FI255 - Tópicos de Óptica e Fotônica II

Óptica Não-Linear

6ª. aula

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UNICAMP - 13 de abril de 2018

Breve revisão da 5ª. Aula

Óptica Não Linear

REVISÃO

Interação luz-matéria sob condições que violam o princípio de superposição linear

Polarização óptica ≡ momento de dipolo por unidade de volume

$$\vec{P} = \epsilon_0 \overleftrightarrow{\chi^{(1)}} \otimes \vec{E} + \epsilon_0 \overleftrightarrow{\chi^{(2)}} \otimes \vec{E}\vec{E} + \epsilon_0 \overleftrightarrow{\chi^{(3)}} \otimes \vec{E}\vec{E}\vec{E} + \ldots$$

$\overleftrightarrow{\chi^{(n)}} \equiv 0$, n = par (meios centro-simétricos)

$\vec{P}$ induz mudança na velocidade da luz no meio e novos comprimentos de onda podem ser gerados

Significado de cada parcela

Vetor polarização

$$\overrightarrow{P^{(1)}} = \epsilon_0 \overleftrightarrow{\chi^{(1)}} \otimes \overrightarrow{E}$$

$$\overrightarrow{P^{(2)}} = \epsilon_0 \overleftrightarrow{\chi^{(2)}} \otimes \vec{E}\vec{E}$$

$$\overrightarrow{P^{(3)}} = \epsilon_0 \overleftrightarrow{\chi^{(3)}} \otimes \vec{E}\vec{E}\vec{E}$$

Componentes da polarização

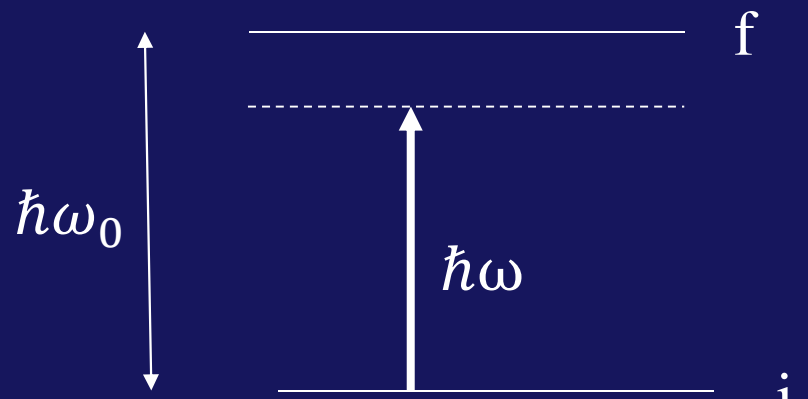
$$P_i^{(1)} = \epsilon_0 \chi_{ij}^{(1)} E_j$$

$$P_i^{(2)} = \epsilon_0 \chi_{ijk}^{(2)} E_j E_k$$

$$P_i^{(3)} = \epsilon_0 \chi_{ijkl}^{(3)} E_j E_k E_l$$

analogamente para os demais têrmos ...

$$\chi^{(1)}(\omega,\omega) = \frac{Ne^2}{m\epsilon_0[(\omega_0^2 - \omega^2) + i\gamma\omega]}$$



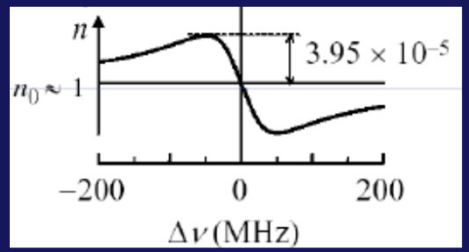
$n_0 \propto Re\ \chi^{(1)}(\omega,\omega)$

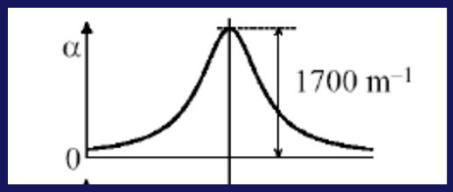
Índice de refração linear

$\alpha_0 \propto Im\ \chi^{(1)}(\omega,\omega)$

Coeficiente de absorção linear

Exemplo: sódio gasoso (2ª. Aula)

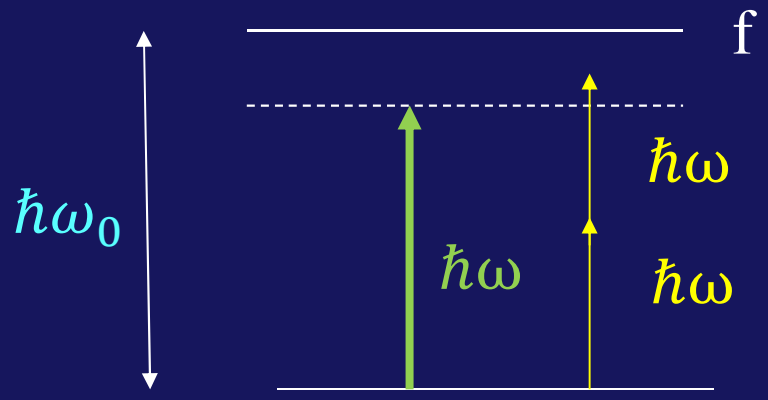




$$P(2\omega,t) = -Nex(2\omega,t) = \epsilon_0 \chi^{(2)} \varepsilon(t)^2$$

$$\chi^{(2)}(2\omega,\omega,\omega) = \frac{Ne^3K}{\epsilon_0 m^2[(\omega_0^2-\omega^2)+i\gamma\omega]^2\,(\omega_0^2-4\omega^2+i2\omega\gamma)}$$

ressonâncias



$\chi^{(2)}$

Second order polarization

Non centro-symmetric materials

REVISÃO

$$P^{(2)} = \varepsilon_0 \chi^{(2)} (E_1 \cos\omega_1 t + E_2 \cos\omega_2 t)^2 =$$

$$P_0^{(2)} + P_{2\omega_1}^{(2)} + P_{2\omega_2}^{(2)} + P_{\omega_1+\omega_2}^{(2)} + P_{\omega_1-\omega_2}^{(2)}$$

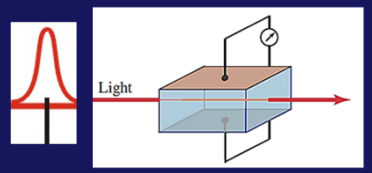
Optical retification

SHG

ω

SFG

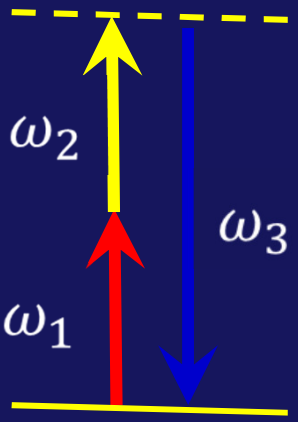
DFG

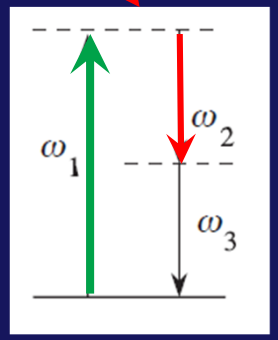


Light pulse

Voltage

time





Bulk crystals (1961)

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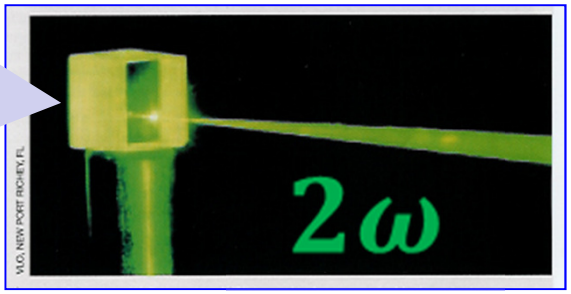
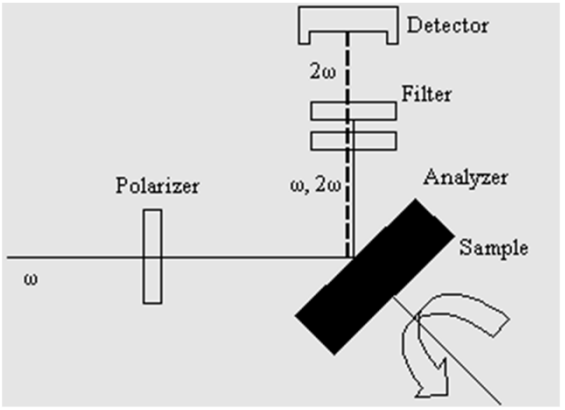
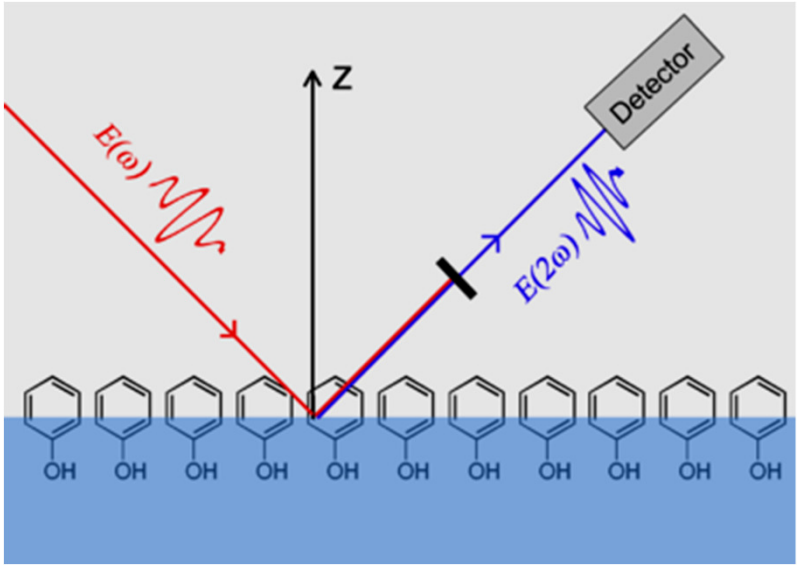
Crystal surface (1962)

ω



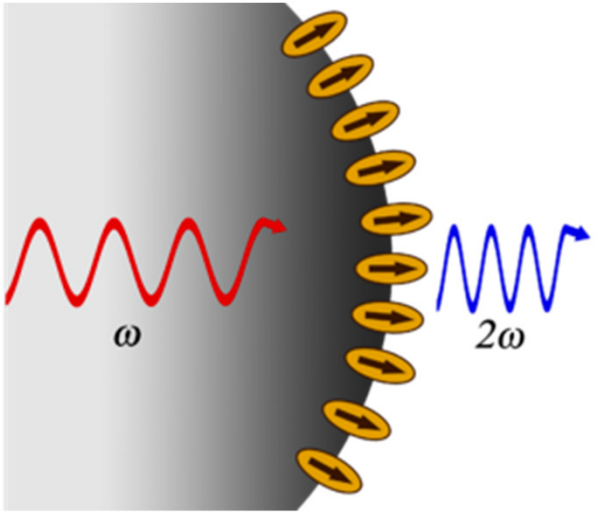


Phenol group at the air-water interface



Many analogous experiments starting in the 70's

Molecules on a curved surface

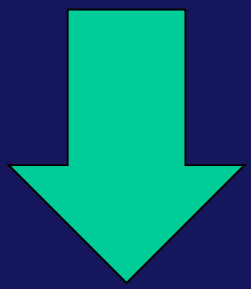


Many contemporary experiments with nanoparticles

Equação de onda para um meio não linear

$$\nabla^2 \vec{E}(\vec{r},t) - \mu\epsilon \frac{\partial^2 \vec{E}(\vec{r},t)}{\partial t^2} = \mu \frac{\partial^2 \vec{P}_{\mathrm{NL}}(\vec{r},t)}{\partial t^2}$$

$\mu = \mu_0$



$\epsilon = \epsilon(\omega)$

$$\nabla^2 \vec{E}(\vec{r},t,\omega_i) - \mu\epsilon(\omega_i) \frac{\partial^2 \vec{E}(\vec{r},t,\omega_i)}{\partial t^2} = \mu \frac{\partial^2 \vec{P}_{\mathrm{NL}}(\vec{r},t,\omega_i)}{\partial t^2}$$

Processos não lineares de segunda ordem

Dois campos incidentes com frequências ω_1 e ω_2

$$P_k^{(2)} = \epsilon_0 \, \chi_{kij}^{(2)} \, E_i \, E_j$$

REVISÃO

$$E_i(z,t,\omega_1) = \frac{1}{2}\left[E_i(z,\omega_1)\,\mathrm{e}^{\mathrm{i}(k_1 z-\omega_1 t)} + \mathrm{cc}\right],$$

$$E_j(z,t,\omega_2) = \frac{1}{2}\left[E_j(z,\omega_2)\,\mathrm{e}^{\mathrm{i}(k_2 z-\omega_2 t)} + \mathrm{cc}\right],$$

$$E_k(z,t,\omega) = \frac{1}{2}\left[E_k(z,\omega)\,\mathrm{e}^{\mathrm{i}(kz-\omega t)} + \mathrm{cc}\right],$$

Campo gerado devido a $\chi_{kij}^{(2)}\,(\omega,\omega_1,\omega_2)$

$$\frac{\mathrm{d}^2\left[E_i(z,\omega_1)\,\mathrm{e}^{\mathrm{i}k_1 z}\right]}{\mathrm{d}z^2}+\mu\epsilon_1\omega_1^2E_i(z,\omega_1)\,\mathrm{e}^{\mathrm{i}k_1 z}=-\mu\,\omega_1^2P_i^{(2)}(z,\omega_1),$$

$$\frac{\mathrm{d}^2\left[E_j(z,\omega_2)\,\mathrm{e}^{\mathrm{i}k_2 z}\right]}{\mathrm{d}z^2}+\mu\epsilon_2\omega_2^2E_j(z,\omega_2)\,\mathrm{e}^{\mathrm{i}k_2 z}=-\mu\,\omega_2^2P_j^{(2)}(z,\omega_2),$$

$$\frac{\mathrm{d}^2\left[E_k(z,\omega)\,\mathrm{e}^{\mathrm{i}kz}\right]}{\mathrm{d}z^2}+\mu\epsilon\omega^2E_k(z,\omega)\,\mathrm{e}^{\mathrm{i}kz}=-\mu\,\omega^2P_k^{(2)}(z,\omega).$$

Sum frequency generation

$\omega = \omega_1 + \omega_2$ $\qquad$ $\vec{k} = \overrightarrow{k_1} + \overrightarrow{k_2}$

$$P_i^{(2)}(z,\omega_1)=\frac{\epsilon_0}{2}\sum_{jk}\chi_{ijk}^{(2)}(\omega_1,-\omega_2,\omega)E_j^*(z,\omega_2)\,E_k(z,\omega)\,\mathrm{e}^{\mathrm{i}(k-k_2)z},$$

$$P_j^{(2)}(z,\omega_2)=\frac{\epsilon_0}{2}\sum_{ki}\chi_{jki}^{(2)}(\omega_2,\omega,-\omega_1)E_k(z,\omega)\,E_i^*(z,\omega_1)\,\mathrm{e}^{\mathrm{i}(k-k_1)z},$$

$$P_k^{(2)}(z,\omega)=\frac{\epsilon_0}{2}\sum_{ij}\chi_{kij}^{(2)}(\omega,\omega_1,\omega_2)E_i(z,\omega_1)\,E_j(z,\omega_2)\,\mathrm{e}^{\mathrm{i}(k_1+k_2)z},$$

$$\Delta k = k - k_1 - k_2$$

$$\frac{\mathrm{d}E_i(z,\omega_1)}{\mathrm{d}z} = \frac{\mathrm{i}\omega_1}{4cn_1}\sum_{jk}\chi^{(2)}_{ijk}(\omega_1,-\omega_2,\omega)E^*_j(z,\omega_2)E_k(z,\omega)\mathrm{e}^{\mathrm{i}\Delta kz},$$

$$\frac{\mathrm{d}E_j(z,\omega_2)}{\mathrm{d}z} = \frac{\mathrm{i}\omega_2}{4cn_2}\sum_{ki}\chi^{(2)}_{jki}(\omega_2,\omega,-\omega_1)E_k(z,\omega)E^*_i(z,\omega_1)\mathrm{e}^{\mathrm{i}\Delta kz},$$

$$\frac{\mathrm{d}E_k(z,\omega)}{\mathrm{d}z} = \frac{\mathrm{i}\omega}{4cn}\sum_{ij}\chi^{(2)}_{kij}(\omega,\omega_1,\omega_2)E_i(z,\omega_1)E_j(z,\omega_2)\mathrm{e}^{-\mathrm{i}\Delta kz},$$

$$\frac{\mathrm{d}E_i(\omega_1)}{\mathrm{d}z} = \mathrm{i}\alpha\frac{\omega_1}{n_1}E^*_j(\omega_2)E_k(\omega)\mathrm{e}^{\mathrm{i}\Delta kz},$$

$$\frac{\mathrm{d}E_j(\omega_2)}{\mathrm{d}z} = \mathrm{i}\alpha\frac{\omega_2}{n_2}E_k(\omega)E^*_i(\omega_1)\mathrm{e}^{\mathrm{i}\Delta kz},$$

$$\frac{\mathrm{d}E_k(\omega)}{\mathrm{d}z} = \mathrm{i}\alpha\frac{\omega}{n}E_i(\omega_1)E_j(\omega_2)\mathrm{e}^{-\mathrm{i}\Delta kz},$$

Transparent crystal

$$\alpha = \frac{1}{4c}\chi^{(2)}_{\mathrm{eff}} = \frac{1}{2c}d^{(2)}_{\mathrm{eff}}$$

Intensidades dos feixes incidentes e feixe gerado

$$I(\omega_1) = \frac{1}{2}\epsilon_0 n_1 c E_i(\omega_1) E_i^*(\omega_1)$$

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$$\frac{\mathrm{d}I(\omega_1)}{\mathrm{d}z} = \frac{1}{2}\epsilon_0 n_1 c \left[E_i(\omega_1)\frac{\mathrm{d}E_i^*(\omega_1)}{\mathrm{d}z} + E_i^*(\omega_1)\frac{\mathrm{d}E_i(\omega_1)}{\mathrm{d}z} \right]$$

Parte imaginária

$$\begin{aligned}\frac{\mathrm{d}I(\omega_1)}{\mathrm{d}z} &= \frac{1}{2}\epsilon_0 c\alpha\omega_1 \left[-\mathrm{i}E_i(\omega_1)E_j(\omega_2)E_k^*(\omega)\,\mathrm{e}^{-\mathrm{i}\Delta kz} \right. \\ &\quad \left. +\mathrm{i}E_i^*(\omega_1)E_j^*(\omega_2)E_k(\omega)\,\mathrm{e}^{\mathrm{i}\Delta kz} \right] \\ &= \epsilon_0 c\alpha\omega_1 I_{\mathrm{im}} \left[\mathrm{i}E_i^*(\omega_1)E_j^*(\omega_2)E_k(\omega)\,\mathrm{e}^{\mathrm{i}\Delta kz} \right].\end{aligned}$$

$$\begin{aligned}\frac{\mathrm{d}I(\omega_2)}{\mathrm{d}z} &= \epsilon_0 c\alpha\omega_2 I_{\mathrm{im}} \left[\mathrm{i}E_i^*(\omega_1)E_j^*(\omega_2)E_k(\omega)\,\mathrm{e}^{\mathrm{i}\Delta kz} \right], \\ \frac{\mathrm{d}I(\omega=\omega_1+\omega_2)}{\mathrm{d}z} &= -\epsilon_0 c\alpha\omega I_{\mathrm{im}} \left[\mathrm{i}E_i^*(\omega_1)E_j^*(\omega_2)E_k(\omega)\,\mathrm{e}^{\mathrm{i}\Delta kz} \right]\end{aligned}$$

$$\frac{\mathrm{d}}{\mathrm{d}z}\left[I(\omega_1)+I(\omega_2)+I(\omega=\omega_1+\omega_2)\right]=0$$

$$\frac{1}{\omega}\frac{\mathrm{d}I(\omega)}{\mathrm{d}z}=-\frac{1}{\omega_1}\frac{\mathrm{d}I(\omega_1)}{\mathrm{d}z}=-\frac{1}{\omega_2}\frac{\mathrm{d}I(\omega_2)}{\mathrm{d}z}$$

Campo gerado

$$\frac{\mathrm{d}E_k(\omega)}{\mathrm{d}z}=\mathrm{i}\alpha\frac{\omega}{n}E_i(\omega_1)E_j(\omega_2)\mathrm{e}^{-\mathrm{i}\Delta kz}$$

Na face de saida da amostra

$$E_k(\omega_1+\omega_2,z=L)=\mathrm{i}(\omega_1+\omega_2)\frac{\alpha}{n}E_i(\omega_1)E_j(\omega_2)\int_0^L\mathrm{e}^{-\mathrm{i}\Delta kz}\,\mathrm{d}z$$

$$=\mathrm{i}(\omega_1+\omega_2)\frac{\alpha}{n}L\,\mathrm{e}^{-\mathrm{i}\frac{\Delta k}{2}L}E_i(\omega_1)E_j(\omega_2)\mathrm{sinc}\left(\frac{\Delta kL}{2}\right),$$

Intensidade gerada

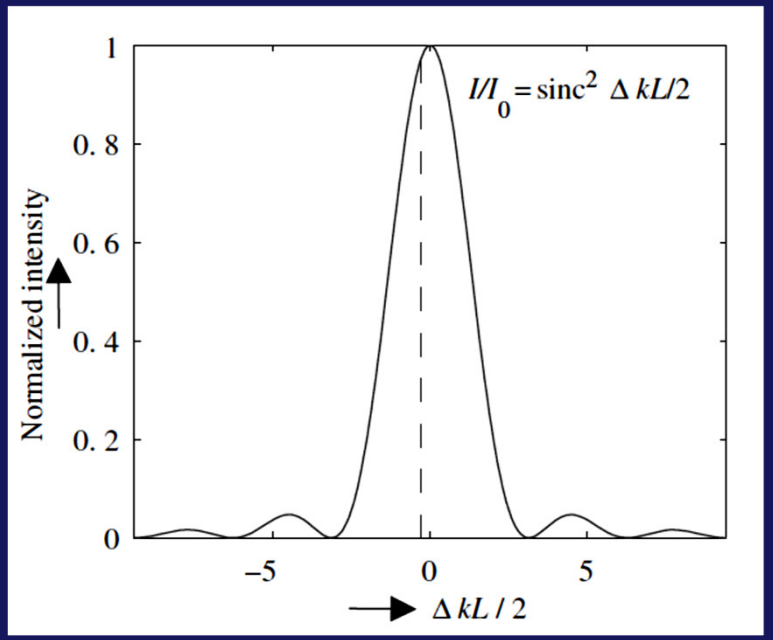
$$I(\omega_1+\omega_2, z=L) = I_0 \,\mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right)$$

$$I_0 = \frac{(\omega_1+\omega_2)^2 L^2 I(\omega_1) I(\omega_2)}{2 n_1 n_2 n \epsilon_0 c^3} (d_{\mathrm{eff}}^{(2)})^2$$

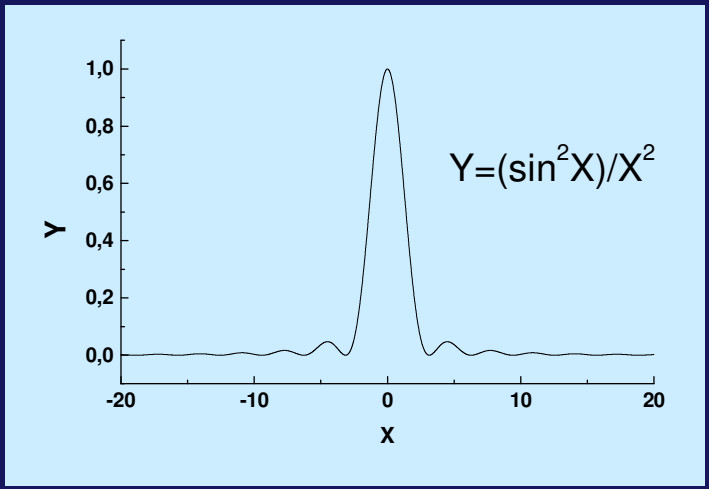
$$I(\omega_1) = \frac{1}{2}\epsilon_0 n_1 c \left|E_i(\omega_1)\right|^2$$

$$I(\omega_2) = \frac{1}{2}\epsilon_0 n_2 c \left|E_j(\omega_2)\right|^2$$

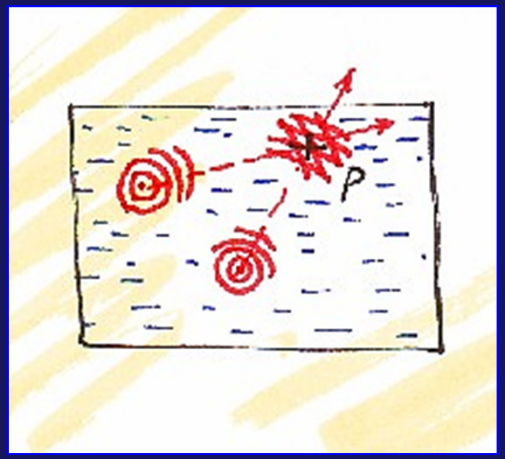
$$\mathrm{sinc}\left(\frac{\Delta kL}{2}\right) = \frac{\sin(\Delta kL/2)}{\Delta kL/2}$$



Por que?



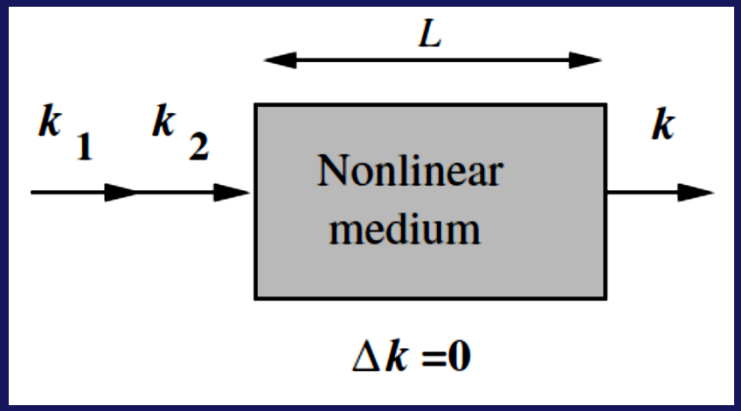
A radiação em um ponto particular é calculada integrando sobre toda a amostra. Contudo as contribuições das várias partes do cristal, em geral, não estão em fase.



Os dipolos induzidos se somam construtivamente dentro do comprimento de coerência

$$l_{coh} = \frac{2\pi}{\Delta k}$$

Phase – Matching colinear



$$\omega = \omega_1 + \omega_2$$

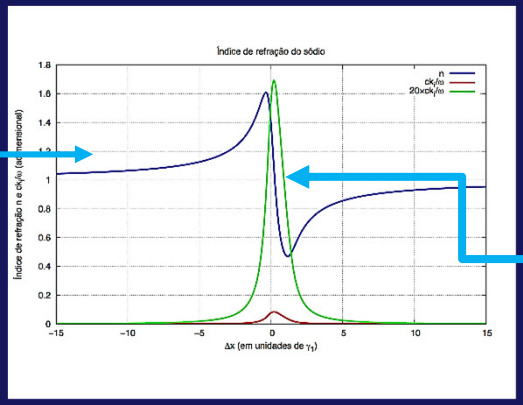
$$\vec{k} = \overrightarrow{k_1} + \overrightarrow{k_2}$$

$$(\omega_1 + \omega_2)\, n(\omega_1 + \omega_2) = \omega_1\, n(\omega_1) + \omega_2\, n(\omega_2)$$

$$\omega_1[n(\omega_1 + \omega_2) - n(\omega_1)] + \omega_2[n(\omega_1 + \omega_2) - n(\omega_2)] = 0$$

Para um meio isotrópico ou materiais cúbicos com dispersão normal

Dispersão normal



Dispersão anômala

$$\omega = \omega_1 + \omega_2$$

$$n(\omega) > \{n(\omega_1), n(\omega_2)\}$$

$$\omega_1[n(\omega_1 + \omega_2) - n(\omega_1)] + \omega_2[n(\omega_1 + \omega_2) - n(\omega_2)] = 0$$

portanto a relação não pode ser satisfeita

Optical anisotropy

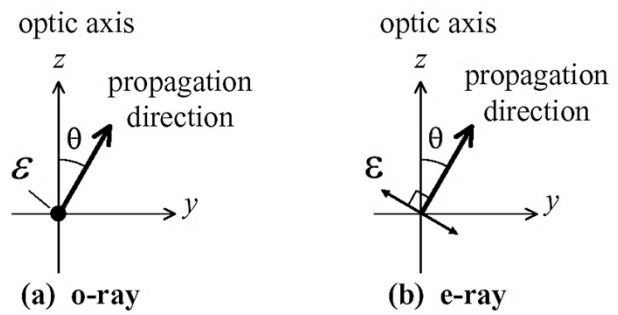


Figure 2.12
Birefringence caused by difference of dielectric constants (and hence refractive index) along the different crystal axes.

$$\begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \varepsilon_{11} & 0 & 0 \\ 0 & \varepsilon_{22} & 0 \\ 0 & 0 & \varepsilon_{33} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

$$\frac{1}{n_e^2(\theta)} = \frac{cos^2\theta}{n_0^2} + \frac{sen^2\theta}{n_e^2}$$

Cristal uniaxial

negativo $n_e(\omega_i) < n_o(\omega_i)$

positivo $n_e(\omega_i) > n_o(\omega_i)$

$$\omega_1[n(\omega_1 + \omega_2) - n(\omega_1)] + \omega_2[n(\omega_1 + \omega_2) - n(\omega_2)] = 0$$

	$n_e > n_o$	$n_e < n_o$
Type I	$e, e \rightarrow o$	$o, o \rightarrow e$
Type II	$o, e \rightarrow o$	$e, o \rightarrow e$

REVISÃO

Applications

Interface structure

Adsorption measurements

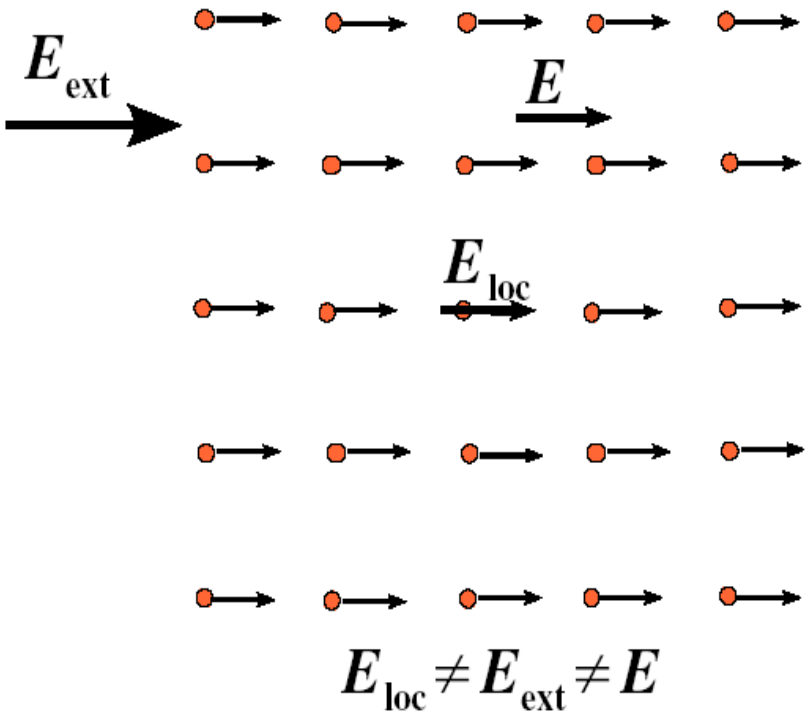
Molecular orientation

Confocal Microscopy:
monolayers, single molecule, single nanoparticles

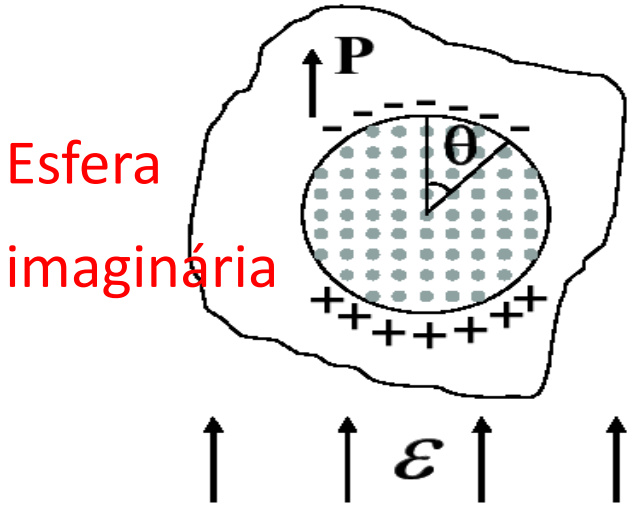
Many applications in biology and medical physics

Nos cálculos que mostrei devemos considerar o efeito do campo local

Considere um meio homogêneo denso submetido a um campo óptico externo



constituintes ativos sentem o campo local



$$E_{loc} = E + \frac{P}{3\epsilon_0}$$

Polarizabilidade

$$p = \epsilon_0 \alpha E_{loc} \quad \Rightarrow \quad P = Np$$

$$P = N\epsilon_0\, \alpha \left(E + \frac{P}{3\epsilon_0} \right)$$

Susceptibilidade linear

$$P = \epsilon_0 \chi^{(1)} E$$

$$\chi^{(1)} = \frac{N\alpha}{1 - \frac{N\alpha}{3}}$$

$$\epsilon^{(1)}(\omega) = 1 + \chi^{(1)}(\omega)$$

Relação de Clausius-Mossoti

$$\frac{\epsilon^{(1)} - 1}{\epsilon^{(1)} + 2} = \frac{N\alpha}{3}$$

$$\chi^{(1)} = \frac{\epsilon^{(1)} + 2}{3} N\alpha(\omega) = L(\omega)\, N\alpha(\omega)$$

Fator de campo local: $L(\omega)$

Vácuo: $\epsilon^{(1)}=1$ → $\chi^{(1)}(\omega) = N\,\alpha(\omega)$

Engrandecimento da susceptibilidade linear

Solução detalhada do oscilador anarmônico

$$\frac{d^2x}{dt^2} + \gamma\frac{dx}{dt} + \omega_0^2 x + ax^2 = \frac{q}{m}E'(t)$$

$$ax^2 \ll \omega_0^2 x$$

E'(t) é o campo local

$$E'(t) = \frac{1}{2}\left[E'(\omega_1)\, e^{-i\omega_1 t} + E'(\omega_2)\, e^{-i\omega_2 t} + cc\right]$$

$$\omega_1, \omega_2 \ll \omega_0$$

$$E'^*(\omega_1) = E'(-\omega_1) \qquad E'^*(\omega_2) = E'(-\omega_2)$$

$$x(t) = x^{(1)}(t) + x^{(2)}(t) + x^{(3)}(t) + \cdots$$

$$x = x^{(1)}(t) = \frac{1}{2}\left[x^{(1)}(\omega_1)\, e^{-i\omega_1 t} + x^{(1)}(\omega_2)\, e^{-i\omega_2 t} + cc\right]$$

$$x^{(1)}(t) = \frac{1}{2}\left[\frac{qE'(\omega_1)/m}{\omega_0^2 - \omega_1^2 - i\gamma\omega_1} e^{-i\omega_1 t} + \frac{qE'(\omega_2)/m}{\omega_0^2 - \omega_2^2 - i\gamma\omega_2} e^{-i\omega_2 t} + cc\right]$$

Polarização e polarizabilidade

$$P_{\mathrm{L}}(t) = \frac{1}{2}\left[P_{\mathrm{L}}(\omega_1)\mathrm{e}^{-\mathrm{i}\omega_1 t} + P_{\mathrm{L}}(\omega_2)\mathrm{e}^{-\mathrm{i}\omega_2 t} + \mathrm{cc}\right]$$

$$P_{\mathrm{L}}(\omega_1) = Nqx^{(1)}(\omega_1) = \frac{\left(Nq^2/m\right)E'(\omega_1)}{\omega_0^2 - \omega_1^2 - \mathrm{i}\gamma\omega_1} = N\alpha^{(1)}(\omega_1)E'(\omega_1)$$

$$P_{\mathrm{L}}(\omega_2) = Nqx^{(1)}(\omega_2) = \frac{\left(Nq^2/m\right)E'(\omega_2)}{\omega_0^2 - \omega_2^2 - \mathrm{i}\gamma\omega_2} = N\alpha^{(1)}(\omega_2)E'(\omega_2)$$

$$\alpha^{(1)}(\omega_1) = \frac{\left(q^2/m\right)}{\omega_0^2 - \omega_1^2 - \mathrm{i}\gamma\omega_1}$$

$$\alpha^{(1)}(\omega_2) = \frac{\left(q^2/m\right)}{\omega_0^2 - \omega_2^2 - \mathrm{i}\gamma\omega_2}.$$

Contribuição anarmônica

$$ax^2 = a\left[x^{(1)} + x^{(2)} + x^{(3)} + \cdots\right]^2 = a(x^{(1)})^2 + 2ax^{(1)}x^{(2)} + \cdots$$

Subtituindo a expressão já obtida

$$x^{(1)}(t) = \frac{1}{2}\left[\frac{qE'(\omega_1)/m}{\omega_0^2 - \omega_1^2 - \mathrm{i}\gamma\omega_1}\mathrm{e}^{-\mathrm{i}\omega_1 t} + \frac{qE'(\omega_2)/m}{\omega_0^2 - \omega_2^2 - \mathrm{i}\gamma\omega_2}\mathrm{e}^{-\mathrm{i}\omega_2 t} + \mathrm{cc}\right]$$

$$a_1 = \frac{qE'(\omega_1)/m}{\omega_0^2 - \omega_1^2 - \mathrm{i}\gamma\omega_1}$$

$$a_2 = \frac{qE'(\omega_2)/m}{\omega_0^2 - \omega_2^2 - \mathrm{i}\gamma\omega_2}$$

$$(x^{(1)})^2 = \frac{1}{4}\left[a_1\mathrm{e}^{-\mathrm{i}\omega_1 t} + a_1^*\mathrm{e}^{\mathrm{i}\omega_1 t} + a_2\mathrm{e}^{-\mathrm{i}\omega_2 t} + a_2^*\mathrm{e}^{\mathrm{i}\omega_2 t}\right]^2 = \frac{1}{2}(a_1a_1^* + a_2a_2^*)$$
$$+ \frac{1}{4}\left[a_1^2\mathrm{e}^{-\mathrm{i}2\omega_1 t} + a_2^2\mathrm{e}^{-\mathrm{i}2\omega_2 t} + 2a_1a_2\mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t} + 2a_1a_2^*\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)t} + \mathrm{cc}\right]$$

Solução de 2ª. ordem

$$\frac{\mathrm{d}^2 x^{(2)}}{\mathrm{d}t^2} + \gamma \frac{\mathrm{d}x^{(2)}}{\mathrm{d}t} + \omega_0^2 x^{(2)} = -a(x^{(1)})^2$$

$$x^{(2)}(t) = x^{(2)}(0) + \frac{1}{2}\left[x^{(2)}(2\omega_1)\mathrm{e}^{-\mathrm{i}2\omega_1 t} + x^{(2)}(2\omega_2)\mathrm{e}^{-\mathrm{i}2\omega_2 t}\right.$$
$$\left. + x^{(2)}(\omega_1+\omega_2)\mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t} + x^{(2)}(\omega_1-\omega_2)\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)t} + \mathrm{cc}\right]$$

$$x^{(2)}(0) = \frac{1}{2}\frac{(-a)(q/m\omega_0)^2 E'(\omega_1) E'^{*}(\omega_1)}{(\omega_0^2 - \omega_1^2 - i\gamma\omega_1)(\omega_0^2 - \omega_1^2 + i\gamma\omega_1)} + \frac{1}{2}\frac{(-a)(q/m\omega_0)^2 E'(\omega_2) E'^{*}(\omega_2)}{(\omega_0^2 - \omega_2^2 - i\gamma\omega_2)(\omega_0^2 - \omega_2^2 + i\gamma\omega_2)}$$

$$x^{(2)}(2\omega_i) = \frac{1}{2}\frac{(-a)(q/m)^2 E'(\omega_i) E'(\omega_i)}{(\omega_0^2 - 4\omega_i^2 - i2\gamma\omega_i)(\omega_0^2 - \omega_i^2 - i\gamma\omega_i)^2}$$

$$x^{(2)}(\omega_1 + \omega_2) = \frac{(-a)(q/m)^2 E'(\omega_1) E'(\omega_2)}{[\omega_0^2 - (\omega_1 + \omega_2)^2 - i\gamma(\omega_1 + \omega_2)]} \times \frac{1}{(\omega_0^2 - \omega_1^2 - i\gamma\omega_1)(\omega_0^2 - \omega_2^2 - i\gamma\omega_2)}$$

$$x^{(2)}(\omega_1 - \omega_2) = \frac{(-a)(q/m)^2 E'(\omega_1) E'^{*}(\omega_2)}{\left[\omega_0^2 - (\omega_1 - \omega_2)^2 - i\gamma(\omega_1 - \omega_2)\right]} \times \frac{1}{(\omega_0^2 - \omega_1^2 - i\gamma\omega_1)(\omega_0^2 - \omega_2^2 + i\gamma\omega_2)}$$

$$P_{\mathrm{NL}}^{(2)}(t) = P_{\mathrm{NL}}^{(2)}(0) + \frac{1}{2}\left[P_{\mathrm{NL}}^{(2)}(2\omega_1)\mathrm{e}^{-\mathrm{i}2\omega_1 t} + P_{\mathrm{NL}}^{(2)}(2\omega_2)\mathrm{e}^{-\mathrm{i}2\omega_2 t} + P_{\mathrm{NL}}^{(2)}(\omega_1+\omega_2)\mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t} + P_{\mathrm{NL}}^{(2)}(\omega_1-\omega_2)\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)t} + \mathrm{cc}\right]$$

$$P_{\mathrm{NL}}^{(2)}(0) = Nqx^{(2)}(0) = N\left[\alpha^{(2)}(0,\omega_1,-\omega_1)E'(\omega_1)E'^{*}(\omega_1) + \alpha^{(2)}(0,\omega_2,-\omega_2)E'(\omega_2)E'^{*}(\omega_2)\right],$$

$$P_{\mathrm{NL}}^{(2)}(2\omega_i) = Nqx^{(2)}(2\omega_i) = N\alpha^{(2)}(2\omega_i,\omega_i,\omega_i)E'(\omega_i)E'(\omega_i)$$

$$P_{\mathrm{NL}}^{(2)}(\omega_1+\omega_2)=Nqx^{(2)}(\omega_1+\omega_2)$$
$$=N\alpha^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2)\,E'(\omega_1)\,E'(\omega_2)$$

$$P_{\mathrm{NL}}^{(2)}(\omega_1-\omega_2)=Nqx^{(2)}(\omega_1-\omega_2)$$
$$=N\alpha^{(2)}(\omega_1-\omega_2,\omega_1,\omega_2)\,E'(\omega_1)\,E'^{*}(\omega_2)$$

$$\alpha^{(2)}(0,\omega_1,-\omega_1)=\frac{(-a)\left(q^3/2m^2\omega_0^2\right)}{\left(\omega_0^2-\omega_1^2-\mathrm{i}\gamma\omega_1\right)\left(\omega_0^2-\omega_1^2+\mathrm{i}\gamma\omega_1\right)}$$

$$\alpha^{(2)}(2\omega_i,\omega_i,\omega_i)=\frac{(-a)\,q^3/2m^2}{\left(\omega_0^2-4\omega_i^2-\mathrm{i}2\gamma\omega_i\right)\left(\omega_0^2-\omega_i^2-\mathrm{i}\gamma\omega_i\right)^2}$$

$$P(\omega) = N[\alpha^{(1)}(\omega)E'(\omega) + \alpha^{(2)}(\omega,\omega_1,\omega_2)E'(\omega_1)E'(\omega_2)+\ldots]$$

$$E'(\omega_i) = \frac{E(\omega_i)}{1 - \frac{N\alpha^{(1)}(\omega_i)}{3\epsilon_0}} = \eta(\omega_i)E(\omega_i)$$

$$P(\omega) = \eta(\omega)\, N\, \alpha^{(1)}(\omega)\, E(\omega) +$$
$$\eta(\omega)\eta(\omega_1)\eta(\omega_2)\, N\alpha^{(2)}(\omega,\omega_1,\omega_2)E(\omega_1)E(\omega_2)+\ldots$$

$$P(\omega) = \epsilon_0\left[\chi^{(1)}(\omega)E(\omega) + \chi^{(2)}(\omega,\omega_1,\omega_2)E(\omega_1)E(\omega_2) + \cdots\right]$$

$$\chi^{(1)}(\omega) = \frac{N\alpha^{(1)}(\omega)}{1 - \frac{N\alpha^{(1)}(\omega)}{3\epsilon_0}} = \eta(\omega)N\alpha^{(1)}(\omega)$$

$$\eta(\omega) = \left(1 - \frac{N\alpha^{(1)}(\omega)}{3\epsilon_0}\right)^{-1}$$

$$\chi^{(2)}(\omega,\omega_1,\omega_2) = \eta(\omega)\,\eta(\omega_1)\,\eta(\omega_2)\,N\alpha^{(2)}(\omega,\omega_1,\omega_2)$$

$$\chi^{(n)}(\omega,\omega_1,\omega_2,\ldots.\omega_n) = \eta(\omega)\eta(\omega_1)\eta(\omega_2)\ldots\eta(\omega_n)\,N\alpha^{(n)}(\omega,\omega_1,\omega_2,\ldots\omega_n)$$

$\alpha^{(n)}(\omega,\omega_1,\omega_2,\ldots\omega_n)$ - Hiper polarizabilidades

$$\chi^{(2)}(2\omega,\omega,\omega) = \left[\frac{am\epsilon_0^2}{2N^2q^3}\right]\chi^{(1)}(2\omega)\ \left(\chi^{(1)}(\omega)\right)^2$$

Expressões análogas para a SOMA e DIFERENÇA de frequências

Meio transparente – longe das ressonâncias

$$\omega_0 \gg 2\omega$$

$$\chi^{(1)}(\omega) \approx \chi^{(1)}(2\omega) \approx \frac{Nq^2}{\epsilon_0 m\omega_0^2}$$

Para estimar o parâmetro anarmônico considere a força restauradora

$$F(x) = -\frac{dU}{dx} = -m_0\omega_0^2 x - m_0 a x^2$$

Considere que

$$m_0\omega_0^2 x = m_0 a x^2$$

quando x tiver aproximadamente o tamanho do átomo d

$$m_0\omega_0^2 d = a m_0 d^2$$

$$a = \frac{\omega_0^2}{d}$$

Portanto d é aproximadamente igual à constante de rede do sólido.

$$N = \frac{1}{d^3}$$

Matéria condensada: $N \approx 10^{22}\ cm^3$

$$\chi^{(1)}(\omega) = \frac{Nq^2}{\epsilon_0 m \omega_0^2}$$

$$\chi^{(2)}(2\omega, \omega, \omega) = \frac{q^3}{\epsilon_0 m^2 \omega_0^4 d^4}$$

$m = 9.1 \times 10^{-31}\ kg$

$\omega_0 = 10^{16}\ rad/s$

$d = 3 \times 10^{-10}\ m$

$\epsilon_0 = 8.85 \times 10^{-12}\ \frac{C^2}{N\,m^2}$

$q = 1.6 \times 10^{-19}\ C$

$$\chi^{(1)}(\omega) \sim 3.5$$

$$\chi^{(2)}(2\omega, \omega, \omega) \sim 6.9 \times 10^{-12}\ m/V$$

$$\chi^{(2)}(2\omega,\omega,\omega) = \frac{am\epsilon_0^2}{2N^2q^3}\,\chi^{(1)}(2\omega)\,\left(\chi^{(1)}(\omega)\right)^2$$

Miller's rule

$$\frac{\chi^{(2)}(2\omega,\omega,\omega)}{\chi^{(1)}(2\omega)\,\left(\chi^{(1)}(\omega)\right)^2} = constante$$

R. C. Miller, Appl. Phys. Lett. 5, 17 (1964)

Verificação para vários sistemas transparentes (1970-2010)

Violação em metamateriais (2015-2016)

Violação da regra de Miller em metamateriais

O'Brien et al., *Predicting nonlinear properties of metamaterials from linear response*. Nature Mater. **2015**, 14, 379-383.

Butet et al., *Evaluation of the nonlinear response of plasmonic metasurfaces: Miller's rule, nonlinear effective susceptibility method, and full wave computation.* J. Opt. Soc. Am. B **2016**, 33, A8-A15.

Metamateriais: apresentam interações coletivas que não são consideradas no modelo do oscilador anarmônico

Amplitude relativa entre as polarizações induzidas

$$\frac{P^{(2)}}{P^{(1)}} = \frac{P(2\omega)}{P(\omega)} = \frac{Nqx^{(2)}(2\omega)}{Nqx^{(1)}(\omega)}$$

$$= \frac{(a\,q/2\,m)E'(\omega)}{(\omega_0^2 - 4\omega^2 - i2\gamma\omega)(\omega_0^2 - \omega^2 - i\gamma\omega)}$$

Longe das ressonâncias

$$\frac{P(2\omega)}{P(\omega)} = \left|\frac{aq}{2m\omega_0^4}\right| E'(\omega)$$

$$\left|\frac{aq}{2m\omega_0^4}\right| E_{at} \approx 1$$

quando: $m_0\omega_0^2 d = m_0 a d^2$

$$\frac{P(2\omega)}{P(\omega)} \sim \frac{E'(\omega)}{E_{at}}$$

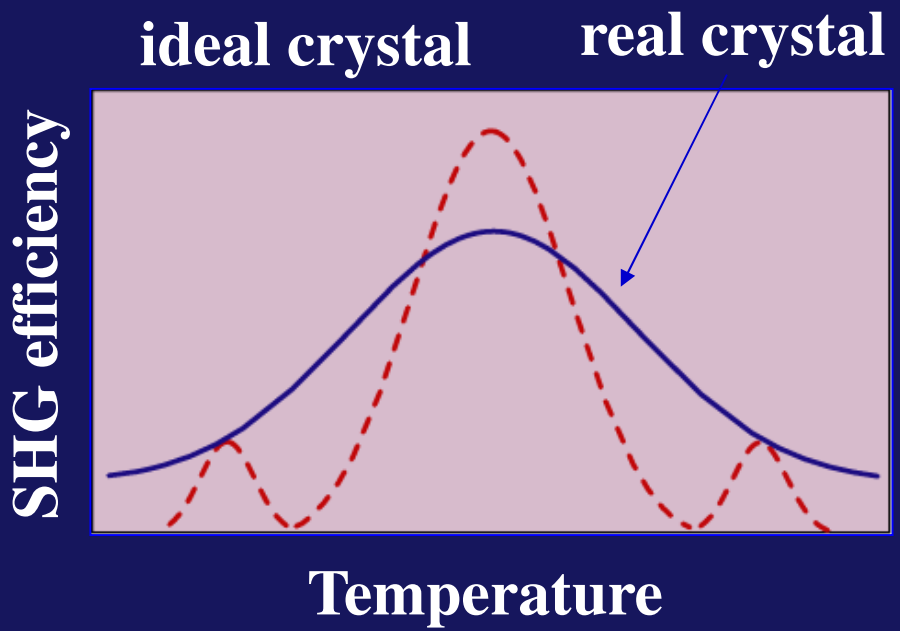
Em geral:

$$\frac{P^{(n+1)}}{P^{(n)}} \sim \frac{E'(\omega)}{E_{at}}$$

Temperature phase-matching

- In general n_e is more temperature dependent than n_o
- Real crystals: broadening of central peak because of slightly varying chemical compositions at different sites of the crystals

$$\Delta k = 2\omega\,(n_{2\omega} - n_{\omega})/c$$

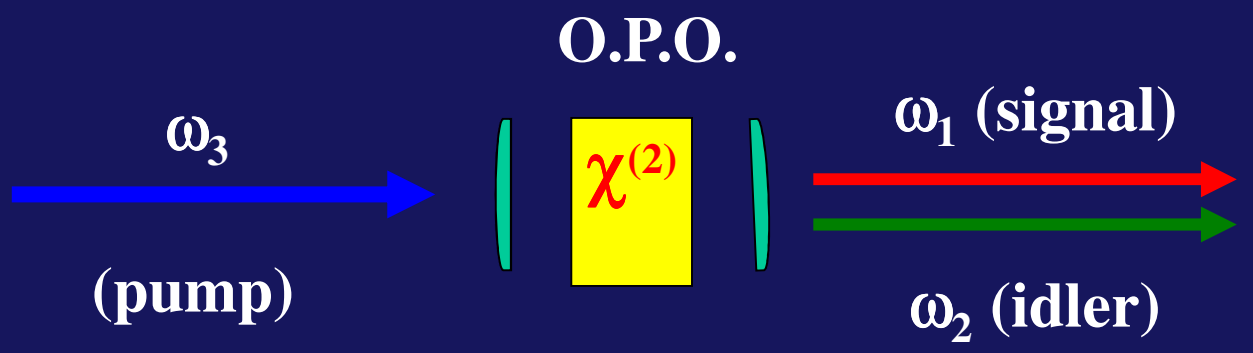


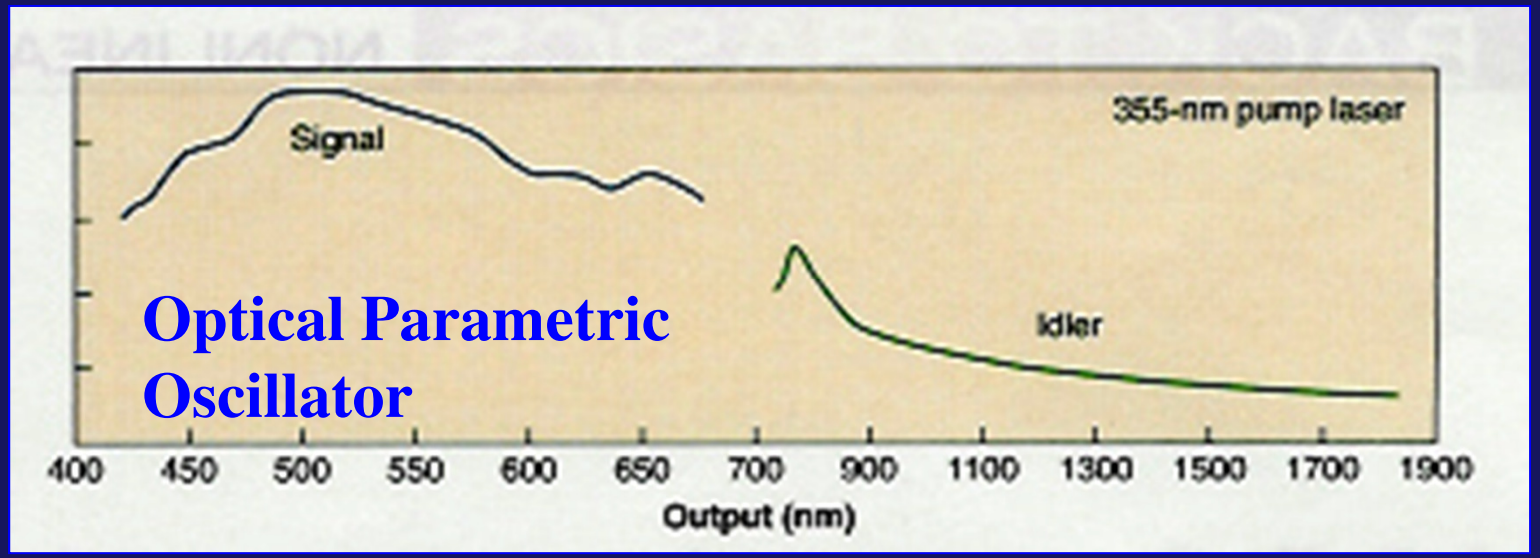
$\chi^{(2)}$ is a tensor hence $P_{2\omega}$ and the incident electric field are not parallel

SHG, SFG, DFG are observed in single pass configuration

Pulsed systems ⇔ High peak power

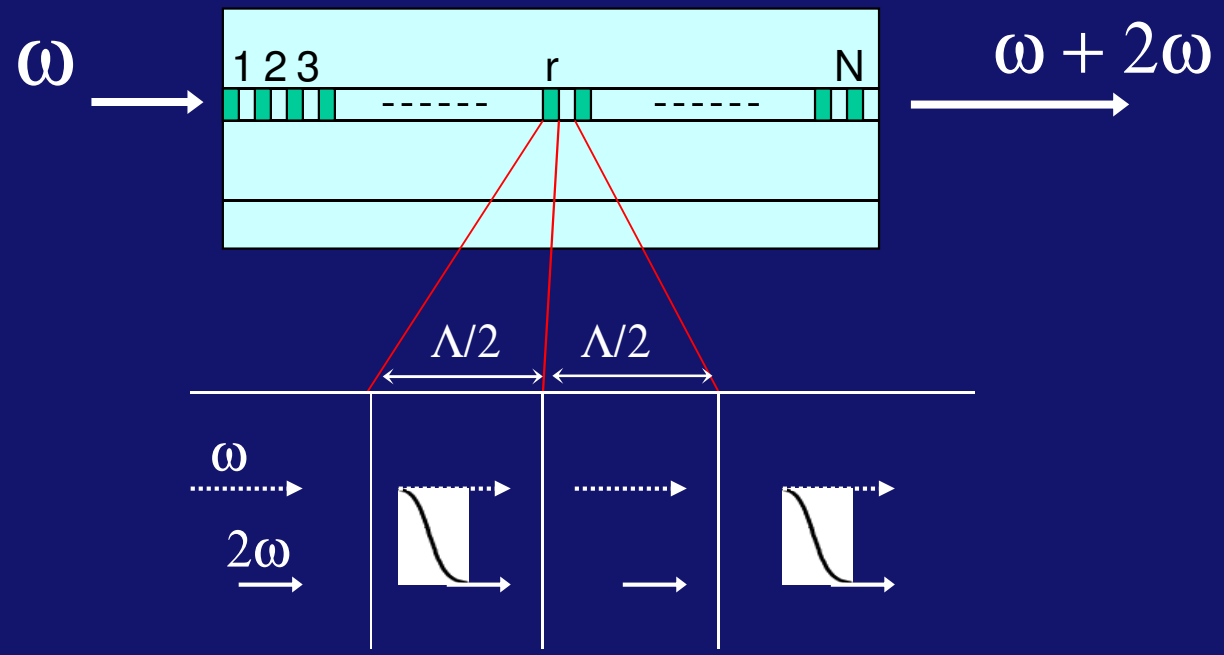
Larger conversion efficiency working with resonant cavities





Quasi Phase-Matching

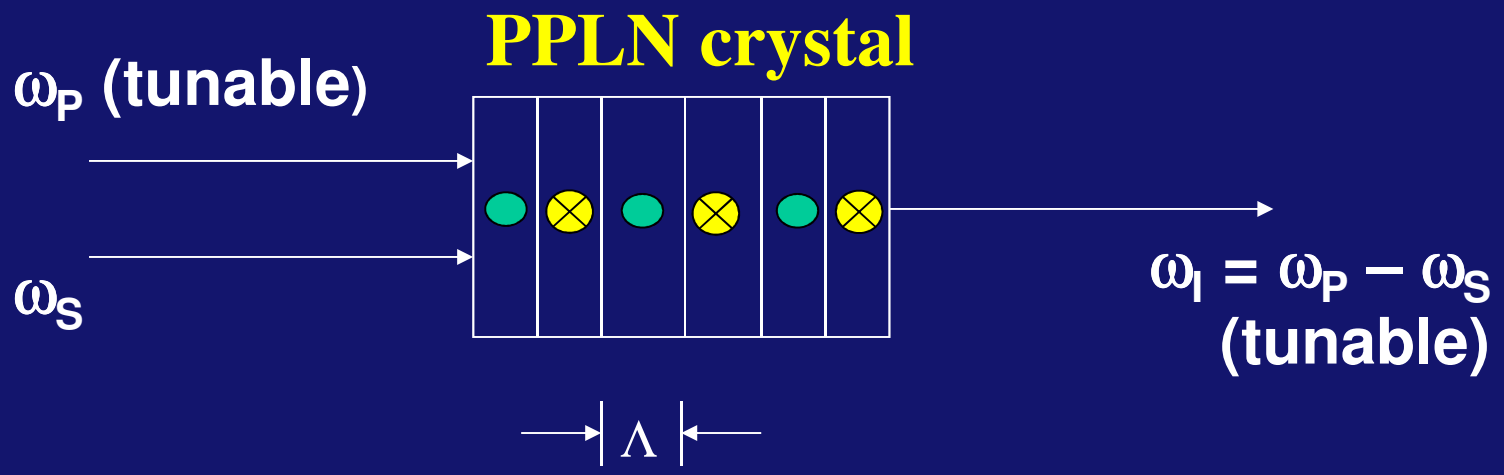
reversal in $\chi^{(2)}$ every coherent length to compensate destructive interference



Bloembergen et al. Phys. Rev. 127 (1962) 1918

First SHG experiment using periodic poled Lithium Niobate (PPLN) – Yamada, APL 62 (1993) 435

Difference frequency generation in QPM

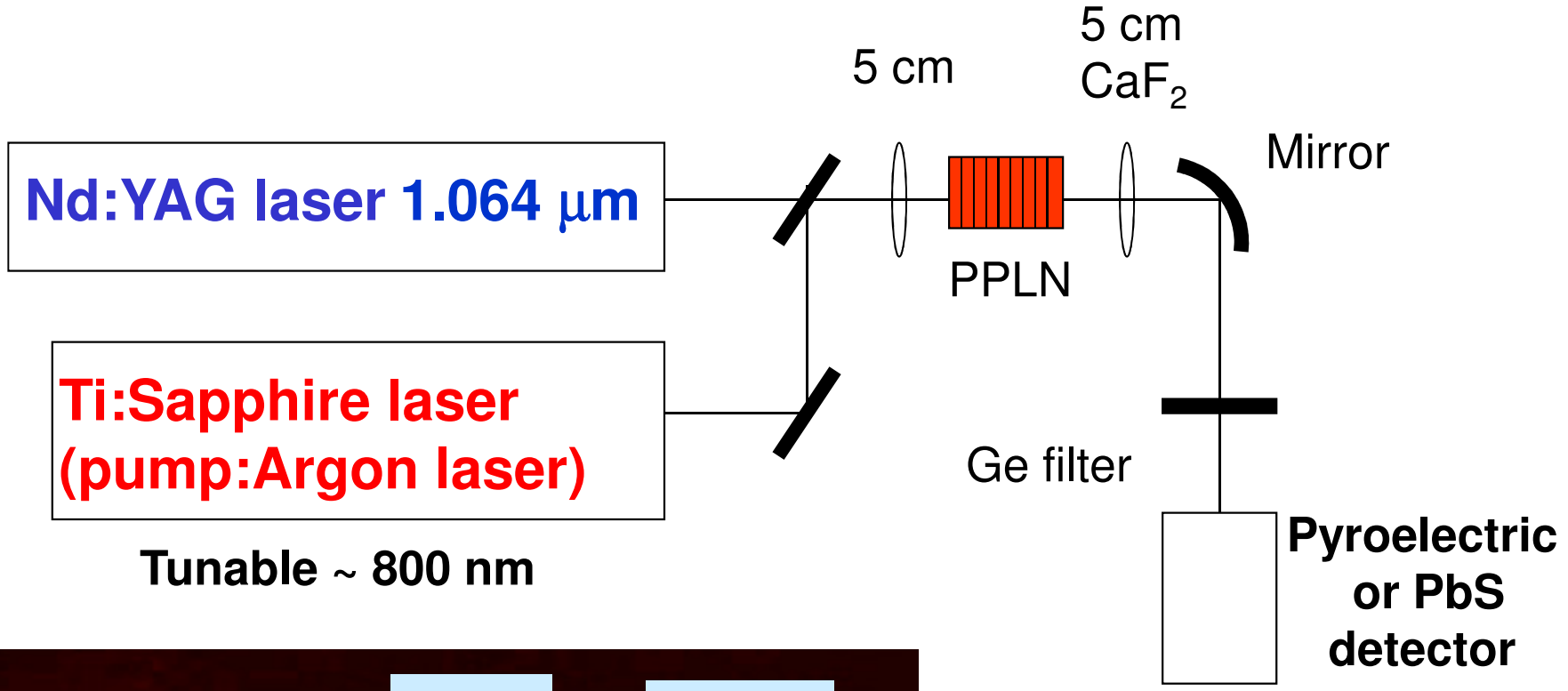


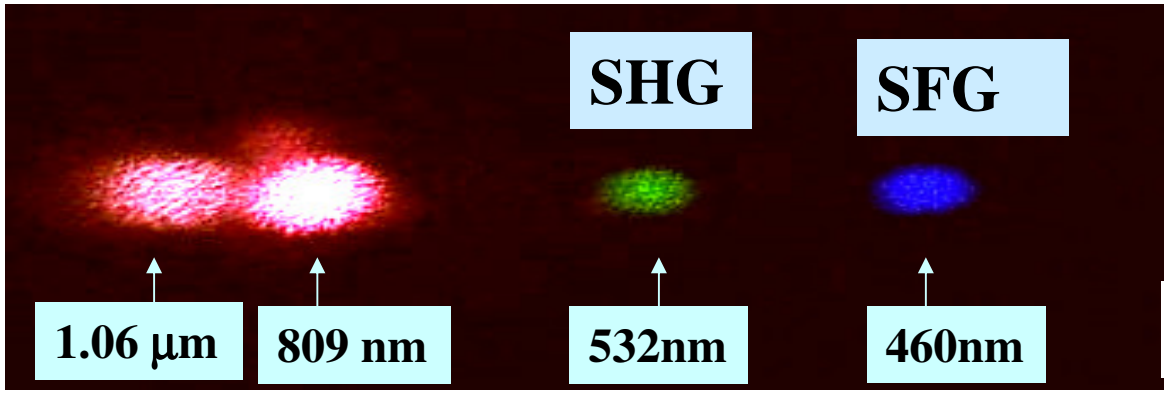
$$\omega_P = \omega_S + \omega_I \qquad k_P - k_S - k_I - \frac{2\pi}{\Lambda} = 0$$

$$\frac{1}{\lambda_P} - \frac{1}{\lambda_S} - \frac{1}{\lambda_I} = 0 \qquad \frac{n_P(T)}{\lambda_P} - \frac{n_S(T)}{\lambda_S} - \frac{n_I(T)}{\lambda_I} - \frac{1}{\Lambda} = 0$$

- n's are calculated using Sellmeier eq for $LiNbO_3$
- Λ is selected to satisfy QPM for a particular case

Experimental setup (UFPE)





$$k_P - k_S - k_I - \frac{2\pi m}{\Lambda} = 0$$

$m = 5$, QPM de 5a Ordem

Experimental results
(T = 25 °C λ_S = 1.0642 μm)

Emission at 3.29 μm

$P_{P,\,in}$ = 350 mW
$P_{S,\,in}$ = 3 W
$P_{I,\,out}$ = 400 μW

$$\eta = \frac{P_I}{P_P\, P_S\, L} \approx$$ **0.03 % / W.cm**

400 μW at 3.29 μm,
enough to detect CH_4 with concentration below ppb

mid infrared - multiple NL interactions with arbitrary wave vector diferences: Fradkin-Kashi et al. PRL 88 (2002)023903]

scheme based on cascaded SFG and DFG:
Xu et al., Opt. Lett. 29 (2004) 292

Proposta de calendário

Listas de exercícios		Provas	
16 / 03	1ª. Lista	11 / 05	1ª. Prova
13 / 04	2ª. Lista	29 / 06	2ª. Prova
18 / 05	3ª. Lista		
15 / 06	4ª. Lista		