NONLINEAR OPTICS

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7 Lectures (7x1h30) 1 Homework 6 tutorial sessions (including one in numerical simulation)

Lecture 4 /7 : learning outcomes

By the end of this lecture, students will be able to...

• Evaluate nonlinear interaction performances/efficiencies under approximations that should be specified, explained and justified (T2)

By the end of this lecture, students will be skilled at...

• deriving and solving the nonlinear wave equation in a parametric situation under the undepleted pump approximation (S3)

• Determining the phase matching conditions for a given nonlinear interaction and achieving/fulfilling this condition by exploiting birefringence properties of materials (S2)

By the end of this lecture, students will understand ...

• Nonlinear optics is an essential tool to create novel optical frequencies(U4) generated through the interaction of incident beams within nonlinear materials

• Nonlinear effects are subject to phase matching conditions (U5)



Lecture 4 - Content

2nd ORDER NONLINEARITIES

- The Manley-Rowe Relations
- Three wave mixing in $\chi^{(2)}$ materials
- Second Harmonic Generation
 - Phase matching consideration
 - Reminder : propagation in a linear anisotropic material
 - Phase matching condition in birefringent materials
- Frequency generation Parametric processes
 - optical parametric fluoresence and amplification
 - optical parametric oscillation : OPO
- Quasi-phase matched materials



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1- The Manley-Rowe relations



General description of the **NL interaction of 3 waves** @ ω_1 , ω_2 , and ω_3 (with $\omega_3 = \omega_1 + \omega_2$) in a <u>2nd order NL lossless</u> medium :





Consistent with quantum-mechanical interpretation

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2 - Three-wave Mixing Interactions in a χ⁽²⁾ material



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Nonlinearities in $\chi^{(2)}$ leads to 3-wave mixing interactions. They are governed by :

• Energy conservation law

 $\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_3$

• Phase-matching condition = Law of conservation of momemtum

$$\overrightarrow{k_1} + \overrightarrow{k_2} = \overrightarrow{k_3}$$

(Important : vectorial relation)



(b)

 \overline{k}_3

 k_2

(a)

2 - Three-wave Mixing Interactions in a χ⁽²⁾ material

Nonlinearities in $\chi^{(2)}$ leads to 3-wave mixing interactions :

Sum-Frequency Generation (SFG)



Second Harmonic Generation (SHG)



Optical Parametric Fluoresence



Optical Parametric Amplification (APO)



Optical Parametric Oscillation (OPO)



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2nd Harmonic Generation



2nd Harmonic Generation



2nd Harmonic Generation

1. Undepleted pump approximation regime



Phase matching condition

About the difficulty to achieve the phase matching condition :

In general, the refractive index for lossless materials shows a NORMAL DISPERSION : the refractive index is increasing with the frequency



The most common procedure for achieving the phase matching condition :



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• Maxwell's equations

no free charges, no free currents, nonmagnetic

$$\begin{cases} \nabla \times \mathcal{E} = -\frac{\partial B}{\partial t} & \nabla \cdot \mathcal{D} = 0 \implies \nabla \cdot \mathbf{D} = i\mathbf{k} \cdot \mathbf{D} = 0 \\ \nabla \times H = \frac{\partial D}{\partial t} & \nabla \cdot B = 0, \implies \nabla \cdot \mathbf{H} = i\mathbf{k} \cdot \mathbf{H} = 0 \end{cases} \stackrel{\mathbf{k} \perp \mathbf{D}}{\mathbf{k} \perp \mathbf{H}} \\ D(\omega) = \underline{\epsilon} \mathbf{E} \quad B = \mu_0 \mathbf{H} \end{cases}$$
Poynting vector : $\mathbf{S} = \mathbf{E} \times \mathbf{H}$

$$\vec{k} \perp \vec{D}$$

$$\vec{k} \perp \vec{D}$$

$$\vec{k} \perp \vec{D}$$

$$\vec{k} \perp \vec{E}$$

$$\alpha : walk-off angle$$

$$\alpha : walk-off angle$$
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Linear Wave Eq. in an anisotropic material

Wave equation

In the time domain

$$abla imes
abla imes oldsymbol{E}(t) + rac{1}{c^2} rac{\partial^2 oldsymbol{E}}{\partial t^2} = -\mu_0 rac{\partial^2 oldsymbol{P}}{\partial t^2}$$

In the frequency domain

$$abla imes \nabla imes \boldsymbol{E}(\omega) - \frac{\omega^2}{c^2} \boldsymbol{E}(\omega) = \omega^2 \mu_0 \boldsymbol{P}(\omega)$$

The complex field amplitude of the plane wave is defined as

$$\boldsymbol{E}(\omega) = E(\omega)e^{\imath \boldsymbol{k}\cdot\boldsymbol{r_e}}$$

Polarization vector



Linear Wave Eq. in an anisotropic material

Wave equation
$$k \times (k \times e) + \frac{\omega^2}{c^2} \underline{\epsilon}(\omega)e = 0$$
 $\vec{k} = |k|\vec{s}$ $D(\omega) = \underline{\epsilon} E$ $\underline{\epsilon} = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \end{pmatrix}$ $\vec{s} = (s_x, s_y, s_z)$ $\underline{\epsilon}(\omega) = \epsilon_0 (1 + \underline{\chi}^{(1)}(\omega))$ New coordinate
systemUnit vector $\underline{\epsilon} = \begin{pmatrix} \epsilon_{xx} & 0 & 0 \\ 0 & \epsilon_{yx} \end{pmatrix}$ Principal
axis systemFresnel's equation $\underline{s} = \begin{pmatrix} \epsilon_{xx} & 0 & 0 \\ 0 & \epsilon_{yx} \end{pmatrix}$ $\underline{s} = \frac{\epsilon_{xx}^2}{n^2 - n_x^2} + \frac{s_y^2}{n^2 - n_y^2} + \frac{s_z^2}{n^2 - n_z^2} = \frac{1}{n^2}$
(in the principal axis system) $n_x n_y n_z$ are the principal axis system $n_x n_y n_z$ are the principal refractive indicesFor any direction of propagation, two waves can propagate
(independently) with phase velocity $v_1 = c/n_1$ and $v_2 = c/n_2$ where
 n_1 and n_2 are solutions of the Fresnel's equationN. Dubreuli - NONLINEAR OPTICS

Linear Wave Eq. in an anisotropic material

The index of ellipsoid : method to determine the refractive indices and the related directions of the electric fields

The electric energy density stored in a medium $w_e = \frac{1}{2} \boldsymbol{E} \cdot \boldsymbol{D} = \frac{1}{2} \sum_{j,k} E_k \epsilon_{kj} E_j$

$$2w_e = \epsilon_{XX}E_X^2 + \epsilon_{YY}E_Y^2 + \epsilon_{ZZ}E_Z^2 + 2\epsilon_{YZ}E_YE_Z + 2\epsilon_{XZ}E_XE_Z + 2\epsilon_{XY}E_XE_Y$$

→ The surface of constant energy forms an ellipsoid

 $\epsilon_{XX}, \epsilon_{YY}, \epsilon_{ZZ} > 0$ In a lossless material

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→ In the principal dielectric axes, the equation is reduced to $2w_e = \epsilon_{xx}E_x^2 + \epsilon_{yy}E_y^2 + \epsilon_{zz}E_z^2$

The constant energy surfaces in the space (D_x, D_y, D_z) form ellipsoids defined by: $2w_e = \frac{D_x^2}{\epsilon_{xx}} + \frac{D_y^2}{\epsilon_{yy}} + \frac{D_z^2}{\epsilon_{zz}}$

New variable :
$$\vec{r} = \vec{D}\sqrt{2w_e\epsilon_0}$$
 $\vec{x}^2 + \frac{y^2}{n_x^2} + \frac{z^2}{n_z^2} =$

It defines the index of ellipsoid equation that is used to determine, for any direction of propagation, the two refractive indices and the associated direction for \vec{D}

Linear Wave Eq. in an anisotropic material

The index of ellipsoid :

$$\vec{r} = \vec{D}\sqrt{2w_e\epsilon_0}$$

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1$$

It defines the index of ellipsoid equation that is used to determine, for any direction of propagation, the two refractive indices and the associated direction for \vec{D}



The two axes of the ellipse formed by the intersection between the plane perpendicular to \vec{s} , the direction of propagation, and the ellipsoid, are equal to $2n_a$ and $2n_b$. Their related field \vec{D}_a and \vec{D}_b are parallel to these two axes.

↑z = optical

S

no

n_e

n,

S

axis

θ

n_o

Ď,

θ

Ζ

n_e

D_θ

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Linear Wave Eq. in an anisotropic material

Case of an uniaxial crystal

It exhibits two identical principal refractive indices : $n_x = n_y = n_o$ and $n_z = n_e \neq n_o$

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1$$

Ordinary wave with **n**_o – polarization state

Extraordinary wave with no polarization state

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$$\boldsymbol{e}_o = \begin{vmatrix} \sin \phi \\ -\cos \phi \\ 0 \end{vmatrix}$$

 $-\cos\theta\cos\phi$ e_{ℓ}

$$\left(\frac{1}{n_{\theta}}\right)^{2} = \left(\frac{\cos\theta}{n_{o}}\right)^{2} + \left(\frac{\sin\theta}{n_{e}}\right)^{2}$$

$$e_{\theta} = \begin{vmatrix} -\cos\theta\sin\phi\\ \sin\theta \end{vmatrix}$$

$$^{2} + \left(\frac{\sin\theta}{n_{e}}\right)^{2}$$
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Stationary Nonlinear Wave Equation

Nonlinear wave equation at steady state

Nonlinear dielectric material

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P}$$

$$\vec{P} = \vec{P}_L + (\vec{P}_{NL})$$
Nonlinear polarization
$$\vec{P}_L(\omega) = \varepsilon_0 \underline{\chi}^{(1)}(\omega) \vec{E}(\omega)$$

Wave equation in frequency domain including a nonlinear polarization:

$$\nabla \times \nabla \times E(r,\omega) = \frac{\omega^2}{c^2} \underline{\epsilon}(r,\omega) E(r,\omega) + \omega^2 \mu_0 P^{(NL)}(r,\omega)$$
Steady-state solution of the form: $E(r,\omega) = A(r)e^{i\boldsymbol{k}\cdot\boldsymbol{r}}e$

$$-\left[k \times (k \times e) + \frac{\omega^2}{c^2} \underline{\epsilon}(\omega)e\right] A(r)$$

$$+i \left[\nabla A \times (k \times e) + k \times (\nabla A \times e)\right] + \nabla \times (\nabla A \times e) = -\omega^2 \mu_0 P_{NL}(z,\omega)e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}$$
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Nonlinear Wave Equation

$$-\left[\boldsymbol{k}\times(\boldsymbol{k}\times\boldsymbol{e})+\frac{\omega^{2}}{c^{2}}\underline{\boldsymbol{\epsilon}}(\omega)\boldsymbol{e}\right]\boldsymbol{A}(\boldsymbol{r})$$
$$+\imath\left[\nabla\boldsymbol{A}\times(\boldsymbol{k}\times\boldsymbol{e})+\boldsymbol{k}\times(\nabla\boldsymbol{A}\times\boldsymbol{e})\right]+\nabla\times(\nabla\boldsymbol{A}\times\boldsymbol{e}) = -\omega^{2}\mu_{0}\boldsymbol{P}_{NL}(\boldsymbol{z},\omega)\boldsymbol{e}^{-\imath\boldsymbol{k}\cdot\boldsymbol{r}}$$

• Simplification :
$$\vec{E} = \vec{E}_o + \vec{E}_{\theta} = \vec{E}_{ordinary} + \vec{E}_{extraordinary}$$

Eigen polarization states of Fresnel Eq. :

$$\boldsymbol{k} \times (\boldsymbol{k} \times \boldsymbol{e}) + \frac{\omega^2}{c^2} \underline{\epsilon}(\omega) \boldsymbol{e} = 0$$

• Slowly-varying amplitude approximation :

$$\left| k \frac{\partial A}{\partial x_i} \right| \gg \left| \frac{\partial^2 A}{\partial x_i \partial y_i} \right|$$

Slow variation of the field amplitude on a typical length of the order of λ



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Nonlinear Wave Equation

$$2i\nabla A \cdot [(k \times e) \times e] = \omega^2 \mu_0 e \cdot \Pi_{NL}(z, \omega) e^{i\Delta k \cdot r}$$

where $P_{NL}(z, \omega) = \Pi_{NL}(z, \omega) e^{ik_p(\omega) \cdot r}$
• Now, vector $k \times (k \times e)$ is
parallel to Poynting vector
Wave equation in the coordinate
system $(\bar{D}, \bar{H}, \bar{k})$
 $\int -\tan \alpha \frac{\partial A}{\partial x} + \frac{\partial A}{\partial z} = \frac{i\omega^2 \mu_0}{2k \cos^2 \alpha} e \cdot \Pi_{NL}(z, \omega) e^{i\Delta k \cdot r}$
Wave vector
 α : walk-off angle

Nonlinear Wave Equation

• Neglecting the walk-off angle, form for wave equation :

$$\begin{array}{c|c} & & & \\$$



 $\Delta \vec{k} =$

the NL polarization and of the electric field N. Dubreuil - NONLINEAR OPTICS • Nonlinear propagation of waves in an <u>anisotropic</u> material can be treated as the propagation in an <u>isotropic</u> material by priorly decompose the incoming field other the eigen polarization modes, the ordinary and the extraordinary fields.

- Energy transfer condition :
 - phase matching condition $\Delta k=0$
 - Non-zero projection between the electric field and the NL polarization $\vec{e} \cdot \vec{P}_{NL} \neq 0$



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Phase-matching condition in birefringent materials

Example : Second Harmonic Generation

The fields at ω and 2ω are decomposed into the sum of ordinary and extraordinary vibration modes :

$$E(\omega) = E_{o}(\omega) + E_{\theta}(\omega)$$

$$E(2\omega) = E_{o}(2\omega) + E_{\theta}(2\omega)$$

$$E_{o}(\omega) = A_{o}(\omega)e_{o}\exp i(k_{o}(\omega).s)$$

$$E_{o}(2\omega) = A_{o}(2\omega)e_{o}\exp i(k_{o}(2\omega).s)$$

$$E_{o}(2\omega) = A_{o}(2\omega)e_{o}\exp i(k_{o}(2\omega).s)$$

$$E_{\theta}(2\omega) = A_{\theta}(2\omega)e_{\theta}\exp i(k_{\theta}(2\omega).s)$$

Nonlinear wave equations :



Phase-matching condition in birefringent materials

Example : Second Harmonic Generation

$$\begin{aligned} \mathcal{P}_{NL}(2\omega) &= \epsilon_0 \underline{\chi}^{(2)}(\omega, \omega) \boldsymbol{E}(\omega) \boldsymbol{E}(\omega), \\ &= \epsilon_0 \underline{\chi}^{(2)}(\omega, \omega) \left[\boldsymbol{E}_o(\omega) \boldsymbol{E}_o(\omega) + \boldsymbol{E}_\theta(\omega) \boldsymbol{E}_\theta(\omega) + \boldsymbol{E}_o(\omega) \boldsymbol{E}_\theta(\omega) + \boldsymbol{E}_\theta(\omega) \boldsymbol{E}_\theta(\omega) \right] \end{aligned}$$

Type I:

$$k_{p}(2\omega) = k_{\theta}(\omega) + k_{\theta}(\omega) = k_{o}(2\omega) \Rightarrow n_{\theta}(\omega) = n_{o}(2\omega)$$

$$k_{p}(2\omega) = k_{o}(\omega) + k_{o}(\omega) = k_{\theta}(2\omega) \Rightarrow n_{o}(\omega) = n_{\theta}(2\omega)$$
Type II:

$$k_{p}(2\omega) = k_{\theta}(\omega) + k_{o}(\omega) = k_{o}(2\omega) \Rightarrow \frac{n_{\theta}(\omega) + n_{o}(\omega)}{2} = n_{o}(2\omega)$$

$$k_{p}(2\omega) = k_{\theta}(\omega) + k_{o}(\omega) = k_{\theta}(2\omega) \Rightarrow \frac{n_{\theta}(\omega) + n_{o}(\omega)}{2} = n_{\theta}(2\omega)$$
NSULUE

Phase-matching condition in birefringent materials

CONCLUSION

Use of birefringence properties of crystals to satisfy the phase matching condition.

- Phase matching condition can be fulfilled by playing with the birefringent properties of the materials
- This condition to be satisfied sets the polarization states of the interacted fields.
- Note that in some cases, it might be impossible to find a proper phase matching configuration



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Phase-matching condition in birefringent materials

CONCLUSION

Case of SHG in uniaxial crystals

• In a negative (positive) uniaxial crystal : the 2ω wave is necessarily polarized along the extraordinary (ordinary) direction

