

FI255 - Tópicos de Óptica e Fotônica II

Óptica Não-Linear

5^a. aula

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UNICAMP - 06 de abril de 2018

Proposta de calendário

Listas de exercícios

16 / 03 1^a. Lista

13 / 04 2^a. Lista

18 / 05 3^a. Lista

15 / 06 4^a. Lista

Provas

11 / 05 1^a. Prova

29 / 06 2^a. Prova

Breve revisão da 4^a. Aula

Óptica Não Linear

REVISÃO

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

Polarização óptica \equiv momento de dipolo por unidade de volume

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

$\overleftrightarrow{\chi}^{(n)} \equiv 0$, $n = \text{par}$ (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

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

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

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

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 termos ...

$$m \frac{d^2 x}{dt^2} + m\gamma \frac{dx}{dt} + m\omega_0^2 x + mKx^2 = -e\varepsilon(t)$$

$$\varepsilon(t) = \varepsilon_0 \cos \omega t = \frac{1}{2} \varepsilon_0 (e^{i\omega t} + e^{-i\omega t})$$

Solução tentativa

$$x(t) = \frac{1}{2} (X_1 e^{i\omega t} + X_2 e^{i2\omega t} + c.c.) \quad X_1 \gg X_2$$

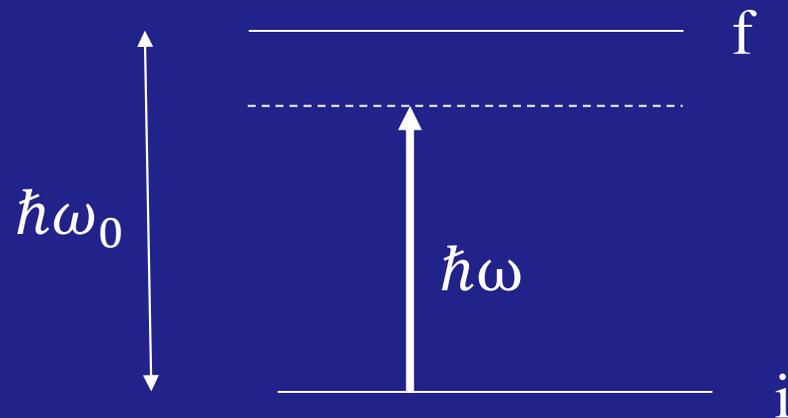
Polarização
induzida

$$X_1 = - \frac{e\varepsilon_0}{m} \frac{1}{(\omega_0^2 - \omega^2) + i\gamma\omega}$$

$$\begin{aligned} P(\omega, t) &= -Nex(\omega, t) = - \frac{Ne}{2} (X_1 e^{i\omega t} + c.c.) \\ &= \varepsilon_0 \chi^{(1)}(\omega) \varepsilon(t) \end{aligned}$$

$$\chi^{(1)}(\omega; \omega) = \frac{Ne^2}{m\varepsilon_0 [(\omega_0^2 - \omega^2) + i\gamma\omega]}$$

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



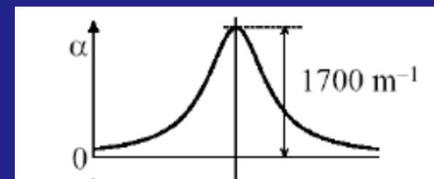
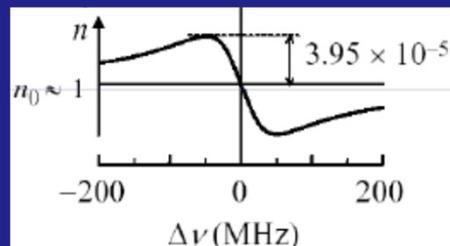
$$n_0 \propto \text{Re } \chi^{(1)}(\omega; \omega)$$

Índice de refração linear

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

Coefficiente de absorção linear

Exemplo: sódio gasoso (2ª. Aula)

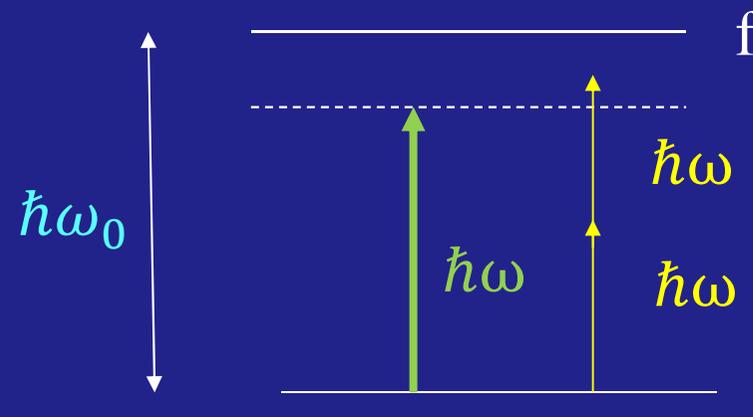


$$P(2\omega, t) = -Nex(2\omega, t) = -\frac{Ne}{2} (X_2 e^{i2\omega t} + c.c.)$$

$$= \epsilon_0 \chi^{(2)} \varepsilon(t)^2$$

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

ressonâncias



$$\varepsilon(t) = \varepsilon_1 \cos \omega_1 t + \varepsilon_2 \cos \omega_2 t$$

$$P = -Ne x = P^{(1)} + P^{(2)} + P^{(3)} \dots$$

Polarização de primeira ordem

$$P^{(1)}(\omega_1) \text{ and } P^{(1)}(\omega_2)$$

Polarização de segunda ordem

$$P^{(2)}(2\omega_1), P^{(2)}(2\omega_2), P^{(2)}(\omega_1 \pm \omega_2), P^{(2)}(0)$$

Polarização de terceira ordem... etc

$\chi^{(2)}$

Second order polarization

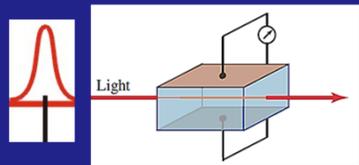
Non centro-symmetric materials

REVISÃO

$$P^{(2)} = \epsilon_0 \chi^{(2)} (E_1 \cos \omega_1 t + E_2 \cos \omega_2 t)^2 =$$

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

Optical rectification

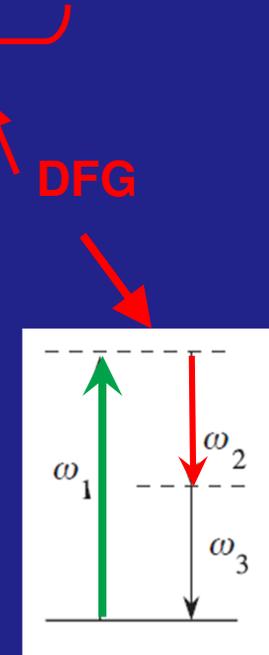
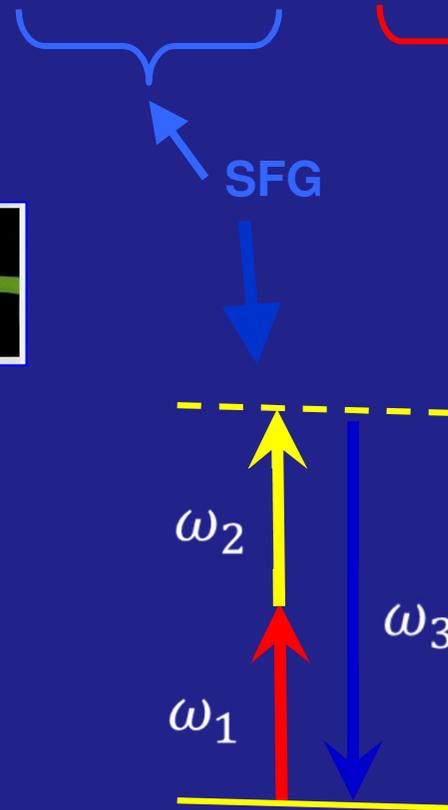
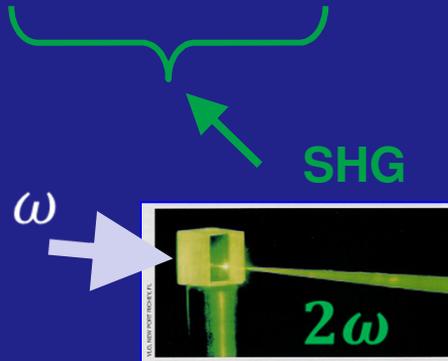


Light pulse

Voltage



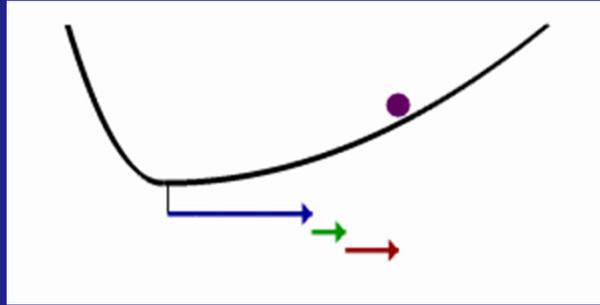
time



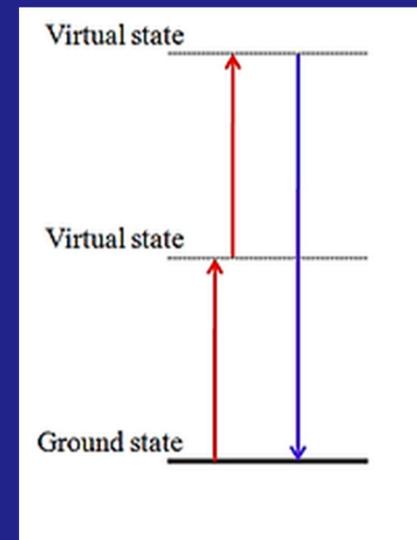
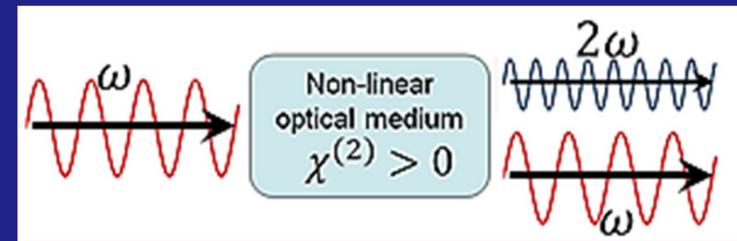
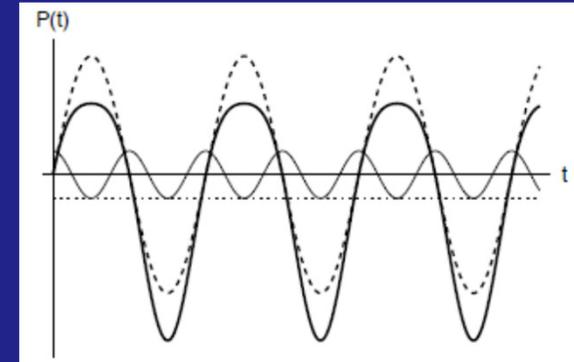
Estudaremos mais adiante alguns efeitos de mais alta ordem, porém, antes disso, veremos em detalhes como descrever matematicamente os efeitos de segunda e terceira ordem.

Nesta aula e na próxima vamos detalhar os cálculos para descrever a intensidade do sinal gerado devido à susceptibilidade $\chi^{(2)}$

As aulas seguintes (7^a, 8^a, 9^a) serão dedicadas aos efeitos de terceira ordem (associados a $\chi^{(3)}$)

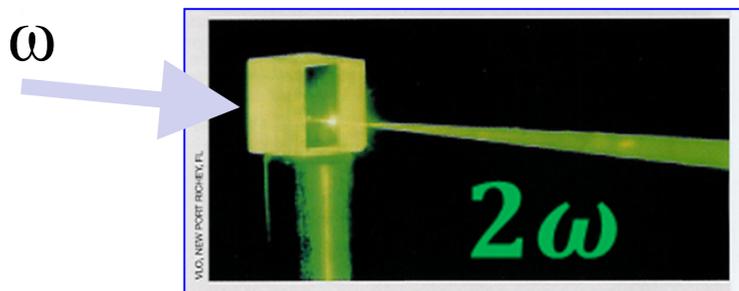


An electron (purple) is being pushed side-to-side by a sinusoidally-oscillating force, i.e. the light's electric field. But because the electron is in an anharmonic potential energy environment (black curve), the electron motion is *not* sinusoidal. The three arrows show the Fourier series of the motion: The blue arrow corresponds to ordinary (linear) susceptibility, the green arrow corresponds to second-harmonic generation, and the red arrow corresponds to optical rectification.

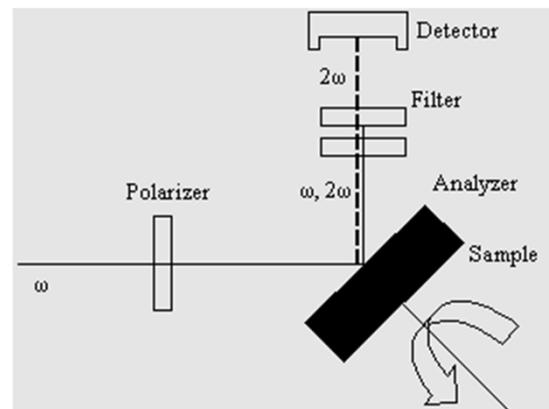


https://en.wikipedia.org/wiki/Second-harmonic_generation

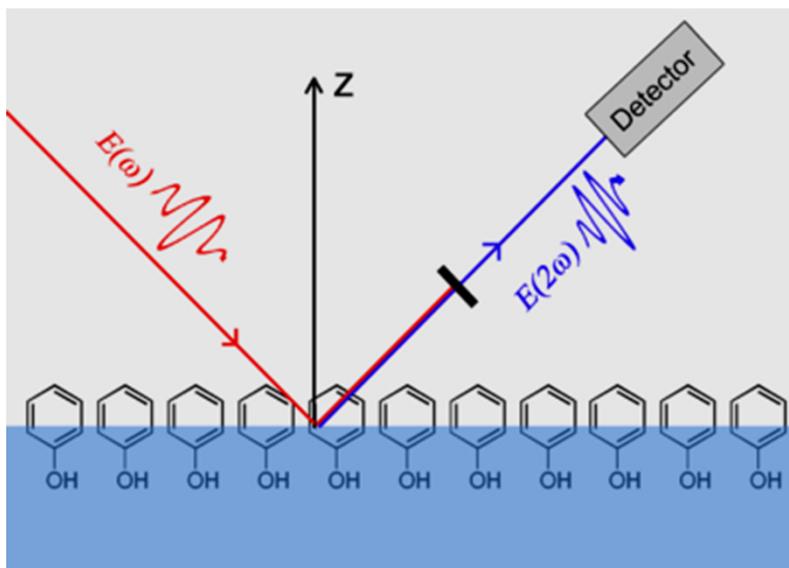
Bulk crystals (1961)



Crystal surface (1962)

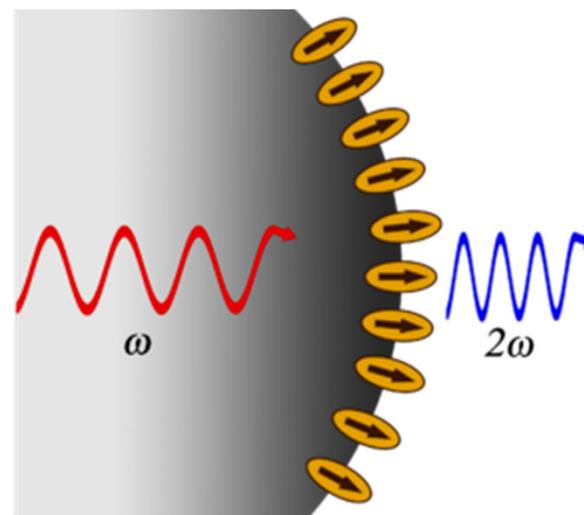


Phenol group at the air-water interface



Many analogous experiments starting in the 70's

Molecules on a curved surface

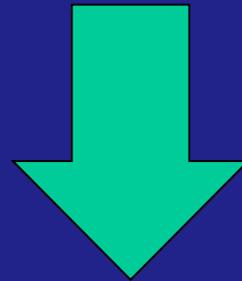


Many contemporary experiments with nanoparticles

Equação de onda para um meio não linear

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

$$\mu = \mu_0$$



$$\epsilon = \epsilon(\omega)$$

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

Processos não lineares de segunda ordem

Dois campos incidentes com frequências ω_1 e ω_2

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

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

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


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

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

$$\frac{\partial^2 E_i(z, t, \omega_1)}{\partial z^2} - \mu \epsilon_1 \frac{\partial^2 E_i(z, t, \omega_1)}{\partial t^2} = \mu \frac{\partial^2 P_i^{(2)}(z, t, \omega_1)}{\partial t^2},$$

$$\frac{\partial^2 E_j(z, t, \omega_2)}{\partial z^2} - \mu \epsilon_2 \frac{\partial^2 E_j(z, t, \omega_2)}{\partial t^2} = \mu \frac{\partial^2 P_j^{(2)}(z, t, \omega_2)}{\partial t^2},$$

$$\frac{\partial^2 E_k(z, t, \omega)}{\partial z^2} - \mu \epsilon \frac{\partial^2 E_k(z, t, \omega)}{\partial t^2} = \mu \frac{\partial^2 P_k^{(2)}(z, t, \omega)}{\partial t^2},$$

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

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

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

$$\frac{d^2 [E_i(z, \omega_1) e^{ik_1 z}]}{dz^2} + \mu \epsilon_1 \omega_1^2 E_i(z, \omega_1) e^{ik_1 z} = -\mu \omega_1^2 P_i^{(2)}(z, \omega_1),$$

$$\frac{d^2 [E_j(z, \omega_2) e^{ik_2 z}]}{dz^2} + \mu \epsilon_2 \omega_2^2 E_j(z, \omega_2) e^{ik_2 z} = -\mu \omega_2^2 P_j^{(2)}(z, \omega_2),$$

$$\frac{d^2 [E_k(z, \omega) e^{ikz}]}{dz^2} + \mu \epsilon \omega^2 E_k(z, \omega) e^{ikz} = -\mu \omega^2 P_k^{(2)}(z, \omega).$$

Sum frequency generation

$$\omega = \omega_1 + \omega_2 \quad \vec{k} = \vec{k}_1 + \vec{k}_2$$

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

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

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

$$\begin{aligned} & \frac{d^2}{dz^2} [E_i(z, \omega_1)e^{ik_1z}] + \mu\epsilon_1\omega_1^2 E_i(z, \omega_1)e^{ik_1z} \\ &= \left[\frac{d^2 E_i(z, \omega_1)}{dz^2} + 2ik_1 \frac{dE_i(z, \omega_1)}{dz} + (\mu\epsilon_1\omega_1^2 - k_1^2) E_i(z, \omega_1) \right] e^{ik_1z} \\ & \approx 2ik_1 e^{ik_1z} \frac{dE_i(z, \omega_1)}{dz}. \end{aligned}$$

$$\begin{aligned} \frac{d^2}{dz^2} [E_j(z, \omega_2)e^{ik_2z}] + \mu\epsilon_2\omega_2^2 E_j(z, \omega_2)e^{ik_2z} &= 2ik_2 e^{ik_2z} \frac{dE_j(z, \omega_2)}{dz}, \\ \frac{d^2}{dz^2} [E_k(z, \omega)e^{ikz}] + \mu\epsilon\omega^2 E_k(z, \omega)e^{ikz} &= 2ike^{ikz} \frac{dE_k(z, \omega)}{dz}. \end{aligned}$$

SVEA

$$\frac{d^2 E(z, \omega)}{dz^2} \ll k \frac{dE(z, \omega)}{dz}$$

$$k_1^2 = \mu\epsilon_1\omega_1^2, \quad k_2^2 = \mu\epsilon_2\omega_2^2, \quad k^2 = \mu\epsilon\omega^2$$

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

$$\frac{dE_i(z, \omega_1)}{dz} = \frac{i\omega_1}{4cn_1} \sum_{jk} \chi_{ijk}^{(2)}(\omega_1, -\omega_2, \omega) E_j^*(z, \omega_2) E_k(z, \omega) e^{i\Delta kz},$$

$$\frac{dE_j(z, \omega_2)}{dz} = \frac{i\omega_2}{4cn_2} \sum_{ki} \chi_{jki}^{(2)}(\omega_2, \omega, -\omega_1) E_k(z, \omega) E_i^*(z, \omega_1) e^{i\Delta kz},$$

$$\frac{dE_k(z, \omega)}{dz} = \frac{i\omega}{4cn} \sum_{ij} \chi_{kij}^{(2)}(\omega, \omega_1, \omega_2) E_i(z, \omega_1) E_j(z, \omega_2) e^{-i\Delta kz},$$

$$\frac{dE_i(\omega_1)}{dz} = i\alpha \frac{\omega_1}{n_1} E_j^*(\omega_2) E_k(\omega) e^{i\Delta kz},$$

$$\frac{dE_j(\omega_2)}{dz} = i\alpha \frac{\omega_2}{n_2} E_k(\omega) E_i^*(\omega_1) e^{i\Delta kz},$$

$$\frac{dE_k(\omega)}{dz} = i\alpha \frac{\omega}{n} E_i(\omega_1) E_j(\omega_2) e^{-i\Delta kz},$$

Transparent crystal

$$\alpha = \frac{1}{4c} \chi_{\text{eff}}^{(2)} = \frac{1}{2c} d_{\text{eff}}^{(2)}$$

Intensidades dos feixes incidentes e feixe gerado

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

$$\frac{dI(\omega_1)}{dz} = \frac{1}{2} \epsilon_0 n_1 c \left[E_i(\omega_1) \frac{dE_i^*(\omega_1)}{dz} + E_i^*(\omega_1) \frac{dE_i(\omega_1)}{dz} \right]$$

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

Parte
imaginária

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

$$\frac{d}{dz} [I(\omega_1) + I(\omega_2) + I(\omega = \omega_1 + \omega_2)] = 0$$

$$\frac{1}{\omega} \frac{dI(\omega)}{dz} = -\frac{1}{\omega_1} \frac{dI(\omega_1)}{dz} = -\frac{1}{\omega_2} \frac{dI(\omega_2)}{dz}$$

Campo gerado

$$\frac{dE_k(\omega)}{dz} = i\alpha \frac{\omega}{n} E_i(\omega_1) E_j(\omega_2) e^{-i\Delta k z}$$

Saida
da
amostra

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

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

Intensidade gerada

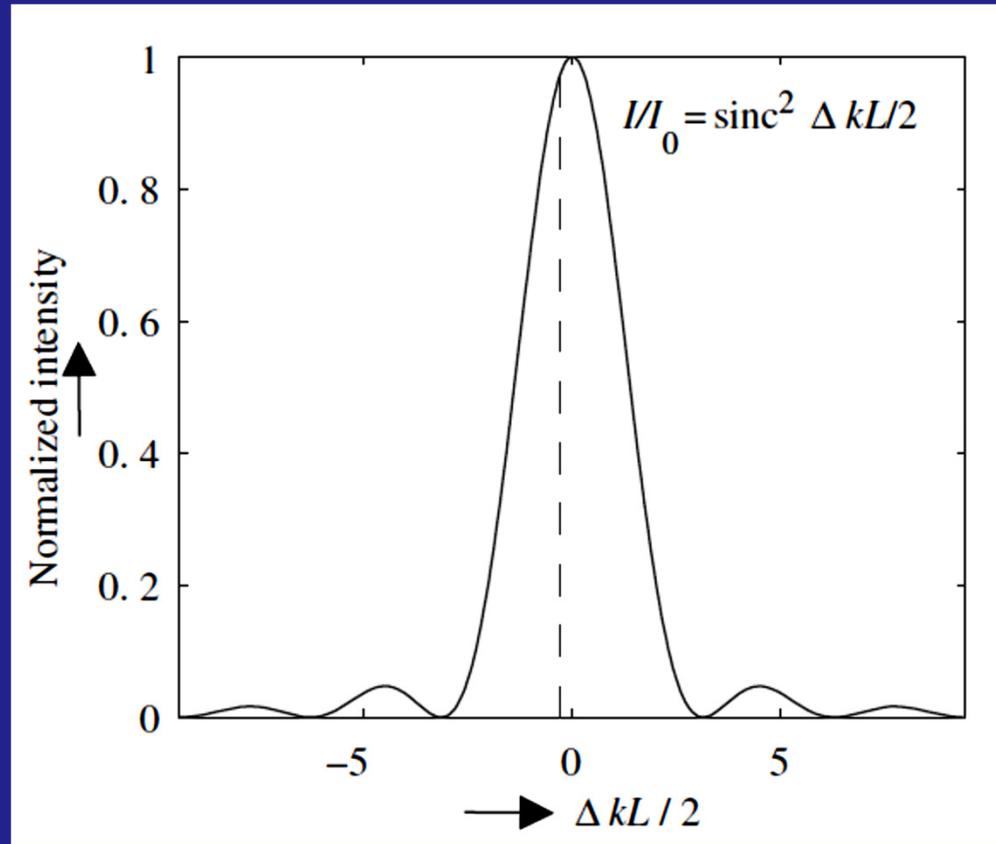
$$I(\omega_1 + \omega_2, z = L) = I_0 \operatorname{sinc}^2 \left(\frac{\Delta k L}{2} \right)$$

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

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

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

$$\operatorname{sinc} \left(\frac{\Delta k L}{2} \right) = \frac{\sin(\Delta k L / 2)}{\Delta k L / 2}$$



$$\theta \approx \frac{\Delta k}{k} = \frac{2\pi/L}{2\pi/\lambda} = \frac{\lambda}{L}$$

ANISOTROPIA ÓPTICA

ÁTOMOS NUM GAZ OU LÍQUIDO NÃO TÊM DIREÇÕES PREFERENCIAIS SE NÃO HOUVER UMA PERTURBAÇÃO EXTERNA

DEVIDO À SIMETRIA DA REDE CRISTALINA OS SÓLIDOS PODEM APRESENTAR ANISOTROPIA ÓPTICA DANDO ORIGEM AO FENÔMENO DE BIRREFRINGÊNCIA

CONSEQUÊNCIA

OS ÍNDICES DE REFRAÇÃO DEPENDEM DA DIREÇÃO DE PROPAGAÇÃO DA LUZ

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2}$$

Optical anisotropy

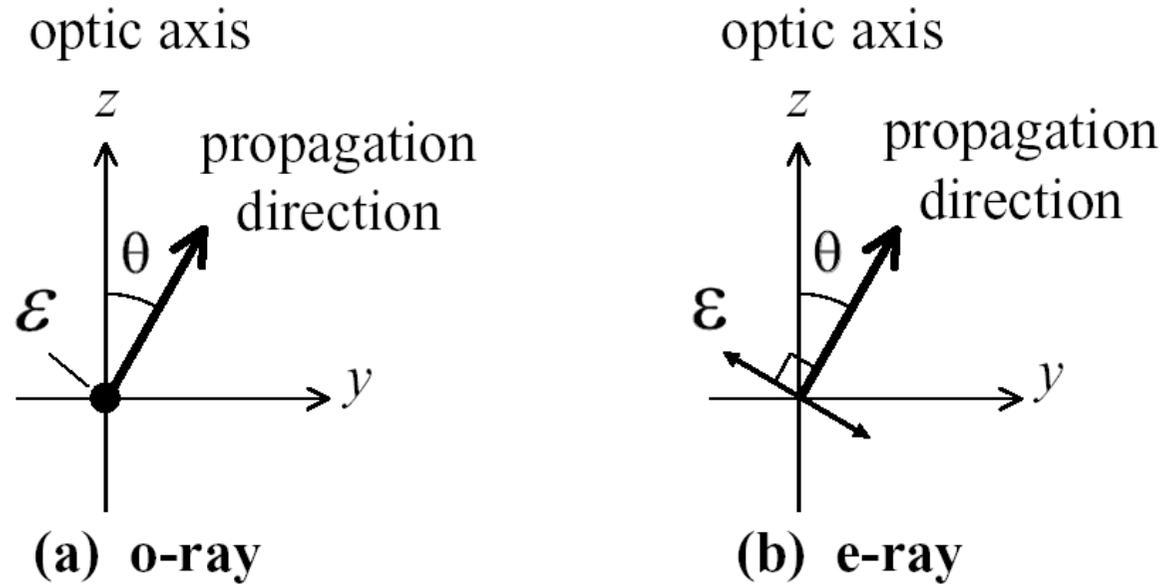


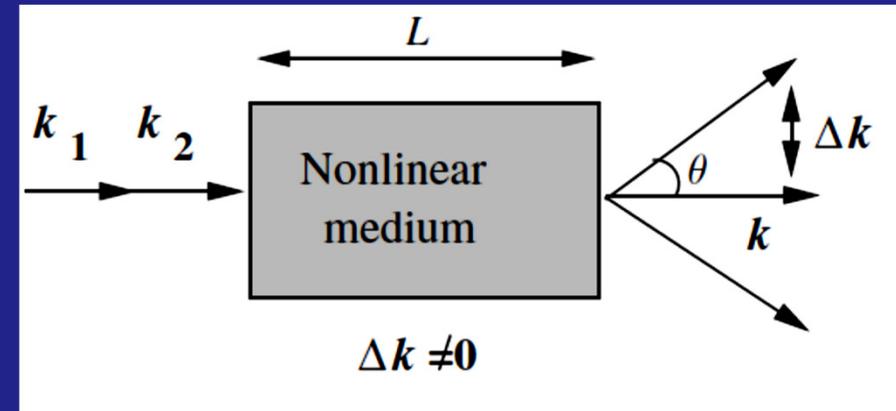
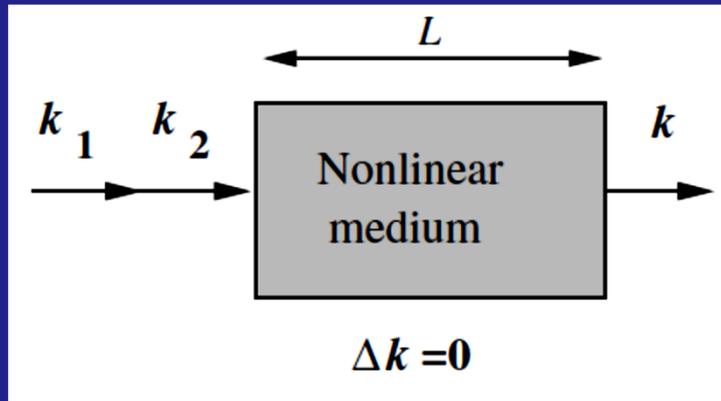
Figure 2.12

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

$$\begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \epsilon_0 \begin{pmatrix} \epsilon_{11} & 0 & 0 \\ 0 & \epsilon_{22} & 0 \\ 0 & 0 & \epsilon_{33} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

Phase Matching

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



Type I Phase Matching

$$n_o(\omega_1 + \omega_2) = \frac{\omega_1}{\omega_1 + \omega_2} n_e(\omega_1) + \frac{\omega_2}{\omega_1 + \omega_2} n_e(\omega_2)$$

Type II Phase Matching

$$n_o(\omega_1 + \omega_2) = \frac{\omega_1}{\omega_1 + \omega_2} n_o(\omega_1) + \frac{\omega_2}{\omega_1 + \omega_2} n_e(\omega_2)$$

Applications

Interface structure

Adsorption measurements

Molecular orientation

**Confocal Microscopy:
monolayers, single molecule, single nanoparticles**

Many applications in biology and medical physics

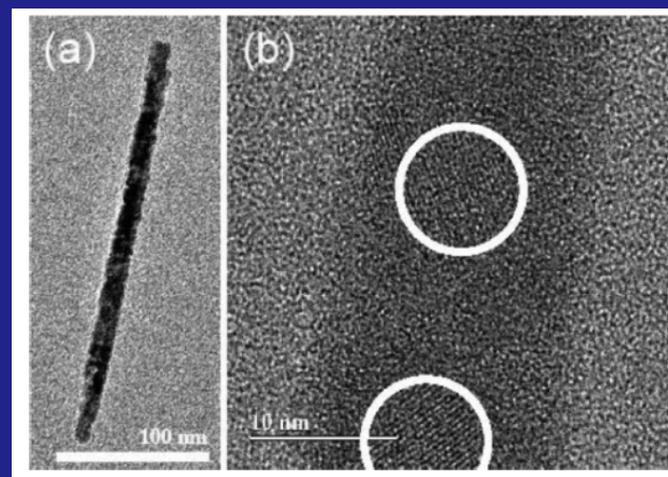
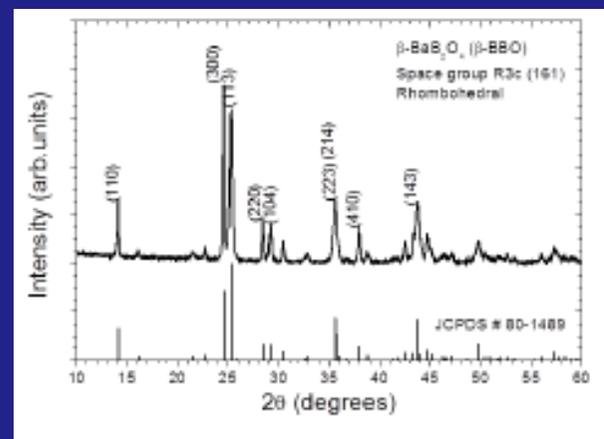
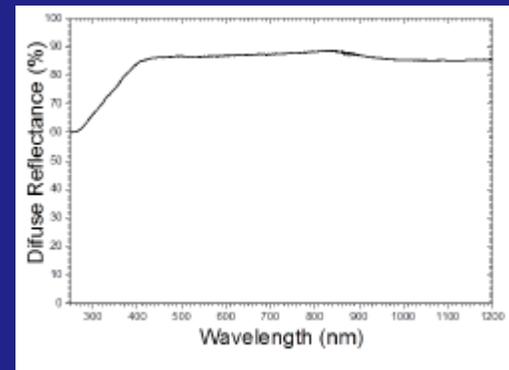


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PROGRAMA DE PÓS-GRADUAÇÃO EM FÍSICA

SECOND-HARMONIC CONFOCAL MICROSCOPY OF
SINGLE β -BBO NANOCRYSTALS

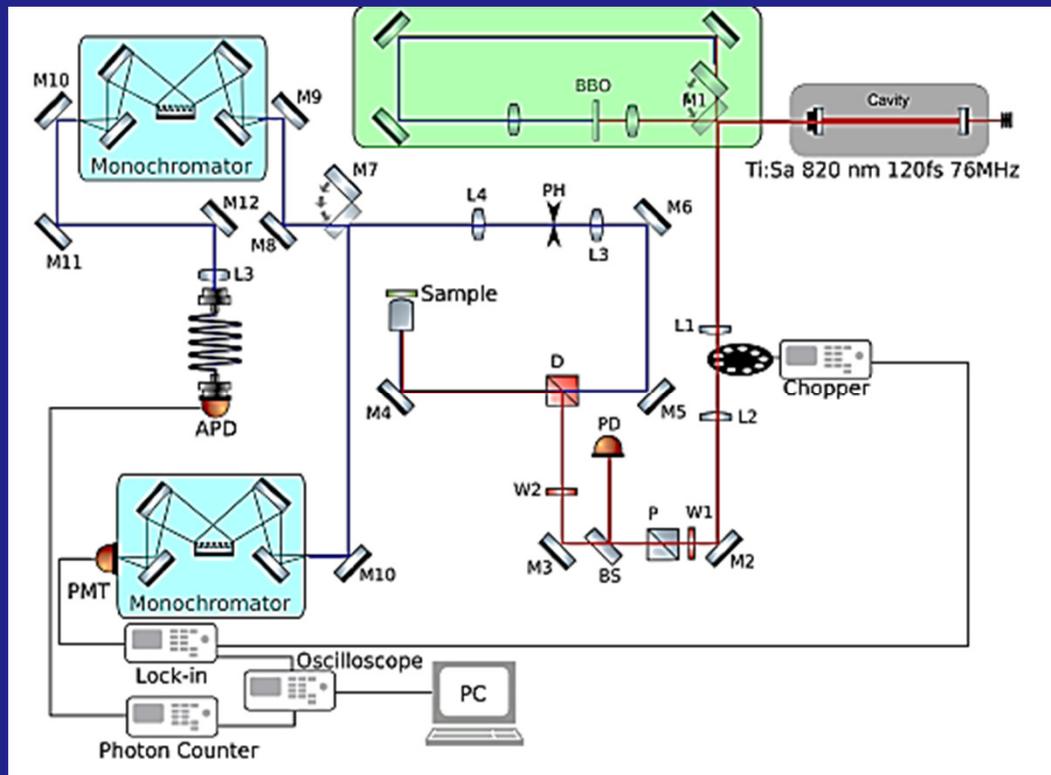
RODRIGO GALVÃO DOS SANTOS

2017

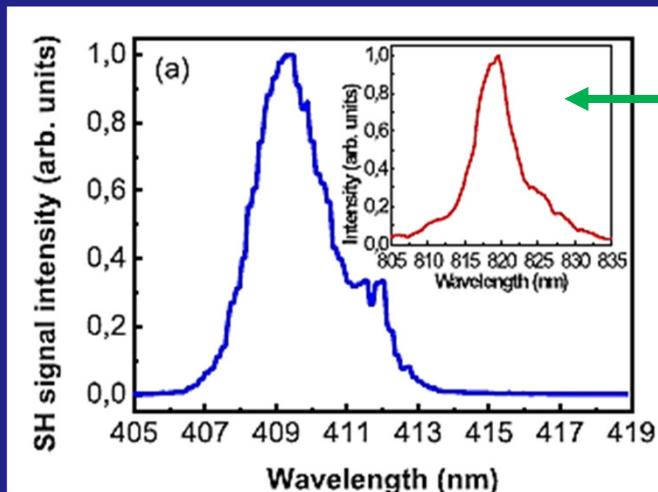
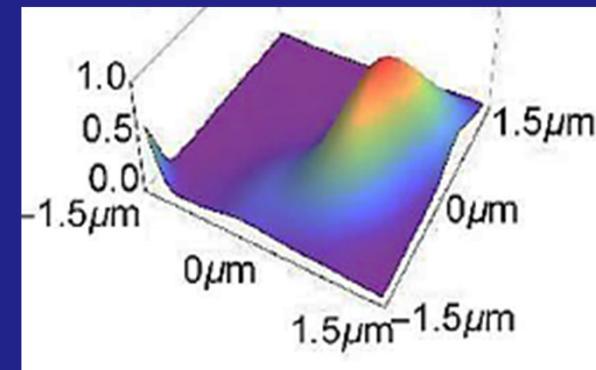


β – BBO nanocrystals

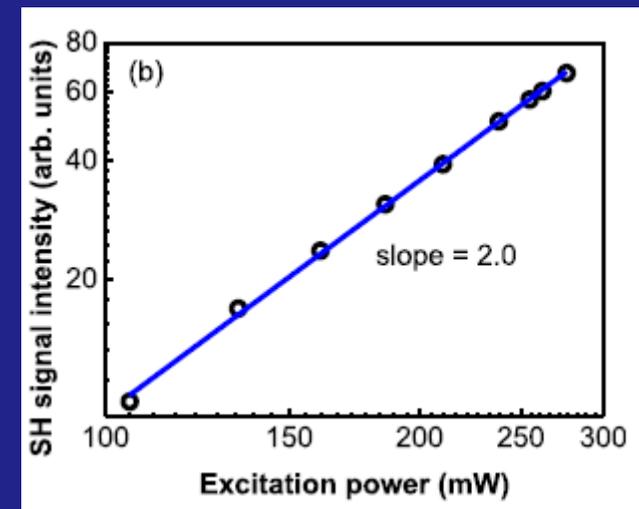
(BaB₂O₄) Beta Barium Borate



Single nanocrystal

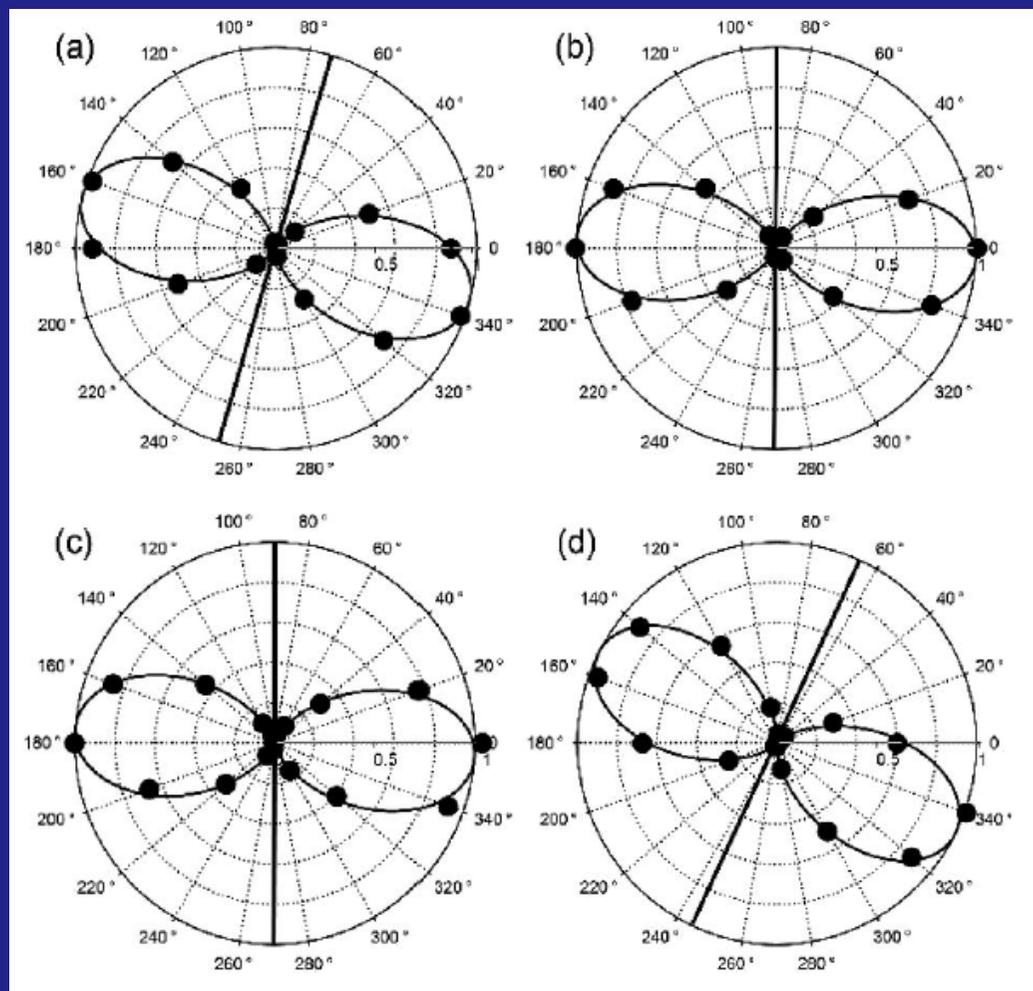


Laser spectrum



Orientação dos nanocristais

A linha reta indica o eixo do NC



References for applications in biology, medical physics, biophotonics

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Proposta de calendário

Listas de exercícios

16 / 03 1^a. Lista

13 / 04 2^a. Lista

18 / 05 3^a. Lista

15 / 06 4^a. Lista

Provas

11 / 05 1^a. Prova

29 / 06 2^a. Prova