nabla \cdot (P_L + P_{NL})$$

$$\nabla \cdot B = 0$$

$$\nabla \times E = \frac{1}{c}\frac{\partial B}{\partial t} \quad \nabla \times H = \frac{1}{c}\frac{\partial D}{\partial t}$$

By using the last two curl equations, we have

$$\nabla^2 E - \nabla(\nabla \cdot E) - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} - \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}(P_L + P_{NL}) = 0$$

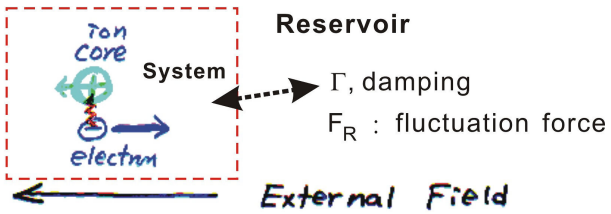
By combining the linear response into second-order temporal derivative of the field, then the equation becomes

$$\boldsymbol{\nabla^2 E - \frac{\varepsilon}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P_{NL}}{\partial t^2}}.$$

This is valid for non-conductive media with $\nabla \cdot E = 0$.

Origin of Optical Nonlinearity (Microscopic Picture of Constitute Equation) with Classical Approach

Consider a dielectric optical material consisting of a collection of charged particles of electrons and ion cores:



In a conductor, the charged carriers are free to move in an electric field. But for a dielectric material, the charged particles are bound together.

Note:

(i) the motion of the charged carriers is transitory when the field is first applied;

(ii) That will induce a collection of induced electric dipole moment.

The induced polarization ***P***(r, t) is defined to be the induced dipole moment per unit volume

(iii) Note light wave oscillates at frequency of $10^{13} \rightarrow 10^{17}$ Hz (from IR to UV);

(iv) Ion cores are massive compared with electrons (This leads to the well-known Born-Oppenheimer (BO) approximation).

Thus an anharmonic oscillator model can be constructed

$$damped\ harmonic\ oscillator + nonlinear\ response\ of\ the\ medium =$$
$$m[\ddot{q}(t) + \gamma\dot{q}(t) + \omega_0^2 q(t) - (\xi^{(2)}q^2 + \xi^{(3)}q^3 + ...)] = -eE(t)$$

Here γ=damping constant; ω_0 = resonant frequency; $\hbar\omega_0 \sim$ 3eV; and q = displacement from the equilibrium position.

Now neglect nonlinear response for a moment and consider harmonic response to an applied electric field

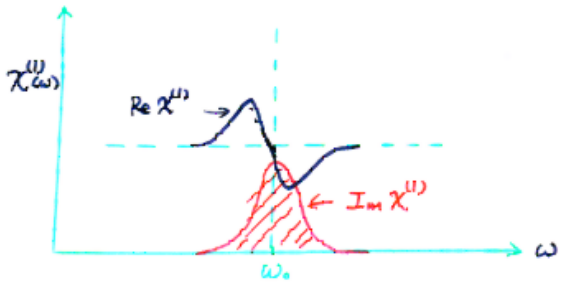
$E(t) = E_0[e^{-i\omega t} + e^{+i\omega t}]$.

Employing the solution *an salz* $q(t) = Ae^{-i\omega t} + c.c.$.

$$m[-\omega^2 A - i\omega\gamma A + \omega_0^2 A]e^{-i\omega t} = -eE_0 e^{-i\omega t}$$
$$\Rightarrow$$
$$q(t) = \frac{-eE_0}{m}\frac{e^{-i\omega t}}{(\omega_0^2 - i\gamma\omega - \omega^2)}$$

Now consider the polarization
P=polarization=dipole moment per unit volume=

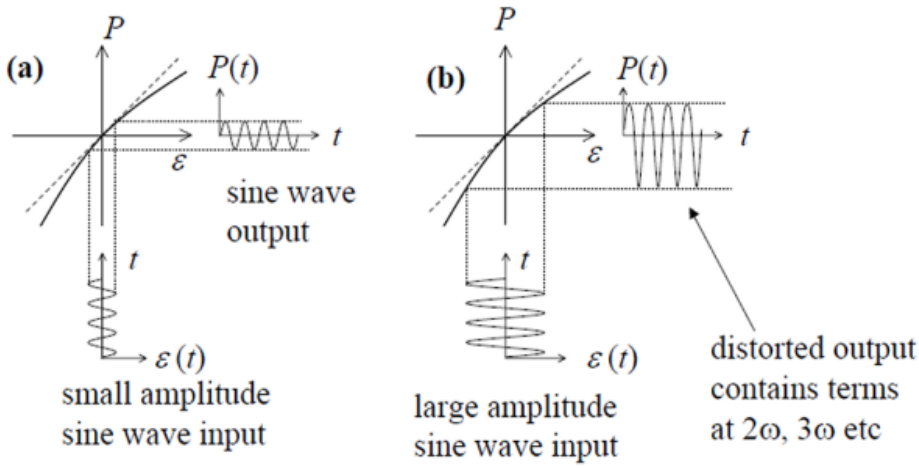
$$N(-e)q = \chi^{(1)}E_0 e^{-i\omega t} + c.c.$$
$$\Rightarrow$$
$$\chi^{(1)} = linear\ susceptibility = \frac{Ne^2}{m}\frac{1}{(\omega_0^2 - i\gamma\omega - \omega^2)}$$



Note that the electric displacement vector ***D***
$D = E + 4\pi P = \varepsilon E$
$\Rightarrow \varepsilon = 1 + 4\pi\chi^{(1)} = n_r + i\kappa$.

We know that the linear dependence of ***P*** on ***E*** is valid only when d is small, where ***P*** oscillates at the same frequency as ***E*** in the linear approximation.

When q is large, the restoring force of the medium could become significantly nonlinear



When q is large, the response is distorted and contains significant components oscillating at the harmonic frequency 2ω, 3ω, etc., and a d.c. component.

$$\ddot{q}(t)+\gamma\dot{q}(t)+\omega_0^2 q(t)+\xi^{(2)}q^2=\frac{-e}{m}[E_{10}(e^{-i\omega_1 t}+e^{i\omega_1 t})+E_{20}(e^{-i\omega_2 t}+e^{i\omega_2 t})]\,.$$

No analytic solution for the above equation. But when $\boldsymbol{\xi^{(2)}q^2}$ is small, perturbation technique can be used.

Let $q(t)=q^{(1)}(t)+q^{(2)}(t)+\ldots$ *and note* $P=-Neq$,

we first linearize the equation to yield

$$q^{(1)}(\omega_i)=\frac{-eE_{i0}}{m}\frac{e^{-i\omega_i t}}{(\omega_0^2-i\gamma\omega_i-\omega_i^2)}\,.$$

$$q^{(1)}(t)=q^{(1)}(\omega_1)+q^{(1)}(\omega_2)+c.c.$$

Then approximate $\boldsymbol{\xi^{(2)}q^2}$ by $\boldsymbol{\xi^{(2)}[q^{(1)}]^2}$. Thus the solutions for $\boldsymbol{q^{(2)}}$ become

$$q^{(2)}=q^{(2)}(2\omega_1)+q^{(2)}(2\omega_2)+q^{(2)}(\omega_1+\omega_2)+q^{(2)}(\omega_1-\omega_2)+q^{(2)}(0)+c.c....,$$

where

$$q^{(2)}(\omega_1\pm\omega_2)=\frac{-2\xi^{(2)}(e/m)^2E_{10}E_{20}}{(\omega_0^2-i\gamma\omega_1-\omega_1^2)(\omega_0^2\mp i\gamma\omega_2-\omega_2^2)}\frac{e^{-i(\omega_1\pm\omega_2)t}}{[\omega_0^2-i\gamma(\omega_1\pm\omega_2)-(\omega_1\pm\omega_2)^2]}$$

$$=\frac{-2\xi^{(2)}(e/m)^2E_{10}E_{20}}{D(\omega_1)D(\omega_2)D(\omega_1\pm\omega_2)}e^{-i(\omega_1\pm\omega_2)t}\,,$$

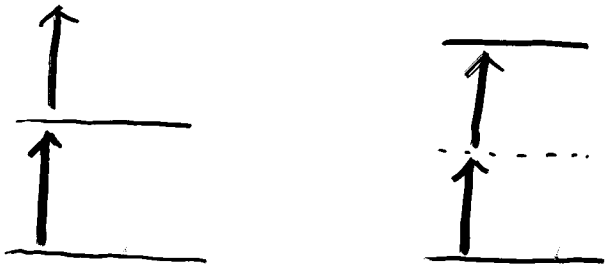
and $\boldsymbol{D(\omega)=(\omega_0^2-i\Gamma\omega-\omega^2)}$

$$q^{(2)}(2\omega_i) = \frac{-2\xi^{(2)}(e/m)^2 E_{i0}{}^2}{D(\omega_i)D(\omega_i)D(2\omega_i)} e^{-i2\omega_i t}$$

Therefore, we can express the nonlinear polarization as

$$\begin{aligned} P^{(2)}(2\omega_i) &\equiv -Neq^{(2)}(2\omega_i) \\ &\equiv \chi^{(2)}(2\omega_i)E(\omega_i)E(\omega_i) \\ &\Rightarrow \qquad . \\ \chi^{(2)} &= \text{sec}\,ond - order\ nonlinear\ susceptibility \\ &= Ne\xi^{(2)}(\frac{e}{m})^2 \frac{1}{D(\omega_i)D(\omega_i)D(2\omega_i)} \end{aligned}$$

The resonances of $\boldsymbol{\chi^{(2)}(2\omega_i)}$ can occur at $\boldsymbol{\omega_i = \omega_0}$ and $\boldsymbol{2\omega_i = \omega_0}$.



$\boldsymbol{P^{(2)}(2\omega_i)}$ oscillates at $\boldsymbol{2\omega_i}$ and will radiate optical field with frequency $\boldsymbol{2\omega_i}$.

Several factors can be used to increase the response of medium

(a) Phase matching: The induced dipoles are forced to radiate in phase and therefore will lead to coherent radiation.

(b) Resonant enhancement: $\boldsymbol{D(\omega_1) \simeq 0}$ ***or*** $\boldsymbol{D(2\omega_1) \simeq 0}$.

(c) $\boldsymbol{\chi^{(2)}}$ depends on $\boldsymbol{\xi^{(2)}}$, *i.e.*, depends on anharmonicity of medium.

When $\boldsymbol{\omega_0 \gg \omega_i}$ far below the absorption edge of the medium, $\boldsymbol{D(\omega_i) \simeq \omega_0^2}$,

$$\left|\frac{P^{(2)}}{P^{(1)}}\right| = \frac{Ne\xi^{(2)}(e/m)^2 E_{i0}{}^2}{D^2(\omega_1)D(2\omega_1)} \frac{D(\omega_1)}{eN(e/m)E_{i0}} \simeq \frac{e\xi^{(2)}E_{i0}}{m\omega_0^4}.$$

Now note that

$$m\omega_0^2 d \sim |-eE_{at}| \gg m\xi^{(2)}d^2 \quad \Rightarrow$$

$$eE_{at} \sim m\omega_0^2 d \ll m\omega_0^2 \left|\frac{\omega_0^2}{\xi^{(2)}}\right| = \frac{m\omega_0^4}{\xi^{(2)}}$$

Therefore, $\left|\frac{P^{(2)}}{P^{(1)}}\right| < \frac{E_{i0}}{E_{at}}$.

In general, $\left|\frac{P^{(n+1)}}{P^{(n)}}\right| < \frac{E_{i0}}{E_{at}}$.

Typically, for $E_{at} \sim 3\times10^{8}\ V/cm$ and $E_{10} \sim 30\ V/cm$ for an optical beam with

$I \sim 2.5\ W/cm^2$, $\left|\frac{P^{(2)}}{P^{(1)}}\right| \sim \frac{E_{i0}}{E_{at}} \sim 10^{-7}$.

1.3 Nonlinear Optical Processes

As an example, let's consider NLO processes that modify the index of refraction of medium. Assuming this medium is subject to an intense electric field, and the polarization of the medium is weak compared to the binding forces between the electrons and nuclei, one can then express the polarization in a power series of the field strength E

$$P = \sum_i P^{(i)} = \chi^{(1)} \cdot E + \chi^{(2)} : EE + \chi^{(3)} \vdots EEE + \ldots . \qquad (1.3.1)$$

Here $\chi^{(2)}_{ijk}$ is a third-rank tensor, denotes the second-order nonlinear optical susceptibility; $\chi^{(3)}_{ijkl}$ is a fourth-rank tensor, denotes the third-order nonlinear optical susceptibility, *etc*.

Now recast Equation (1.3.1) as

$$P = \sum_i P^{(i)} = \chi^{(1)} \cdot E + \chi^{(2)} : EE + \chi^{(3)} \vdots EEE + \ldots = \chi_{eff}(E) \cdot E .$$

Here $\chi_{eff}(E)$ is an effective optical susceptibility and will depend on the field strength. Now let

$$E = dc + optical\ \ field\ of\ \ frequency\ \omega,$$

then

$$P = \sum_i P^{(i)} = \chi^{(1)}[E(0) + E_0\cos(\omega t + kz)] + \chi^{(2)}[E(0) + E_0\cos(\omega t + kz)]^2 + \chi^{(3)}[E(0) + E_0\cos(\omega t + kz)]^3 + \ldots .$$

Consider $E_0\cos(\omega t + kz)$-term only:

$$P = \sum_i P^{(i)} = \chi^{(1)}E_0\cos(\omega t + kz) + 2\chi^{(2)}E(0)E_0\cos(\omega t + kz) + 3\chi^{(3)}E(0)^2E_0\cos(\omega t + kz) + \frac{3}{4}\chi^{(3)}{E_0}^3\cos(\omega t + kz) = \chi_{eff}E_0\cos(\omega t + kz)$$

We can now define a nonlinear index of refraction to be

$$n^2 = 1 + 4\pi\chi_{eff} = 1 + 4\pi[\chi^{(1)} + 2\chi^{(2)}E(0) + 3\chi^{(3)}E(0)^2 + \frac{3}{4}\chi^{(3)}E_0^2.$$

The change of the index of refraction caused by the external field becomes

$$\begin{aligned} n^2 - n_0^2 &= (n - n_0)(n + n_0) \\ &= 8\pi\chi^{(2)}E(0) + 12\pi\chi^{(3)}E(0)^2 + 3\pi\chi^{(3)}E_0^2 \\ \Rightarrow n &= n_0 + \frac{4\pi\chi^{(2)}}{n_0}E(0) + \frac{6\pi\chi^{(3)}}{n_0}E(0)^2 + \frac{3\pi\chi^{(3)}}{2n_0}E_0^2 \end{aligned} \qquad (1.3.2)$$

We can also define the light intensity in cgs unit as

$I = \frac{c\sqrt{\varepsilon}}{8\pi}|E_0|^2$ and rewrite Equation (1.3.2) as

$$\begin{aligned} n &= n_0 + \frac{4\pi\chi^{(2)}}{n_0}E(0) + \frac{6\pi\chi^{(3)}}{n_0}E(0)^2 + \frac{12\pi^2\chi^{(3)}}{c{n_0}^2}I \\ &= n_0 + n_1E(0) + n_2(0)E(0)^2 + n_2(\omega)I(\omega) \end{aligned}.$$

(a) $n_1 = 4\pi\chi^{(2)}/n_0$ denotes a linear electro-optical effect, discovered by Pockels.

(b) $n_2(0) = 6\pi\chi^{(3)}/n_0$ describes quadratic electro-optical effect, which is often also called dc Kerr effect.

(c) $n_2(\omega) = 12\pi^2\chi^{(3)}/(c{n_0}^2)$ describes the optical Kerr effect, which is the basis of all high-speed all-optical switching.

1.3.1 Second-Order Nonlinear Optical Effects: $\chi^{(2)}_{ijk}(-\omega_3; \omega_1, \omega_2)$

This is a three-wave mixing process where the waves exchange energy with one another through the interaction of the nonlinear medium.

(i) Second-Harmonic Generation (SHG) $\chi^{(2)}_{ijk}(-2\omega; \omega, \omega)$: $\omega + \omega \to 2\omega$

(ii) Linear Electro-Optical Effect (EO) $\chi^{(2)}_{ijk}(-\omega; \omega, 0)$: $\omega + dc \to \omega$: Note the

change in refractive index due to the linear electro-optical effect is commonly defined as $\Delta(\frac{1}{n})_{ij} = [\frac{1}{\varepsilon(E)}]_{ij} - [\frac{1}{\varepsilon(0)}]_{ij} = r_{ijk} E_k(0)$, where

$r_{ijk} = -8\pi\chi^{(2)}/n_0^4$ $(in\ cgs)$ is also called EO coefficient.

(iii) Optical Rectification (OR) $\chi_{ijk}^{(2)}(-0;\omega,-\omega)$: $\omega-\omega\to 0$: The interaction of an optical wave at frequency ω with itself, generates a dc polarization in the medium. Recall the Kleinman symmetry (to be discussed in the following chapter), $\chi_{ijk}^{(2)}(-0;-\omega,\omega) = \chi_{ijk}^{(2)}(-\omega;0,-\omega)$ indicates the optical rectification and the dc electro-optical effects are clearly related.

(iv) Sum-Frequency Generation (SFG) $\chi_{ijk}^{(2)}(-\omega_3;\omega_1,\omega_2)$: $\omega_1+\omega_2\to\omega_3$

(v) Difference-Frequency Generation (DFG) $\chi_{ijk}^{(2)}(-\omega_2;\omega_3,-\omega_1)$: $\omega_3-\omega_1\to\omega_2$:

The optical parametric amplification (OPA)

$\chi_{ijk}^{(2)}(-\omega_2;\omega_3,-\omega_1)$: $\omega_3(pump)-\omega_1(weak\ signal)\to\omega_1(amplified\ signal),\omega_2(idler)$

, which is similar to DFG..

1.3.2 Third-Order Nonlinear Optical Effects: $\chi_{ijkl}^{(3)}(-\omega_4;\omega_1,\omega_2,\omega_3)$

(i) Third-Harmonic Generation (THG) $\chi_{ijkl}^{(3)}(-3\omega;\omega,\omega,\omega)$: $\omega+\omega+\omega\to 3\omega$

$P_i^{(3)}(3\omega) = [\frac{1}{4}]\chi_{ijkl}^{(3)}(-3\omega;\omega,\omega,\omega)E_j(\omega)E_k(\omega)E_l(\omega)$.

We have to include the number of distinct permutation possibilities of the incident fields, which is equal to 1 in this case. Note that since $E_j(\omega,t)$ and $P_j^{(3)}(3\omega,t)$ are real, we must let $E_j(\omega,t) = [\frac{1}{2}][E_j(\omega)e^{i(\omega t-k_\omega z)} + E_j^*(\omega)e^{-i(\omega t-k_\omega z)}]$ and

$P^{(3)}{}_i(3\omega,t) = [\frac{1}{2}][P_i(3\omega)e^{i(3\omega t-k_{3\omega}z)} + P_i^*(3\omega)e^{-i(3\omega t-k_{3\omega}z)}]$.

Therefore, we find

$$2[from\ P(3\omega)]\times\frac{1[\#distinct\ driving-field\ permutation]}{2\times2\times2[from\ each\ incident\ field]} = \frac{1}{4}$$

(ii) Self-Action

These phenomena are the third-order NLO processes with a third-order NLO polarization

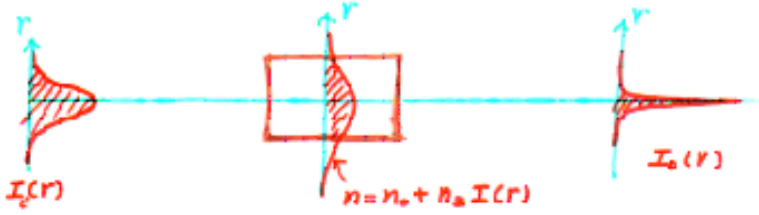
$$P_i^{(3)}(\omega) = [\frac{3}{4}]\,\mathrm{Re}[\chi_{iikk}^{(3)}(-\omega;\omega,\omega,-\omega)]E_i(\omega)\,|\,E_k(\omega)\,|^2$$

The nonlinear polarization is created in the medium at the same frequency as the incident beam

$$\begin{aligned} E(\omega,t) &= E_0(t)e^{i(\omega t - k_\omega z)} + c.c. \\ &= \bar{E}_0(t)e^{i\phi_0(t)} \cdot e^{i(\omega t - k_\omega z)} \end{aligned}.$$

A. Self-Focusing/Defocusing

These phenomena can be understood by combining the effect of $n_2(\omega)$ and the spatial variation of the laser intensity:



When $\chi^{(3)} > 0,\quad n_2 = 12\pi^2\chi^{(3)}/(cn_0^2) > 0$. Then $n(r) = n_0 + n_2 I(r)$ acts like a focusing lens.

B. Self-Phase Modulation (SPM)

When consider the temporal variation of the incident laser pulse, the temporal behavior of the induced refractive index change can be generated

$\Delta n(t) = n - n_0 = n_2 I(t)$.

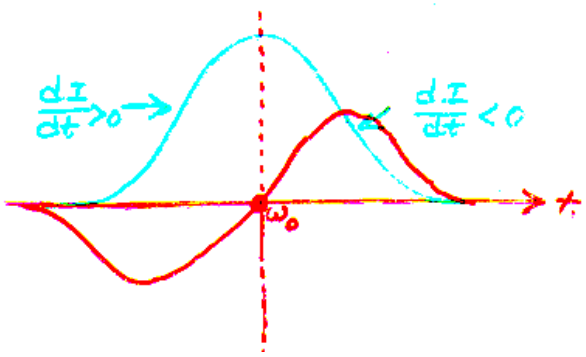
This can lead to a broadening of the frequency profile of the laser pulse field.

$$\begin{aligned} \phi(t,z) &= \textit{phase front of the pulse} \\ &= \omega_0 t - \frac{n\omega_0}{c} z = \omega_0 t - \frac{n_0 + n_2 I(t)}{c}\omega_0 z \end{aligned}.$$

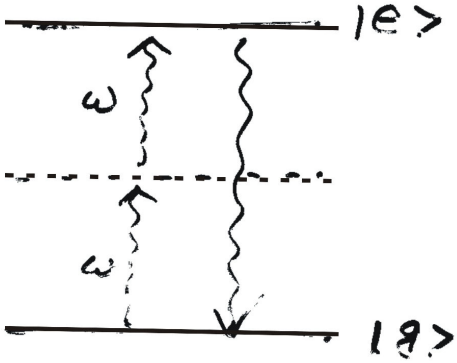
Defining the instantaneous frequency as

$\tilde{\omega}(t) \equiv d\phi(t)/dt = \omega_0 - \frac{n_2 z}{c}\omega_0 \frac{dI(t)}{dt}$, which shall depend on t.

If $n_2(\omega) > 0$, the instantaneous frequency first experiences a frequency upchirping at the leading edge of the pulse and then down chirping at the trailing edge of the pulse as depicted in the following diagram.



(iii) Two-Photon Absorption (TPA)



This NLO phenomenon can be explained with a NLO polarization induced in a medium

$$P_i^{(3)}(\omega) = [\frac{3}{4}]\,\mathrm{Im}[\chi_{iikk}^{(3)}(-\omega;\omega,\omega,-\omega)]E_i(\omega)\,|\,E_k(\omega)\,|^2 .$$

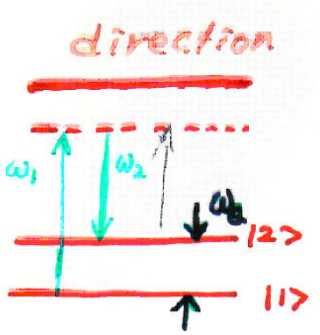
iv) Degenerate Four-Wave Mixing (DFWM)

$$P_i^{(3)}(\omega) = [\frac{3}{4}]\,\chi_{ijkl}^{(3)}(-\omega;\omega,\omega,-\omega)E_j(\omega)E_k(\omega)E_l{}^*(\omega)$$

Note that: Although $E_j(\omega), E_k(\omega), E_l(\omega)$ have the same frequency, they can have different propagation direction and polarization. The generated beam can also propagate along a different direction with different polarization.

v) Coherent Raman Effects

If $\omega_1 - \omega_2 = \omega_Q$ =Raman active mode, we can observe resonant effect. This is a useful spectroscopic technique to determining the structure of a molecular medium.



Several technical variations can be developed from this concept, which include:

v.1 Raman Induced Kerr Effect (RIKES)

$$P_i^{(3)}(\omega_i) = [\frac{3}{4}]\,\mathrm{Re}[\chi_{iikk}^{(3)}(-\omega_1;\omega_1,\omega_2,-\omega_2)]E_i(\omega_1)\,|\,E_k(\omega_2)\,|^2$$

v.2 Coherent Stokes Raman Spectroscopy (CSRS)

$$P_i^{(3)}(\omega_s = 2\omega_2 - \omega_1) = [\frac{3}{2}]\,\mathrm{Im}[\chi_{ijkl}^{(3)}(-\omega_s;\omega_2,\omega_2,-\omega_1)]E_j(\omega_2)E_k(\omega_2)E_l^*(\omega_1)$$

where $\omega_1 > \omega_2$

v.3 Coherent Anti-Stokes Raman Spectroscopy (CARS)

$$P_i^{(3)}(\omega_A = 2\omega_1 - \omega_2) = [\frac{3}{2}]\,\mathrm{Im}[\chi_{ijkl}^{(3)}(-\omega_A;\omega_1,\omega_1,-\omega_2)]E_j(\omega_1)E_k(\omega_1)E_l^*(\omega_2)$$

where $\omega_1 > \omega_2$

v.4 Electric Field-Induced Second-Harmonic Generation (EFISH)

When an isotropic medium is biased in a dc electric field, its centro-symmetric structure of electronic distribution can be distorted, renders it to become non centrosymmetric structure. The material can therefore generate second harmonic output at 2ω when it is excited with an optical pulse with a frequency of ω. The phenomenon, which is similar to the optical second-harmonic generation from a noncentrosymmetric crystal, is called Electric Field-Induced Second-Harmonic Generation (EFISH) and in fact belongs to a third-order NLO process, describing by the following NLO polarization induced in the medium.

$$P_i^{(3)}(\omega_3 = 2\omega_1) = [\frac{3}{2}][\chi_{ijkl}^{(3)}(-\omega_3;\omega_1,\omega_1,0)]E_j(\omega_1)E_k(\omega_1)E_l^*(0)$$

C. Resonant Nonlinearity

Under resonant condition ($\omega \sim \omega_0$), one can also observe a large dynamic

nonlinearity derived from an optical absorption process, which at sufficiently high intensity becomes light-intensity dependent

$$\alpha_0(\omega) \rightarrow (I\ increase) \rightarrow \alpha(\omega, I) = \frac{\alpha_0(\omega)}{1+(I/I_s)}.$$

From

$n(\omega) = \frac{c}{\pi} P\int_0^\infty \frac{d\omega' \alpha(\omega')}{\omega'^2 - \omega^2}$, $n(\omega)$ is also light-intensity dependent. In fact, any process which can lead to an intensity-dependent absorption shall give rise to a dynamic third-order optical nonlinearity.

Summary of Chapter 1