

Óptica não linear



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Tópicos das aulas

- 1) Introdução à óptica não linear, P_{NL} e χ^{NL}
- 2) Equação de ondas não linear (SVEA)
- 3) Cálculo da susceptibilidade óptica
- 4) Índice de refração dependente da intensidade

Referências:

- R. W. Boyd, Nonlinear Optics, 3ª ed.
- Y. R. Shen, The Principles of Nonlinear Optics
- N. Bloembergen, Nonlinear Optics

<http://www.fotonica.ifsc.usp.br> presentations

Aula 1 – Introdução à óptica não linear



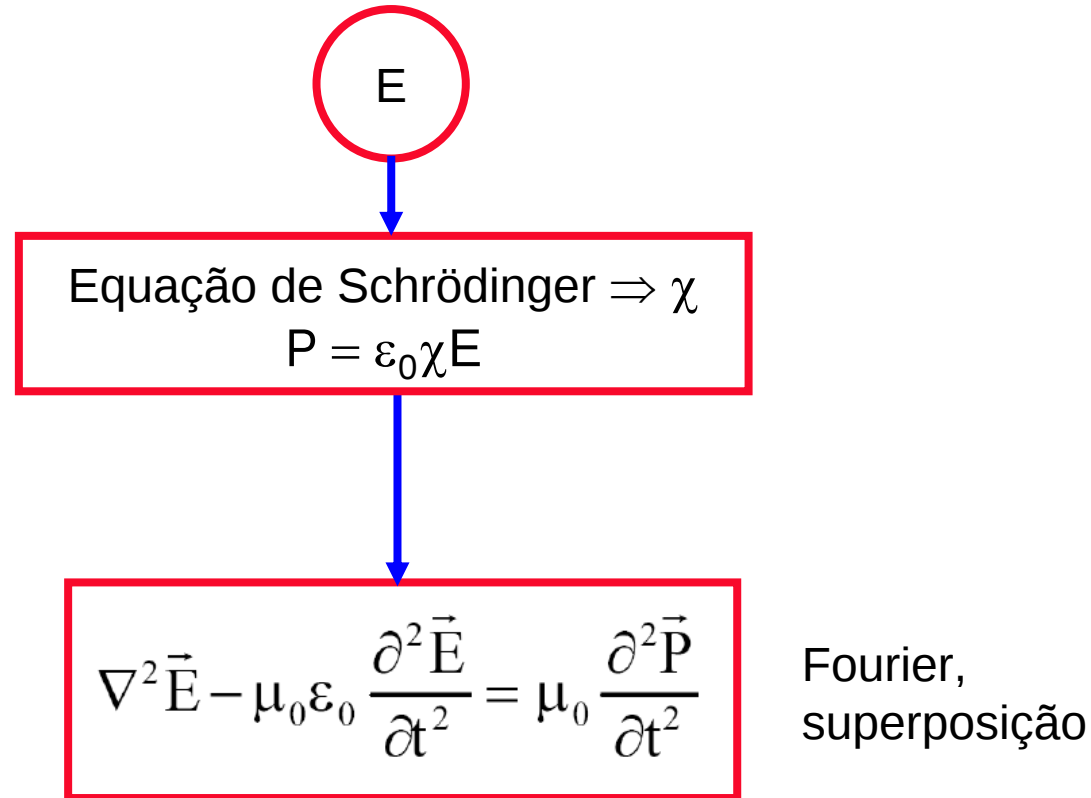
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1.1) Polarização não linear

- Óptica não linear: estuda fenômenos devidos às modificações das propriedades ópticas de um material pela presença de luz.
- Efeitos ópticos não lineares já eram observados no século 19: efeito Pockels, efeito Kerr. Efeito Raman em 1923.
- Os altos campos produzidos por lasers possibilitaram, a partir de 1960, a descoberta de novos efeitos não lineares: geração de harmônicos de várias ordens, espalhamento Raman estimulado, auto-focalização, etc.

Interação luz-matéria – tratamento semi-clássico



- 
- As propriedades ópticas de um material são definidas pela susceptibilidade elétrica, χ .
 - Num meio linear $\mathbf{P}(t) = \epsilon_0 \chi^{(1)} \mathbf{E}(t)$.

$\chi^{(1)}$ independe de E

$\text{Im}(\chi^{(1)})$ é proporcional a k

$\text{Re}(\chi^{(1)})$ é proporcional a n

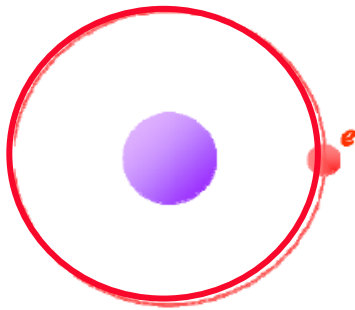
Birrefringência e dicroísmo

- Num meio não linear

$$\begin{aligned}\mathbf{P}(t) &= \epsilon_0 \chi(\mathbf{E}) \mathbf{E}(t) = \epsilon_0 \chi^{(1)} \mathbf{E}(t) + \epsilon_0 \chi^{(2)} \mathbf{E}^2(t) + \epsilon_0 \chi^{(3)} \mathbf{E}^3(t) \\ &= \mathbf{P}^{(1)}(t) + \mathbf{P}^{(2)}(t) + \mathbf{P}^{(3)}(t)\end{aligned}$$

Meio transparente (sem absorção) e sem dispersão,
isto é, com resposta instantânea

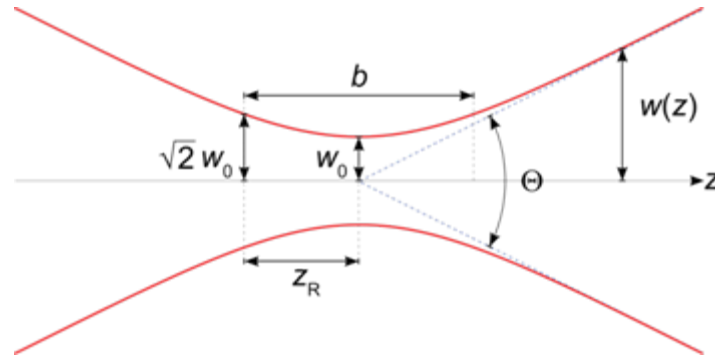
Na óptica não linear, o campo deve ser geralmente alto



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

$$r \sim 4 \text{ \AA}$$

$$E_{\text{atômico}} = 10^{10} \text{ V/m}$$



$$P = 20 \text{ W} \quad w_0 = 20 \text{ \mu m}$$

$$I_0 = 2P/\pi(w_0)^2 \Rightarrow I_0 = 3 \times 10^{10} \text{ W/m}^2$$

$$I_0 = (1/2)cn\epsilon_0(E_0)^2 \Rightarrow E_0 = 4 \times 10^6 \text{ V/m}$$

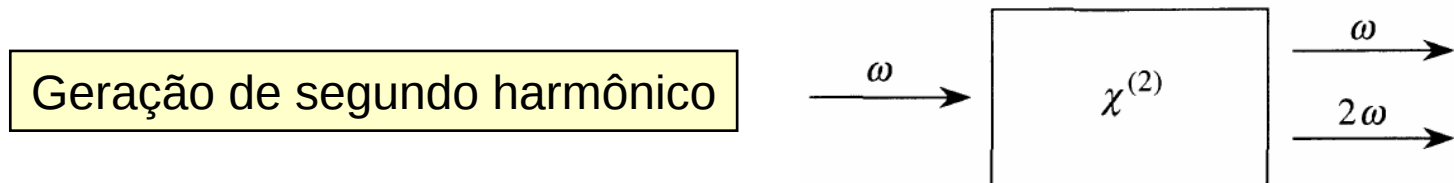
$$I_0 = 10 \text{ GW/cm}^2 = 10^{14} \text{ W/m}^2 \Rightarrow E_0 = 10^8 \text{ V/m}$$

Laser pulsado!

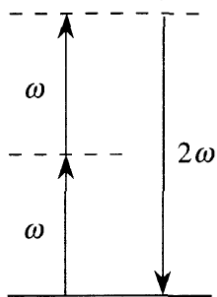
Meio transparente com resposta instantânea

$$P(t) = \epsilon_0 \chi(E) E(t) = \epsilon_0 \chi^{(1)} E(t) + \epsilon_0 \chi^{(2)} E^2(t) + \epsilon_0 \chi^{(3)} E^3(t)$$

1.2) Breve descrição dos processos não lineares



Franken, P.A., Hill, A.E., Peters, C.W., Weinreich, G., 1961. *Phys. Rev. Lett.* 7, 118.

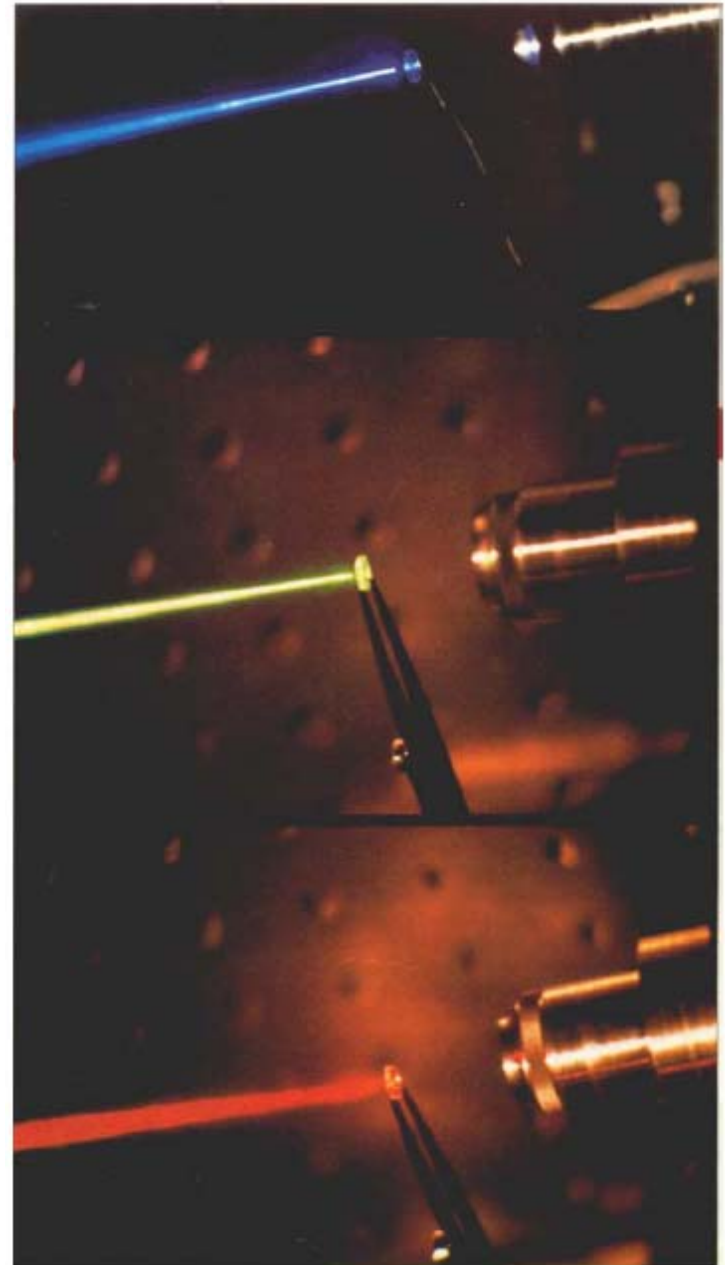


$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} E(t)^2$$

$$E(t) = E e^{-i\omega t} + \text{c.c.}$$

$$P^{(2)}(t) = 2\epsilon_0 \chi^{(2)} E E^* + (\epsilon_0 \chi^{(2)} E^2 e^{-i2\omega t} + \text{c.c.})$$

Geração de segundo harmônico



Geração de soma e diferença de frequências

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

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

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} \left[E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} \right. \\ \left. + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.} \right] + 2\epsilon_0 \chi^{(2)} [E_1 E_1^* + E_2 E_2^*]$$

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

$$P(2\omega_1) = \epsilon_0 \chi^{(2)} E_1^2 \quad (\text{GSH})$$

$$P(2\omega_2) = \epsilon_0 \chi^{(2)} E_2^2 \quad (\text{GSH})$$

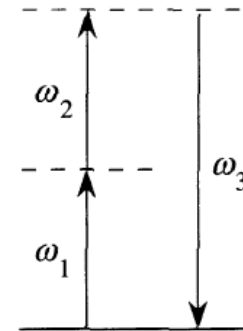
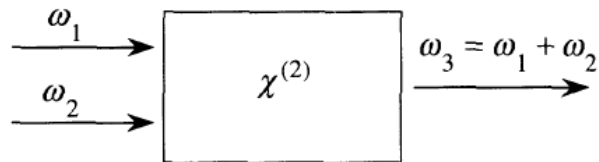
$$P(\omega_1 + \omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2 \quad (\text{GSF})$$

$$P(\omega_1 - \omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2^* \quad (\text{GDF})$$

$$P(0) = 2\epsilon_0 \chi^{(2)} [E_1 E_1^* + E_2 E_2^*] \quad (\text{RO})$$

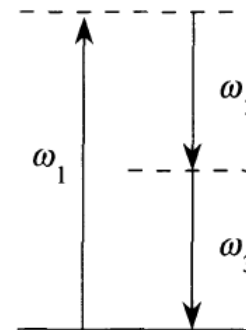
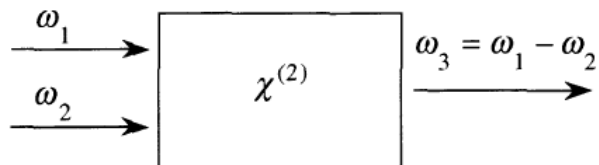
$\omega_1 + \omega_2$: geração de soma de frequências

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

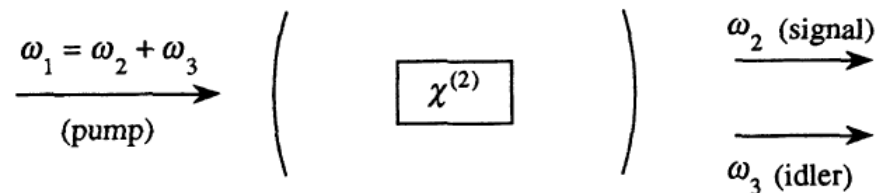


$\omega_1 - \omega_2$: geração de diferença de frequências

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



Oscilação paramétrica



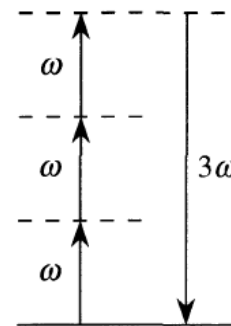
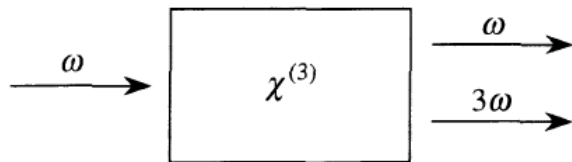
Polarização de terceira ordem

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)$$

Caso monocromático: $E(t) = E_0 \cos \omega t$

$$P^{(3)}(t) = \frac{1}{4} \epsilon_0 \chi^{(3)} E_0^3 \cos 3\omega t + \frac{3}{4} \epsilon_0 \chi^{(3)} E_0^3 \cos \omega t$$

3ω : geração de terceiro harmônico

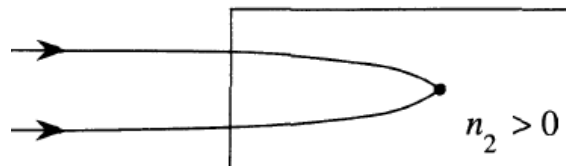


ω : índice de refração dependente da intensidade

$$n = n_0 + n_2 I$$

$$n_2 = (3/4) \chi^{(3)} \mu_0 / n_0^2$$

Auto-focalização



Polarização de terceira ordem – caso geral

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t) \quad E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + \text{c.c.}$$

44 componentes de frequências diferentes:

$$\begin{aligned} &\omega_1, \omega_2, \omega_3, 3\omega_1, 3\omega_2, 3\omega_3, (\omega_1 + \omega_2 + \omega_3), (\omega_1 + \omega_2 - \omega_3), \\ &(\omega_1 + \omega_3 - \omega_2), (\omega_2 + \omega_3 - \omega_1), (2\omega_1 \pm \omega_2), (2\omega_1 \pm \omega_3), (2\omega_2 \pm \omega_1), \\ &(2\omega_2 \pm \omega_3), (2\omega_3 \pm \omega_1), (2\omega_3 \pm \omega_2), \end{aligned}$$

$$P^{(3)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}$$

$$P(\omega_1) = \epsilon_0 \chi^{(3)} [3E_1 E_1^* + 6E_2 E_2^* + 6E_3 E_3^*] E_1$$

$$P(\omega_2) = \epsilon_0 \chi^{(3)} [6E_1 E_1^* + 3E_2 E_2^* + 6E_3 E_3^*] E_2$$

$$P(\omega_3) = \epsilon_0 \chi^{(3)} [6E_1 E_1^* + 6E_2 E_2^* + 3E_3 E_3^*] E_3$$

$$P(3\omega_1) = \epsilon_0 \chi^{(3)} E_1^3 \quad P(3\omega_2) = \epsilon_0 \chi^{(3)} E_2^3 \quad P(3\omega_3) = \epsilon_0 \chi^{(3)} E_3^3$$


$$P(\omega_1 + \omega_2 + \omega_3) = 6\varepsilon_0\chi^{(3)}E_1E_2E_3$$

$$P(\omega_1 - \omega_2 + \omega_3) = 6\varepsilon_0\chi^{(3)}E_1E_2^*E_3$$

$$P(2\omega_1 + \omega_2) = 3\varepsilon_0\chi^{(3)}E_1^2E_2$$

$$P(\omega_1 + 2\omega_2) = 3\varepsilon_0\chi^{(3)}E_1E_2^2$$

$$P(\omega_1 + 2\omega_3) = 3\varepsilon_0\chi^{(3)}E_1E_3^2$$

$$P(2\omega_1 - \omega_2) = 3\varepsilon_0\chi^{(3)}E_1^2E_2^*$$

$$P(-\omega_1 + 2\omega_2) = 3\varepsilon_0\chi^{(3)}E_1^*E_2^2$$

$$P(-\omega_1 + 2\omega_3) = 3\varepsilon_0\chi^{(3)}E_1^*E_3^2$$

$$P(\omega_1 + \omega_2 - \omega_3) = 6\varepsilon_0\chi^{(3)}E_1E_2E_3^*$$

$$P(-\omega_1 + \omega_2 + \omega_3) = 6\varepsilon_0\chi^{(3)}E_1^*E_2E_3$$

$$P(2\omega_1 + \omega_3) = 3\varepsilon_0\chi^{(3)}E_1^2E_3$$

$$P(2\omega_2 + \omega_3) = 3\varepsilon_0\chi^{(3)}E_2^2E_3$$

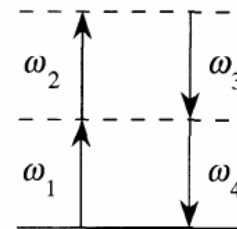
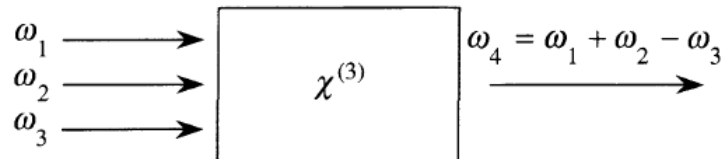
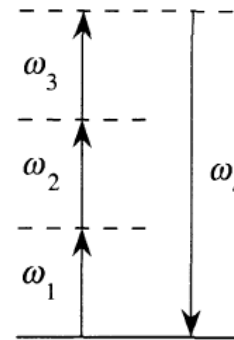
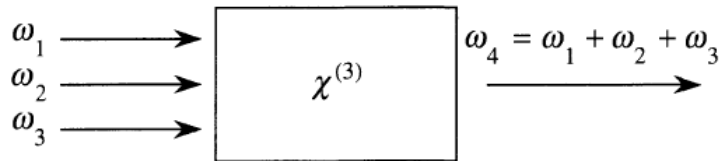
$$P(\omega_2 + 2\omega_3) = 3\varepsilon_0\chi^{(3)}E_2E_3^2$$

$$P(2\omega_1 - \omega_3) = 3\varepsilon_0\chi^{(3)}E_1^2E_3^*$$

$$P(2\omega_2 - \omega_3) = 3\varepsilon_0\chi^{(3)}E_2^2E_3^*$$

$$P(-\omega_2 + 2\omega_3) = 3\varepsilon_0\chi^{(3)}E_2^*E_3^2$$

Exemplos

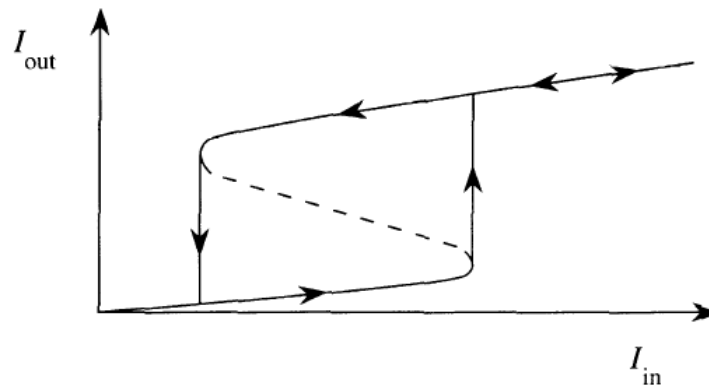
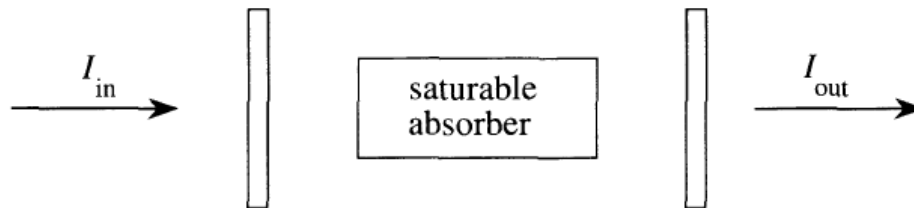


Processos não paramétricos

Absorção saturável

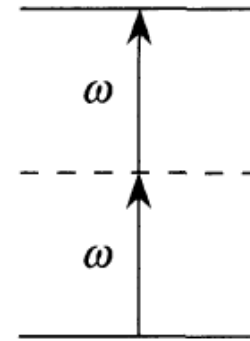
$$\alpha = \frac{\alpha_0}{1 + I/I_s}$$

Bistabilidade óptica

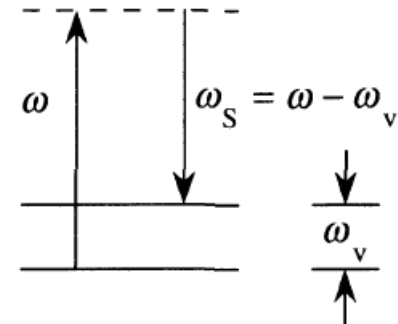
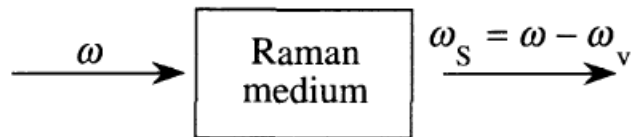


Absorção de dois fótons

$$\sigma = \sigma^{(2)} I, \quad R = \frac{\sigma I}{\hbar \omega} \Rightarrow R = \frac{\sigma^{(2)} I^2}{\hbar \omega}$$



Espalhamento Raman estimulado



A equação de ondas

$$\vec{\nabla}_x \vec{E} = -\partial \vec{B} / \partial t$$

$$\vec{B} = \mu \vec{H} = \mu_0 \vec{H}$$

$$\vec{\nabla}_x \vec{H} = \cancel{\vec{E}} + \frac{\partial \vec{D}}{\partial t}$$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P} = \epsilon_0 (1 + \chi) \vec{E} = \epsilon \vec{E}$$

$$\vec{\nabla} \times (\vec{\nabla} \times \vec{E}) = -\vec{\nabla} \times \left(\frac{\partial \vec{B}}{\partial t} \right) = -\frac{\partial}{\partial t} (\vec{\nabla} \times \vec{B}) = -\mu_0 \frac{\partial}{\partial t} (\vec{\nabla} \times \vec{H})$$

$$\nabla^2 \vec{E} = \mu_0 \frac{\partial^2}{\partial t^2} \vec{D} = \mu_0 \frac{\partial^2 (\epsilon_0 \vec{E} + \vec{P})}{\partial t^2}$$

$$\nabla^2 \vec{E} - \mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

1.3) Definição formal da susceptibilidade não linear

$$\nabla^2 \vec{E} - \mu_0 \varepsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

$$\vec{P} = \varepsilon_0 \tilde{\chi}^{(1)} : \vec{E} + \varepsilon_0 \tilde{\chi}^{(2)} : \vec{E} \vec{E} + \varepsilon_0 \tilde{\chi}^{(3)} : \vec{E} \vec{E} \vec{E} + \dots$$

$$\nabla^2 \vec{E} - \mu_0 \varepsilon_0 (1 + \tilde{\chi}^{(1)}) : \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}^{NL}}{\partial t^2}$$

$$\tilde{\varepsilon} = (1 + \tilde{\chi}^{(1)})$$

Constante dielétrica

$$P_i^{(1)} = \varepsilon_0 \sum_j \chi_{ij}^{(1)} E_j,$$

$$P_i^{(2)} = \varepsilon_0 \sum_{j,k} \chi_{ijk}^{(2)} E_j E_k,$$

$$P_i^{(3)} = \varepsilon_0 \sum_{j,k,l} \chi_{ijkl}^{(3)} E_j E_k E_l$$

$$\vec{E}(\vec{r}, t) = \sum_{\ell} \vec{E}_{\ell}(\vec{k}_{\ell}, \omega_{\ell}) = \sum_{\ell} \vec{A}_{\ell}(\vec{r}, \omega) \exp\{i(\vec{k}_{\ell} \cdot \vec{r} - \omega_{\ell} t)\}$$

$$\vec{P}^{(1)}(\vec{r}, t) = \sum_{\ell} \vec{P}_{\ell}^{(1)}(\vec{k}_{\ell}, \omega_{\ell}) = \epsilon_0 \sum_{\ell} \tilde{\chi}^{(1)}(\omega_{\ell}) : \vec{E}_{\ell}(\vec{k}_{\ell}, \omega_{\ell})$$

$$\vec{P}^{NL}(\vec{r}, t) = \sum_{n \geq 2} \vec{P}^{(n)}(\vec{r}, t) = \sum_m \vec{P}^{NL}(\vec{k}_m, \omega_m)$$

$$\nabla^2 \vec{E} + \frac{\omega^2}{c^2} \vec{\epsilon} : \vec{E}(\vec{k}, \omega) = -\mu_0 \omega^2 \vec{P}^{NL}(\vec{k}_m, \omega_m = \omega)$$

$$\left(\nabla^2 + \frac{\omega_1^2}{c^2} \vec{\epsilon}_1 : \right) \vec{E}_1(\vec{k}_1, \omega_1) = -\mu_0 \omega_1^2 \vec{P}^{NL}(\omega_1)$$

$$= -\mu_0 \epsilon_0 \omega_1^2 \tilde{\chi}^{(2)}(\omega_1 = \omega - \omega_2) \vec{E}_2^*(\vec{k}_2, \omega_2) \vec{E}(\vec{k}, \omega)$$

$$\left(\nabla^2 + \frac{\omega_2^2}{c^2} \vec{\epsilon}_2 : \right) \vec{E}_2(\vec{k}_2, \omega_2) = -\mu_0 \omega_2^2 \vec{P}^{NL}(\omega_2)$$

$$= -\mu_0 \epsilon_0 \omega_2^2 \tilde{\chi}^{(2)}(\omega_2 = \omega - \omega_1) \vec{E}_1^*(\vec{k}_1, \omega_1) \vec{E}(\vec{k}, \omega)$$

$$\left(\nabla^2 + \frac{\omega^2}{c^2} \vec{\epsilon} : \right) \vec{E}(\vec{k}, \omega) = -\mu_0 \omega^2 \vec{P}^{NL}(\omega)$$

$$= -\mu_0 \epsilon_0 \omega^2 \tilde{\chi}^{(2)}(\omega = \omega_1 + \omega_2) \vec{E}_1(\vec{k}_1, \omega_1) \vec{E}_2(\vec{k}_2, \omega_2)$$



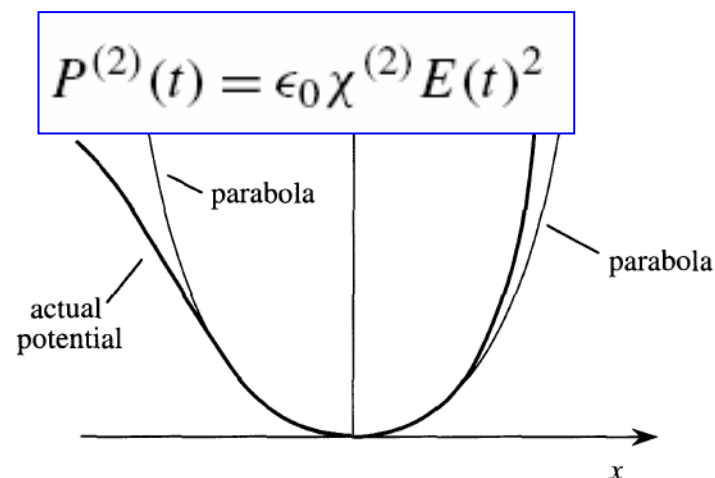
1.4) Susceptibilidade de um oscilador não harmônico

Meio não centro-simétrico

$$m \frac{d^2x}{dt^2} + 2m\gamma \frac{dx}{dt} + Kx + max^2 = -eE$$

$$U = - \int F_{\text{restoring}} dx = \frac{1}{2} m \omega_0^2 x^2 + \frac{1}{3} max^3$$

$$E(t) = E_1 \exp(-i\omega_1 t) + E_2 \exp(-i\omega_2 t) + \text{c.c.}$$



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

$$x^{(1)} = x^{(1)}(\omega_1) + x^{(1)}(\omega_2)$$

$$x^{(1)}(\omega_i) = \frac{-e/m}{\omega_0^2 - \omega_i^2 - i\omega_i\gamma} E_i = \frac{-e/m}{D(\omega_i)} E_i$$

$$x^{(2)} = x^{(2)}(\omega_1 + \omega_2) + x^{(2)}(\omega_1 - \omega_2) + x^{(2)}(2\omega_1) + x^{(2)}(2\omega_2) + x^{(2)}(0)$$

$$D(\omega_i) = \omega_0^2 - \omega_i^2 - i\omega_i\gamma$$

$$x^{(2)}(2\omega_1) = \frac{-a(e/m)^2}{D^2(\omega_1)D(2\omega_1)} E_1^2 \exp\{-2i\omega_1 t\}$$

$$x^{(2)}(2\omega_2) = \frac{-a(e/m)^2}{D^2(\omega_2)D(2\omega_2)} E_2^2 \exp\{-2i\omega_2 t\}$$

$$x^{(2)}(\omega_1 + \omega_2) = \frac{-2a(e/m)^2}{D(\omega_1)D(\omega_2)D(\omega_1 + \omega_2)} E_1 E_2 \exp\{-i(\omega_1 + \omega_2)t\}$$

$$x^{(2)}(\omega_1 - \omega_2) = \frac{-2a(e/m)^2}{D(\omega_1)D(-\omega_2)D(\omega_1 - \omega_2)} E_1 E_2^* \exp\{-i(\omega_1 - \omega_2)t\}$$

$$x^{(2)}(0) = -2a(e/m)^2 \left[\frac{E_1 E_1^*}{D(0)D(\omega_1)D(-\omega_1)} + \frac{E_2 E_2^*}{D(0)D(\omega_2)D(-\omega_2)} \right]$$

$$P^{(1)}(\omega_i) = \epsilon_0 \chi^{(1)}(\omega_i) E(\omega_i) = -N e x^{(1)}(\omega_i) \Rightarrow \chi^{(1)}(\omega_i) = \frac{N(e^2/m\epsilon_0)}{D(\omega_i)}$$

$$P^{(2)}(2\omega_i) = \epsilon_0 \chi^{(2)}(2\omega_i; \omega_i, \omega_i) E^2(\omega_i) = -N e x^{(2)}(2\omega_i) \Rightarrow \chi^{(2)}(2\omega_i; \omega_i, \omega_i) = \frac{N a(e^3/m^2\epsilon_0)}{D(2\omega_i)D^2(\omega_i)}$$

$$\Rightarrow \chi^{(2)}(2\omega_i; \omega_i, \omega_i) = \frac{m(\epsilon_0)^2 a}{N^2 e^3} [\chi^{(1)}(2\omega_i)] [\chi^{(1)}(\omega_i)]^2$$

$$\chi^{(2)}(\omega_1 + \omega_2; \omega_1, \omega_2) = \frac{N a(e^3/m^2\epsilon_0)}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)} = \frac{m(\epsilon_0)^2 a}{N^2 e^3} [\chi^{(1)}(\omega_1 + \omega_2)] [\chi^{(1)}(\omega_1)] [\chi^{(1)}(\omega_2)]$$

$$\chi^{(2)}(\omega_1 - \omega_2; \omega_1, -\omega_2) = \frac{N a(e^3/m^2\epsilon_0)}{D(\omega_1)D(-\omega_2)D(\omega_1 - \omega_2)} = \frac{m(\epsilon_0)^2 a}{N^2 e^3} [\chi^{(1)}(\omega_1 - \omega_2)] [\chi^{(1)}(\omega_1)] [\chi^{(1)}(-\omega_2)]$$

$$\chi^{(2)}(0; \omega_1, -\omega_1) = \frac{N a(e^3/m^2\epsilon_0)}{D(0)D(\omega_1)D(-\omega_1)} = \frac{m(\epsilon_0)^2 a}{N^2 e^3} [\chi^{(1)}(0)] [\chi^{(1)}(\omega_1)] [\chi^{(1)}(-\omega_1)]$$



1.5) Propriedades da susceptibilidade não linear

Consideremos a mistura de ondas ω_1 , ω_2 e $\omega_3 = \omega_1 + \omega_2$

$$P_i^{(2)}(\omega_n + \omega_m) = \epsilon_0 \sum_{j,k} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m)$$

$$\chi_{ijk}^{(2)}(\omega_1, \omega_3, -\omega_2), \quad \chi_{ijk}^{(2)}(\omega_1, -\omega_2, \omega_3), \quad \chi_{ijk}^{(2)}(\omega_2, \omega_3, -\omega_1),$$

$$\chi_{ijk}^{(2)}(\omega_2, -\omega_1, \omega_3), \quad \chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2), \quad \text{and} \quad \chi_{ijk}^{(2)}(\omega_3, \omega_2, \omega_1)$$

$(6+6)*27 = 324$ componentes tensoriais! Mas existem restrições.....

Número de valores independentes de $\chi_{ijk}^{(2)}$

324 possíveis

Realidade do campo

$324/2 \Rightarrow 162$ possíveis

$$\chi_{ijk}^{(2)}(-\omega_n - \omega_m, -\omega_n, -\omega_m) = \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)^*$$

Permutação intrínseca

$$P_i^{(2)}(\omega_n + \omega_m) = \varepsilon_0 \sum_{j,k} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m)$$

$$\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) = \chi_{ikj}^{(2)}(\omega_n + \omega_m, \omega_m, \omega_n)$$

81 possíveis

Meios sem perda

$\chi^{(2)}$ é real pois γ é nulo

$$\chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{jki}^{(2)}(-\omega_1 = \omega_2 - \omega_3) = \chi_{jki}^{(2)}(\omega_1 = -\omega_2 + \omega_3)^*$$

27 possíveis

Simetria de Kleinman

Se $\omega_{m,n} \ll \omega_0$, χ independe de ω

18 possíveis

GSH, notação contraída

$$d_{ijk} = \frac{\epsilon_0}{2} \chi_{ijk}^{(2)}$$

$$P_i(\omega_n + \omega_m) = \sum_{jk} \sum_{(nm)} 2d_{ijk} E_j(\omega_n) E_k(\omega_m)$$

10 possíveis

Simetria cristalina

<10 possíveis

Aula 2 - Equação de ondas não linear



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IFSC - USP

2.1) Equação de ondas não linear

Equações de Maxwell

$$\vec{\nabla} \cdot \vec{D} = \rho$$

$$\vec{\nabla} \cdot \vec{B} = 0$$

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}$$

$$\vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t}$$

$$\vec{B} = \mu_0 \vec{H}$$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$$

$$\epsilon_0 = 8.854 \times 10^{-12} \text{ F/m} \text{ e } \mu_0 = 4\pi \times 10^{-7} \text{ H/m}$$

$$\vec{\nabla} \times (\vec{\nabla} \times \vec{E}) = -\vec{\nabla} \times \left(\frac{\partial \vec{B}}{\partial t} \right) = -\frac{\partial}{\partial t} (\vec{\nabla} \times \vec{B}) = -\mu_0 \frac{\partial}{\partial t} (\vec{\nabla} \times \vec{H})$$

$$\vec{\nabla}_x (\vec{\nabla}_x \vec{E}) = \vec{\nabla} (\vec{\nabla} \cdot \vec{E}) - \nabla^2 \vec{E}$$

$$\nabla^2 \vec{E} = \mu_0 \frac{\partial^2}{\partial t^2} \vec{D} = \mu_0 \frac{\partial^2 (\epsilon_0 \vec{E} + \vec{P})}{\partial t^2}$$

$$\nabla^2 \vec{E} - \mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

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

$$\nabla^2 \vec{E} - \mu_0 \epsilon_0 (1 + \tilde{\chi}^{(1)}) : \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}^{NL}}{\partial t^2}$$

$$\vec{E}(\vec{r}, t) = \sum_{\ell} \vec{E}_{\ell}(\vec{k}_{\ell}, \omega_{\ell}) = \sum_{\ell} \vec{A}_{\ell}(\vec{r}, \omega) \exp\{i(\vec{k}_{\ell} \cdot \vec{r} - \omega_{\ell} t)\}$$

$$\vec{P}^{(1)}(\vec{r}, t) = \sum_{\ell} \vec{P}_{\ell}^{(1)}(\vec{k}_{\ell}, \omega_{\ell}) = \epsilon_0 \sum_{\ell} \tilde{\chi}^{(1)}(\omega_{\ell}) : \vec{E}_{\ell}(\vec{k}_{\ell}, \omega_{\ell})$$

$$\vec{P}^{NL}(\vec{r}, t) = \sum_{n \geq 2} \vec{P}^{(n)}(\vec{r}, t) = \sum_m \vec{P}^{NL}(\vec{k}_m, \omega_m)$$

$$\nabla^2 \vec{E} + \frac{\omega^2}{c^2} \vec{\epsilon} : \vec{E}(\vec{k}, \omega) = -\mu_0 \omega^2 \vec{P}^{NL}(\vec{k}_m, \omega_m = \omega)$$

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

Constante dieléctrica

2.2) Equações para a soma de frequências

$$\left(\nabla^2 + \frac{\omega_1^2}{c^2} \vec{\epsilon}_1 : \right) \vec{E}_1(\vec{k}_1, \omega_1) = -\mu_0 \omega_1^2 \vec{P}^{NL}(\omega_1)$$

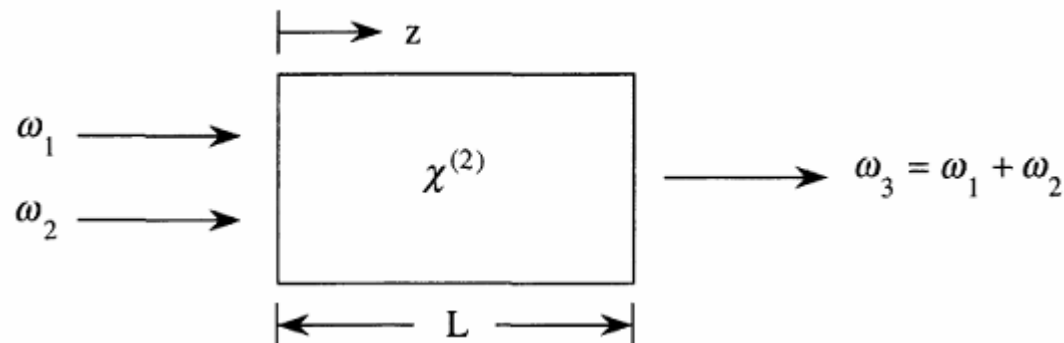
$$= -\mu_0 \epsilon_0 \omega_1^2 \tilde{\chi}^{(2)}(\omega_1 = \omega - \omega_2) \vec{E}_2^*(\vec{k}_2, \omega_2) \vec{E}(\vec{k}, \omega)$$

$$\left(\nabla^2 + \frac{\omega_2^2}{c^2} \vec{\epsilon}_2 : \right) \vec{E}_2(\vec{k}_2, \omega_2) = -\mu_0 \omega_2^2 \vec{P}^{NL}(\omega_2)$$

$$= -\mu_0 \epsilon_0 \omega_2^2 \tilde{\chi}^{(2)}(\omega_2 = \omega - \omega_1) \vec{E}_1^*(\vec{k}_1, \omega_1) \vec{E}(\vec{k}, \omega)$$

$$\left(\nabla^2 + \frac{\omega^2}{c^2} \vec{\epsilon} : \right) \vec{E}(\vec{k}, \omega) = -\mu_0 \omega^2 \vec{P}^{NL}(\omega)$$

$$= -\mu_0 \epsilon_0 \omega^2 \tilde{\chi}^{(2)}(\omega = \omega_1 + \omega_2) \vec{E}_1(\vec{k}_1, \omega_1) \vec{E}_2(\vec{k}_2, \omega_2)$$



SVEA

$$\nabla^2 \vec{E} + \frac{\omega^2}{c^2} \vec{\epsilon}^{(1)} : \vec{E}(\vec{k}, \omega) = -\mu_0 \omega^2 \vec{P}^{NL}(\vec{k}_m, \omega_m = \omega)$$

Caso escalar unidimensional: $E(\omega, z) = A(\omega, z) \exp\{i(kz - \omega t)\}$

$$\frac{d^2 E}{dz^2} = \frac{d}{dz} \left[\left(\frac{dA}{dz} + ikA \right) \exp\{i(kz - \omega t)\} \right] = \left(\frac{d^2 A}{dz^2} + 2ik \frac{dA}{dz} - k^2 A \right) \exp\{i(kz - \omega t)\}$$

$$\frac{d^2 A}{dz^2} \ll k \frac{dA}{dz} \Rightarrow$$

$$\frac{d^2 E}{dz^2} + \frac{\omega^2 n^2}{c^2} E = 2ik \frac{dA}{dz} \exp\{i(kz - \omega t)\} = -\mu_0 \omega^2 \vec{P}^{NL}(\vec{k}_m, \omega_m = \omega)$$

$$\frac{d^2 E}{dz^2} + \frac{\omega^2 n^2}{c^2} E = 2ik \frac{dA}{dz} \exp\{i(kz - \omega t)\} = -\mu_0 \omega^2 P^{NL}(\vec{k}_m, \omega_m = \omega)$$

$$\frac{dA(\omega, z)}{dz} = \frac{i\mu_0 \omega^2}{2k} P^{NL}(\omega, z) \exp\{-i(kz - \omega t)\}$$

famosa e popular SVEA

Geração de soma de frequências

$$\frac{dA_3(z)}{dz} = \frac{i\mu_0 \omega_3^2}{2k_3} P^{NL}(\omega_3) \exp\{-i(k_3 z - \omega_3 t)\}$$

$$k_3 = \frac{n_3 \omega_3}{c}$$

$$P^{NL}(\omega_3) = \varepsilon_0 \chi^{(2)}(\omega_3 = \omega_1 + \omega_2) E_1 E_2 + \varepsilon_0 \chi^{(2)}(\omega_3 = \omega_2 + \omega_1) E_2 E_1 = 2\varepsilon_0 \chi^{(2)}(\omega_3) E_1 E_2$$

$$E_i(z, t) = A_i \exp\{i(k_i z - \omega_i t)\} \quad i = 1, 2$$

$$\Delta k = k_1 + k_2 - k_3$$

$$\frac{dA_3(z)}{dz} = \frac{i\mu_0 \varepsilon_0 \omega_3^2}{2k_3} 2\chi^{(2)}(\omega_3) A_1 A_2 \exp\{i\Delta k z\} = \frac{i\omega_3^2}{k_3 c^2} \chi^{(2)}(\omega_3) A_1 A_2 \exp\{i\Delta k z\}$$

Procedendo da mesma forma,

$$\frac{dA_1(z)}{dz} = \frac{i\mu_0\omega_1^2}{2k_1} P^{\text{NL}}(\omega_1) \exp\{-i(k_1z - \omega_1t)\}$$

$$P^{\text{NL}}(\omega_1) = 2\varepsilon_0\chi^{(2)}(\omega_1 = \omega_3 - \omega_2) E_3E_2^*$$

$$\frac{dA_1(z)}{dz} = \frac{i\omega_1^2}{k_1c^2} \chi^{(2)}(\omega_1) A_3A_2^* \exp\{-i\Delta kz\}$$

$$\frac{dA_2(z)}{dz} = \frac{i\omega_2^2}{k_2c^2} \chi^{(2)}(\omega_2) A_3A_1^* \exp\{-i\Delta kz\}$$

Se utilizarmos a conjectura de Kleinman,

$$\chi^{(2)} = \chi^{(2)}(\omega_1) = \chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_3)$$

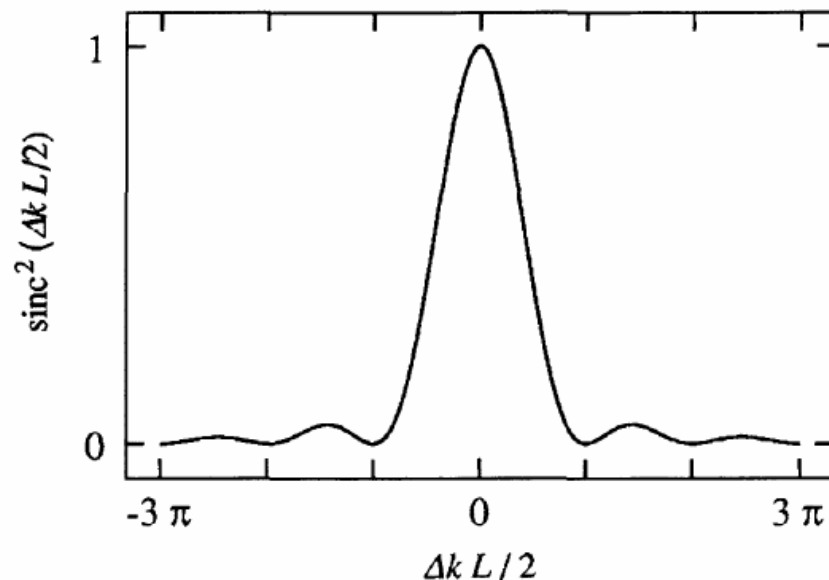
$$\frac{dA_3(z)}{dz} = \frac{i\omega_3^2}{k_3 c^2} \chi^{(2)} A_1 A_2 \exp\{i\Delta k z\}$$

Sem depleção:

$$A_3(z) = \frac{\omega_3}{cn_3} \chi^{(2)} A_1 A_2 \frac{[\exp\{i\Delta k z\} - 1]}{\Delta k}$$

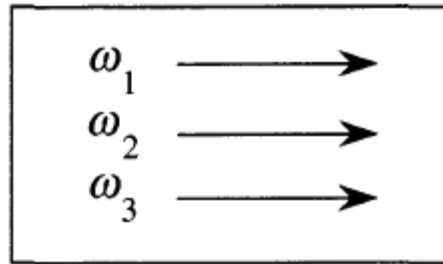
$$I_3(z) = \frac{1}{2} cn_3 \varepsilon_0 |A_3|^2 = \frac{\omega_3^2}{n_1 n_2 n_3 c^3} |\chi^{(2)}|^2 I_1 I_2 \left[\frac{\text{sen}(\Delta k z/2)}{\Delta k z/2} \right]^2 z^2$$

onde $I_1 = \frac{1}{2} cn_1 \varepsilon_0 |A_1|^2,$
 $I_2 = \frac{1}{2} cn_2 \varepsilon_0 |A_2|^2$



2.3) Relações de Manley-Rowe

Vamos considerar 3 ondas: ω_1 , ω_2 e $\omega_3 = \omega_1 + \omega_2$



$$I_i = \frac{1}{2} \text{cn}_i \epsilon_0 A_i A_i^*$$

$$\frac{dI_i}{dz} = \frac{1}{2} \text{cn}_i \epsilon_0 \left(A_i^* \frac{dA_i}{dz} + A_i \frac{dA_i^*}{dz} \right)$$

$$\frac{dA_1(z)}{dz} = \frac{i\omega_1^2}{k_1 c^2} \chi^{(2)}(\omega_1) A_3 A_2^* \exp\{-i\Delta k z\}$$

Considerando a irradiância total:

$$I = I_1 + I_2 + I_3$$

e utilizando a conjectura de Kleinman, $\chi^{(2)} = \chi^{(2)}(\omega_1) = \chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_3)$

$$\frac{dI}{dz} = \frac{dI_1}{dz} + \frac{dI_2}{dz} + \frac{dI_3}{dz} = -\epsilon_0 (\omega_1 + \omega_2 - \omega_3) \chi^{(2)} \text{Im} \left[A_1^* A_3 A_2^* \exp\{-i\Delta k z\} \right] = 0$$

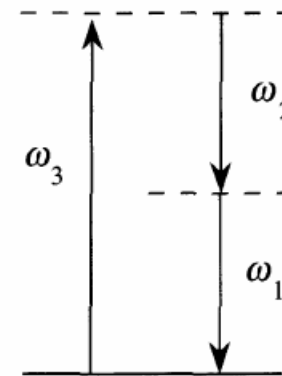
Além disso,

$$\frac{d}{dz} \left(\frac{I_1}{\omega_1} \right) = \frac{d}{dz} \left(\frac{I_2}{\omega_2} \right) = - \frac{d}{dz} \left(\frac{I_3}{\omega_3} \right)$$

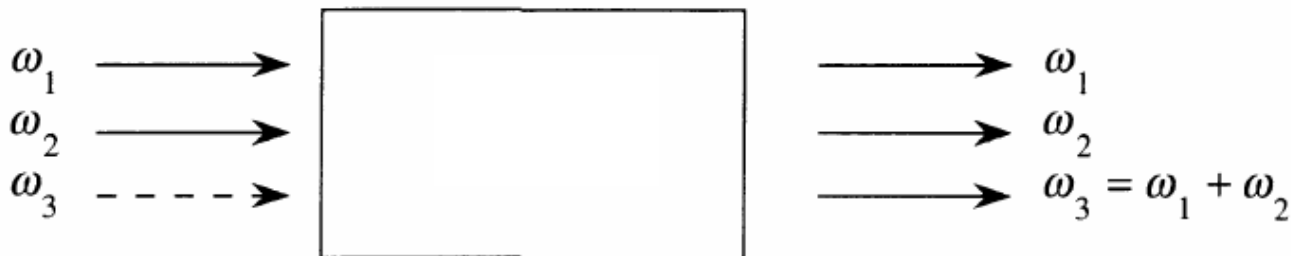
Relações de
Manley-Rowe

$I/\omega = n^0$ de fótons/área/tempo

$$\frac{d}{dz} \left(\frac{I_1}{\omega_1} \right) = \frac{d}{dz} \left(\frac{I_2}{\omega_2} \right) = - \frac{d}{dz} \left(\frac{I_3}{\omega_3} \right)$$



2.4) Geração de soma de frequências



ω_2 muito forte, ω_1 fraco

$$\frac{dA_1}{dz} = \frac{i\omega_1^2}{k_1 c^2} \chi^{(2)} A_3 A_2^* \exp\{-i\Delta k z\} = K_1 A_3 \exp\{-i\Delta k z\}$$

$$K_1 = \frac{i\omega_1^2}{k_1 c^2} \chi^{(2)} A_2^*$$

$$\frac{dA_3}{dz} = \frac{i\omega_3^2}{k_3 c^2} \chi^{(2)} A_2 A_1 \exp\{i\Delta k z\} = K_3 A_1 \exp\{i\Delta k z\}$$

$$K_3 = \frac{i\omega_3^2}{k_3 c^2} \chi^{(2)} A_2$$

Considerando inicialmente o caso em que $\Delta k=0$ (phase matching)

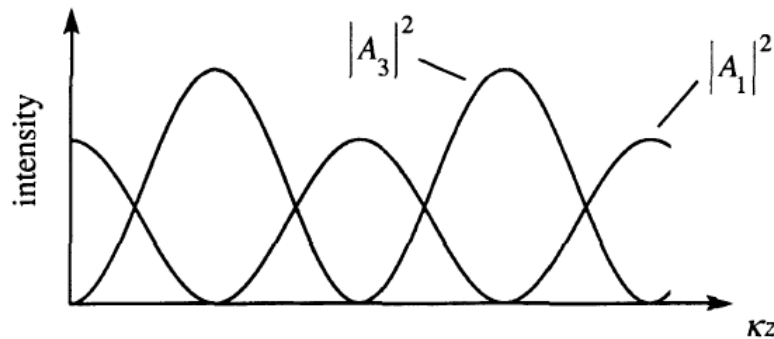
$$\frac{d^2 A_1}{dz^2} = K_1 \frac{dA_3}{dz} = K_1 K_3 A_1 = -\kappa^2 A_1$$

$$\kappa^2 = \frac{\omega_3^2 \omega_1^2}{k_3 k_1 c^4} |\chi^{(2)}|^2 |A_2|^2$$

que representa a equação de um oscilador harmônico.

Usando a condição inicial $A_3(0)=0$ e $A_1(0)$ dado, obtemos:

$$A_1(z) = A_1(0) \cos \kappa z \qquad A_3(z) = -A_1(0) \frac{\kappa}{K_1} \sin \kappa z$$



$$\frac{dA_1}{dz} = K_1 A_3 \exp\{-i\Delta kz\}$$

$$\frac{dA_3}{dz} = K_3 A_1 \exp\{i\Delta kz\}$$

Consideremos a seguir o caso em que $\Delta k \neq 0$ (sem phase matching) e tomemos como soluções tentativa:

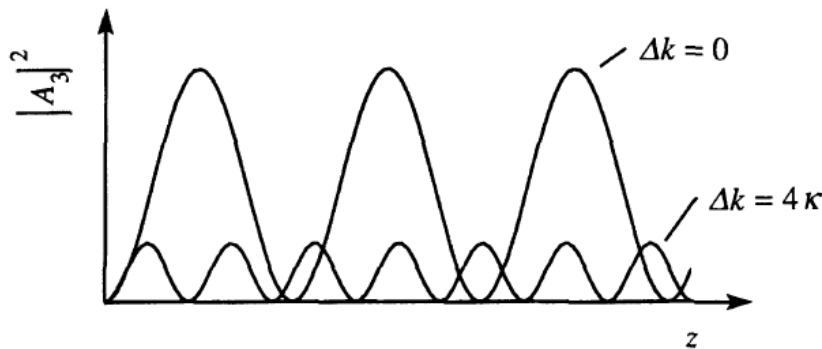
$$A_1(z) = (Fe^{igz} + Ge^{-igz})e^{-i\Delta kz/2} \quad \text{e} \quad A_3(z) = (Ce^{igz} + De^{-igz})e^{i\Delta kz/2}$$

$$g = \sqrt{\kappa^2 + \frac{1}{4}\Delta k^2}$$

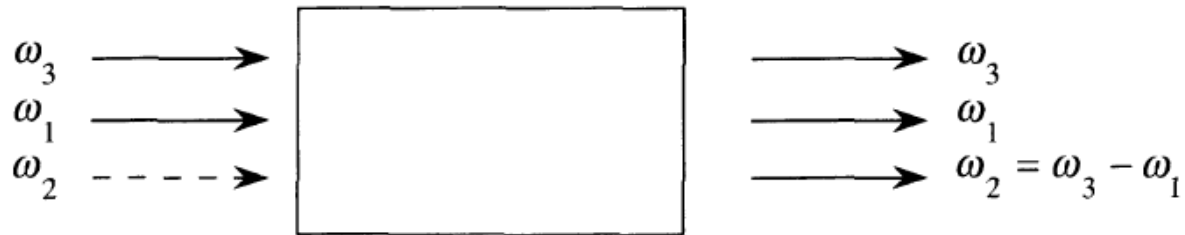
Usando a condição inicial $A_3(0)=0$ e $A_1(0)$ dado, obtemos:

$$A_1(z) = \left[A_1(0) \cos gz + \left(\frac{K_1}{g} A_3(0) + \frac{i \Delta k}{2g} A_1(0) \right) \sin gz \right] e^{-(1/2)i \Delta k z}$$

$$A_3(z) = \frac{K_3}{g} A_1(0) \sin gz e^{(1/2)i \Delta k z} \quad |A_3(z)|^2 = |A_1(0)|^2 \frac{|K_3|^2}{g^2} \sin^2 gz$$



2.5) Geração de diferença de frequências e amplificação paramétrica



ω_3 muito forte, ω_2 nulo em $z = 0$

$$\frac{dA_1}{dz} = \frac{i\omega_1^2}{k_1 c^2} \chi^{(2)} A_3 A_2^* \exp\{i\Delta k z\}$$

$$\frac{dA_2}{dz} = \frac{i\omega_2^2}{k_2 c^2} \chi^{(2)} A_3 A_1^* \exp\{i\Delta k z\}$$

$$\Delta k = k_3 - k_1 - k_2$$

Considerando inicialmente o caso em que $\Delta k=0$ (phase matching)

$$\frac{d^2 A_2}{dz^2} = \frac{i\omega_2^2}{k_2 c^2} \chi^{(2)} A_3 \frac{dA_1^*}{dz}$$

$$\frac{dA_1^*}{dz} = \frac{-i\omega_1^2}{k_1 c^2} \chi^{(2)} A_3^* A_2$$

$$\frac{d^2 A_2}{dz^2} = \frac{\omega_2^2 \omega_1^2}{k_2 k_1 c^4} |\chi^{(2)}|^2 A_3 A_3^* A_2 = \kappa^2 A_2$$

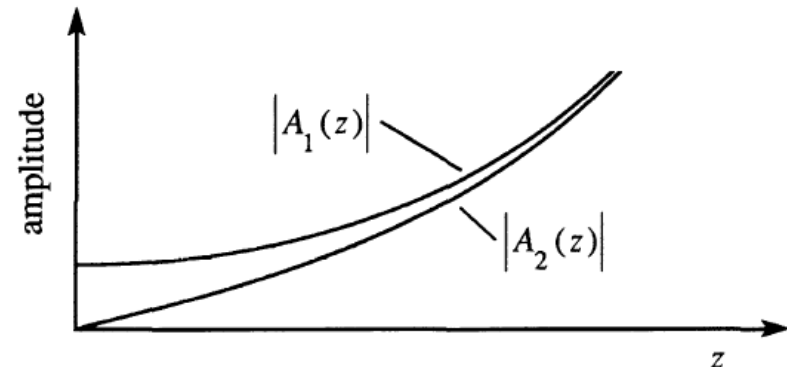
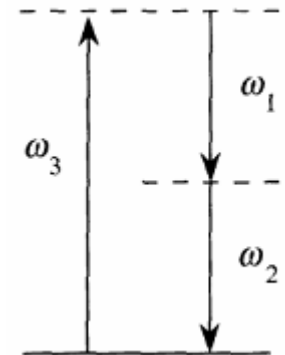
$$\kappa^2 = \frac{\omega_2^2 \omega_1^2}{k_2 k_1 c^4} |\chi^{(2)}|^2 |A_3|^2$$

$$A_2(z) = C \sinh \kappa z + D \cosh \kappa z$$

Usando a condição inicial $A_2(0)=0$ e $A_1(0)$ arbitrário, obtemos:

$$A_1(z) = A_1(0) \cosh \kappa z$$

$$A_2(z) = i \left(\frac{n_1 \omega_2}{n_2 \omega_1} \right)^{1/2} \frac{A_3}{|A_3|} A_1^*(0) \sinh \kappa z$$



Considerando o caso em que $\Delta k \neq 0$ (sem phase matching)

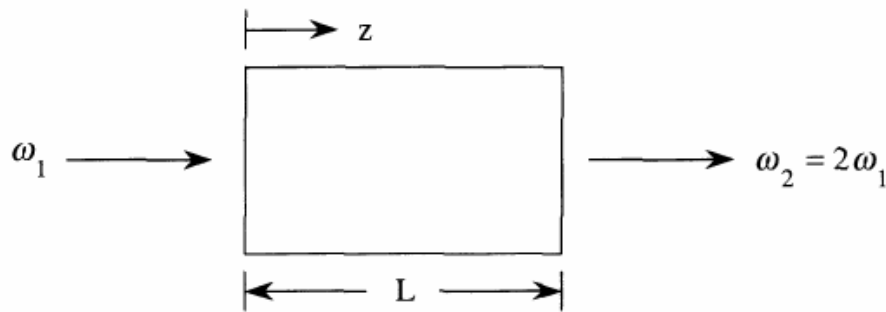
$$A_1(z) = \left[A_1(0) \left(\cosh gz - \frac{i \Delta k}{2g} \sinh gz \right) + \frac{\kappa_1}{g} A_2^*(0) \sinh gz \right] e^{i \Delta k z / 2}$$

$$A_2(z) = \left[A_2(0) \left(\cosh gz - \frac{i \Delta k}{2g} \sinh gz \right) + \frac{\kappa_2}{g} A_1^*(0) \sinh gz \right] e^{i \Delta k z / 2}$$

$$g = \left[\kappa_1 \kappa_2^* - \left(\frac{\Delta k}{2} \right)^2 \right]^{1/2}$$

$$\kappa_j = \frac{i \omega_j^2}{k_j c^2} \chi^{(2)} A_j$$

2.6) Geração de segundo harmônico



$$k_j = n_j \omega_j / c \quad n_j = [\epsilon^{(1)}(\omega_j)]^{1/2}$$

$$E(z, t) = E_1(z, t) + E_2(z, t)$$

$$E_j(z, t) = E_j(z) e^{-i\omega_j t} + \text{c.c.}$$

$$E_j(z) = A_j(z) e^{ik_j z}$$

$$P^{\text{NL}}(z, t) = P_1(z, t) + P_2(z, t)$$

$$P_j(z, t) = P_j(z) e^{-i\omega_j t} + \text{c.c.}, \quad j = 1, 2$$

$$P_1(z) = 2\epsilon_0 \chi^{(2)} E_2(z) E_1^*(z) = 2\epsilon_0 \chi^{(2)} A_2 A_1^* e^{i(k_2 - k_1)z}$$

$$P_2(z) = \epsilon_0 \chi^{(2)} E_1^2(z) = \epsilon_0 \chi^{(2)} A_1^2 e^{2ik_1 z}$$

$$\frac{dA(\omega, z)}{dz} = \frac{i\mu_0\omega^2}{2k} P^{NL}(\omega, z) \exp\{-i(kz - \omega t)\}$$

$$\frac{dA_1}{dz} = \frac{i\mu_0\omega_1^2}{2k_1} 2\varepsilon_0\chi^{(2)} A_2 A_1^* \exp\{i(k_2 - 2k_1)z\} = \frac{i\omega_1}{n_1 c} \chi^{(2)} A_2 A_1^* \exp\{-i\Delta k z\}$$

$$\frac{dA_2}{dz} = \frac{i\mu_0\omega_2^2}{2k_2} \varepsilon_0\chi^{(2)} A_1^2 \exp\{i\Delta k z\} = \frac{i\omega_2}{n_2 c} \chi^{(2)} A_1^2 \exp\{i\Delta k z\} \quad \Delta k = 2k_1 - k_2$$

Como $I_j = (1/2)cn_j\varepsilon_0 |E_j|^2$, definimos:

$$A_1 = \sqrt{\frac{2I}{n_1 c \varepsilon_0}} u_1 e^{i\phi_1} \quad A_2 = \sqrt{\frac{2I}{n_2 c \varepsilon_0}} u_2 e^{i\phi_2} \quad I = I_1 + I_2$$

tal que: $u_1^2(z) + u_2^2(z) = 1$. Substituindo nas equações para A_1 e A_2 ,

$$\frac{du_1}{d\zeta} = u_1 u_2 \sin \theta,$$

$$\frac{du_2}{d\zeta} = -u_1^2 \sin \theta,$$

$$\frac{d\theta}{d\zeta} = \Delta s + \frac{\cos \theta}{\sin \theta} \frac{d}{d\zeta} (\ln u_1^2 u_2)$$

onde: $\zeta = z/l$, $l = \sqrt{\frac{n_1^2 n_2 c^3 \epsilon_0}{2I}} \frac{1}{\omega_1 \chi^{(2)}}$, $\theta = 2\phi_1 - \phi_2 + \Delta k z$ e $\Delta s = \Delta k l$

Considerando o caso em que $\Delta k = \Delta s = 0$ (phase matching)

$$\frac{d\theta}{d\zeta} = \frac{\cos \theta}{\sin \theta} \frac{d}{d\zeta} (\ln u_1^2 u_2) \Rightarrow \frac{d}{d\zeta} \ln (\cos \theta u_1^2 u_2) = 0.$$

$$u_1^2 u_2 \cos \theta = \Gamma$$

$$u_1^2(z) + u_2^2(z) = 1$$

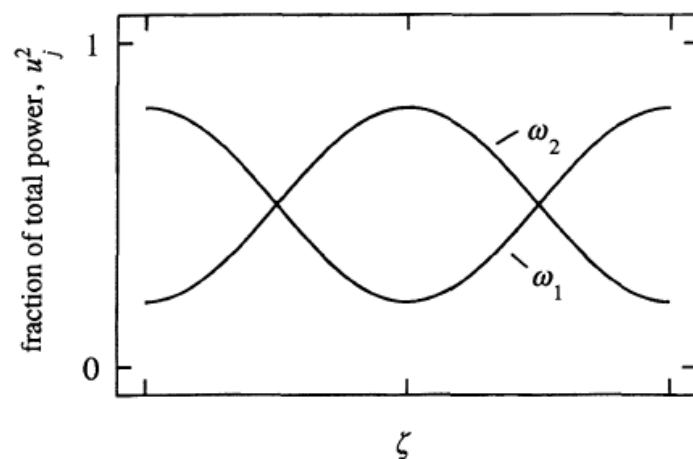
$$\frac{du_2}{d\zeta} = -u_1^2 \sin \theta = \pm(1 - u_2^2)(1 - \cos^2 \theta)^{1/2}$$

$$\frac{du_2}{d\zeta} = \pm(1 - u_2^2) \left(1 - \frac{\Gamma^2}{u_1^4 u_2^2}\right)^{1/2} = \pm(1 - u_2^2) \left(1 - \frac{\Gamma^2}{(1 - u_2^2)^2 u_2^2}\right)^{1/2}$$

$$u_2 \frac{du_2}{d\zeta} = \pm[(1 - u_2^2)^2 u_2^2 - \Gamma^2]^{1/2}$$

$$\frac{du_2^2}{d\zeta} = \pm 2[(1 - u_2^2)^2 u_2^2 - \Gamma^2]^{1/2}$$

Equação padrão –
funções elípticas de Jacobi



$$u_1^2 u_2 \cos \theta = \Gamma$$

Consideremos o caso $\Gamma = 0$, com $\cos \theta = 0$, em particular $\sin \theta = -1$

$$\frac{du_1}{d\zeta} = u_1 u_2 \sin \theta,$$

$$\frac{du_1}{d\zeta} = -u_1 u_2$$

$$\frac{du_2}{d\zeta} = -u_1^2 \sin \theta,$$

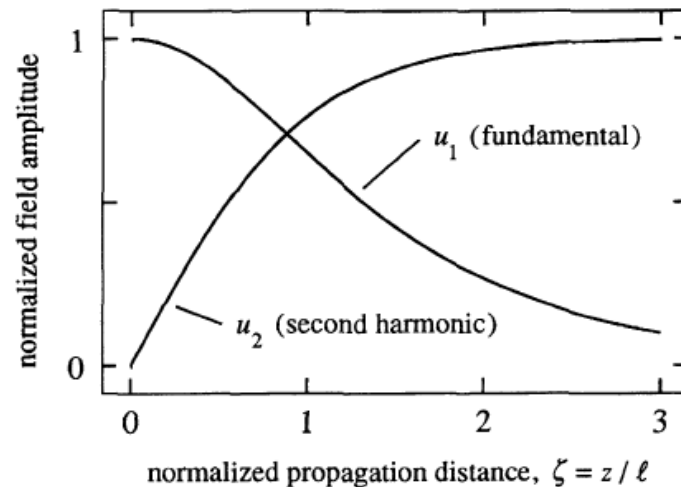
$$\frac{du_2}{d\zeta} = u_1^2 = 1 - u_2^2$$

cuja solução é: $u_2 = \tanh(\zeta + \zeta_0)$. Para $u_1(0) = 1$ e $u_2(0) = 0$,

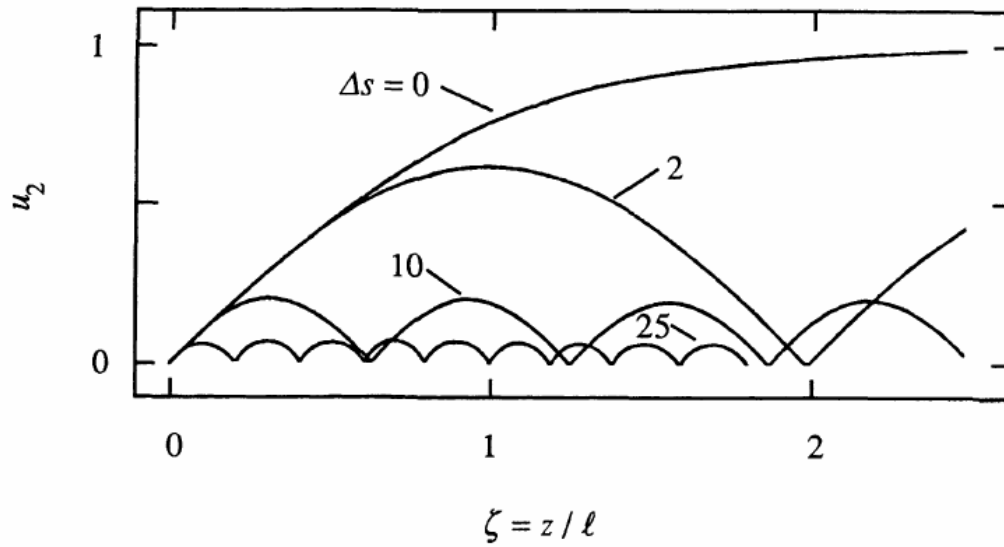
$$u_2(\zeta) = \tanh \zeta$$

$$u_1(\zeta) = \operatorname{sech} \zeta$$

$$l = \sqrt{n_1 n_2} \frac{c}{\omega_1 |A_1(0)| \chi^{(2)}}$$



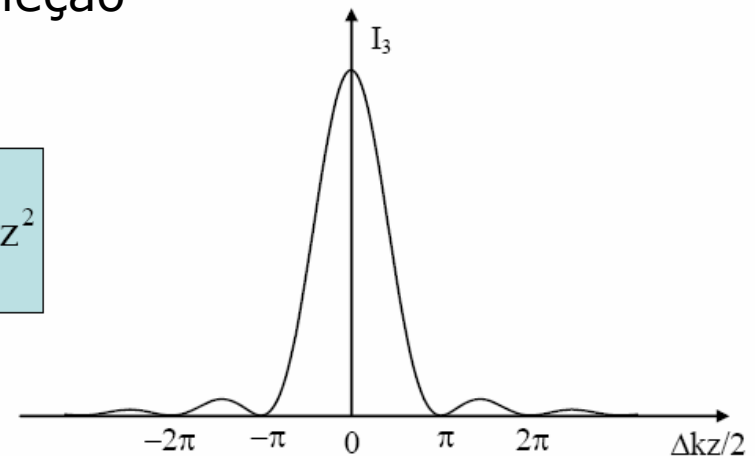
Considerando o caso em que $\Delta k = \Delta s \neq 0$ (sem phase matching)



2.7) Considerações sobre casamento de fase

Geração de segundo harmônico sem depleção

$$I_3(z) = \frac{1}{2} \epsilon_0 n_3 |A_3|^2 = \frac{\omega_3^2}{n_1 n_2 n_3 c^3} |\chi^{(2)}|^2 I_1 I_2 \left[\frac{\sin(\Delta k z / 2)}{\Delta k z / 2} \right]^2 z^2$$

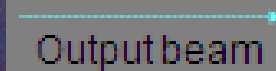
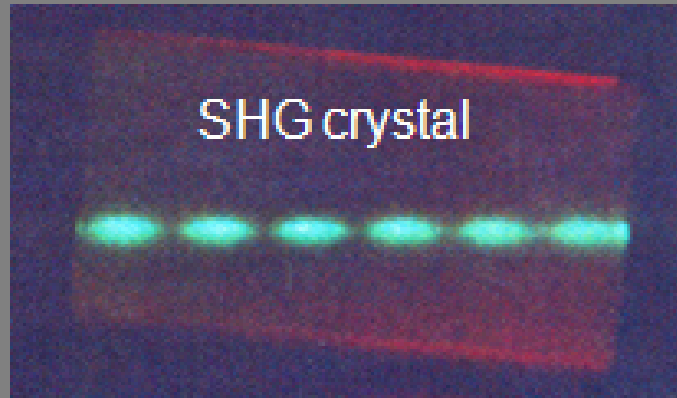
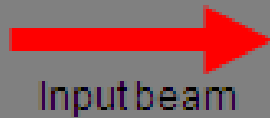


$$[2\omega n(2\omega)/c - 2\omega n(\omega)/c] \ell_c = 2k_0 [n(2\omega) - n(\omega)] \ell_c = 2\pi \quad \ell_c = \frac{\lambda}{2[n(2\omega) - n(\omega)]}$$

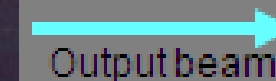
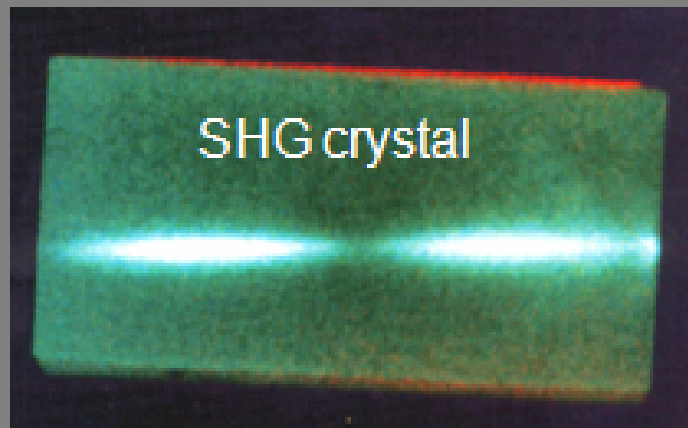
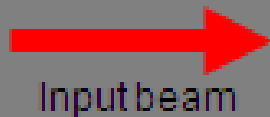
Assim, se a diferença de índices for 0,05, por exemplo, o comprimento de coerência será de apenas $10 \lambda_0$.

Light created in real crystals

Far from
phase-matching:



Closer to
phase-matching:



Note that SH beam is brighter as phase-matching is achieved.

Second-Harmonic Generation

SHG KDP crystals at Lawrence
Livermore National Laboratory

These crystals convert as much
as 80% of the input light to its
second harmonic. Then
additional crystals produce the
third harmonic with similar
efficiency!



TABLE 2.7.1 Linear optical classification of the various crystal systems

System	Linear optical classification
Triclinic, monoclinic, orthorhombic	Biaxial
Trigonal, tetragonal, hexagonal	Uniaxial
Cubic	Isotropic

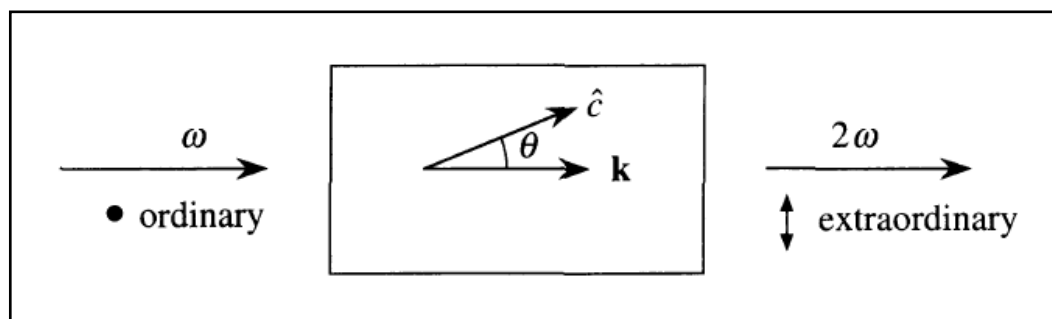
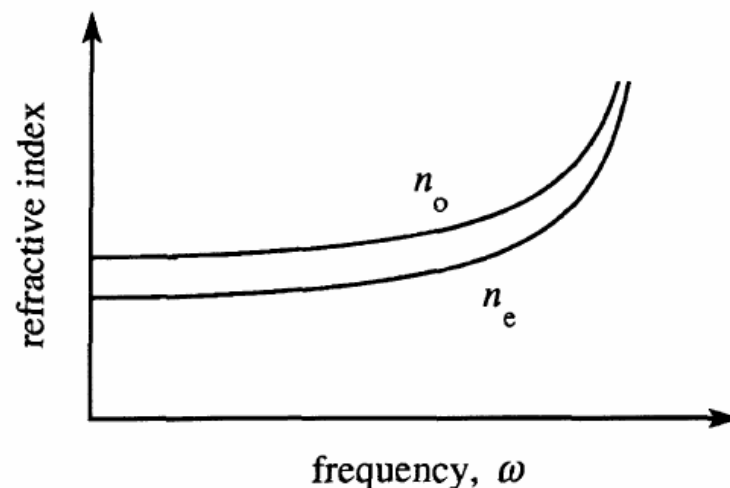
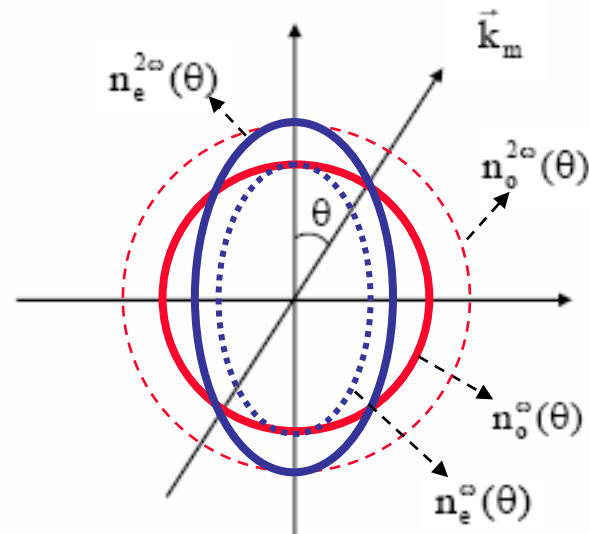
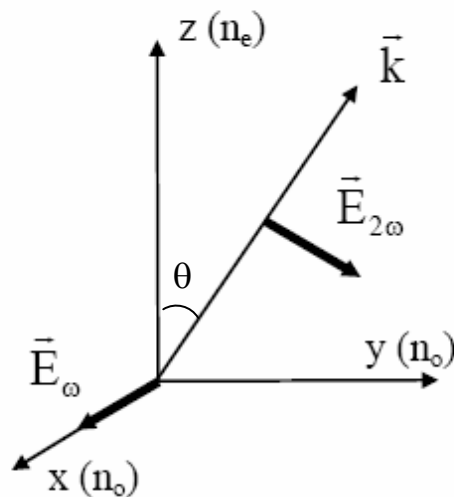


TABLE 2.7.2 Phase-matching methods for uniaxial crystals

	Positive uniaxial ($n_e > n_o$)	Negative uniaxial ($n_e < n_o$)
Type I	$n_3^o \omega_3 = n_1^e \omega_1 + n_2^e \omega_2$	$n_3^e \omega_3 = n_1^o \omega_1 + n_2^o \omega_2$
Type II	$n_3^o \omega_3 = n_1^o \omega_1 + n_2^e \omega_2$	$n_3^e \omega_3 = n_1^e \omega_1 + n_2^o \omega_2$



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

$$\sin^2 \theta_m = \frac{[n_o^\omega]^{-2} - [n_o^{2\omega}]^2}{[n_e^\omega]^{-2} - [n_o^{2\omega}]^2}$$

Para o caso de um cristal de KDP (KH_2PO_4) temos $n_e^\omega = 1.466$, $n_e^{2\omega} = 1.487$, $n_o^\omega = 1.506$ e $n_o^{2\omega} = 1.534$, para $\lambda = 6943 \text{ \AA}$, que é o comprimento de onda de operação de um laser de rubi. Com estes valores obtemos $\theta_m = 50.4^\circ$.

O casamento de fases onde $n(2\omega) = n_o(\omega)$ é chamado do tipo I. Existe ainda o tipo II, onde dois feixes fundamentais tem polarizações ortogonais tal que $n(2\omega) = 1/2 [n_o(\omega) + n_e(\omega)]$.

Aula 3 – Cálculo semi-clássico da susceptibilidade não linear



Sérgio Carlos Zilio
IFSC - USP

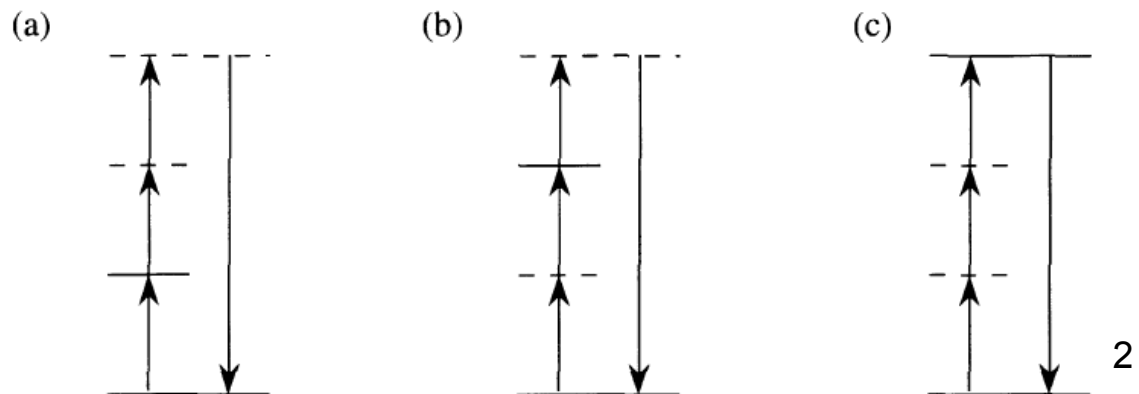
3.1) Introdução

Para que se incomodar em calcular χ ?

- a) As expressões obtidas mostram a dependência de $\chi^{(n)}$ com os parâmetros do material (momento de dipolo de transição e níveis de energia)
- b) Mostram também as simetrias internas de $\chi^{(n)}$
- c) Permitem a obtenção de valores numéricos de $\chi^{(n)}$

Mostram ainda o engrandecimento por ressonância porque aparecem os níveis reais envolvidos no processo

Estratégia para o
enhancement do THG



3.2) Cálculo da susceptibilidade não linear com a equação de Schrödinger

$$i\hbar \frac{\partial \psi}{\partial t} = \hat{H} \psi.$$

$$\hat{H} = \hat{H}_0 + \hat{V}(t)$$

$$\hat{V}(t) = -\hat{\mu} \cdot \tilde{\mathbf{E}}(t) \quad \hat{\mu} = -e\hat{\mathbf{r}}$$

$$\tilde{\mathbf{E}}(t) = \sum_p \mathbf{E}(\omega_p) e^{-i\omega_p t}$$

Átomo livre

$$\psi_n(\mathbf{r}, t) = u_n(\mathbf{r}) e^{-i\omega_n t}$$

$$\hat{H}_0 u_n(\mathbf{r}) = E_n u_n(\mathbf{r}) \quad E_n = \hbar\omega_n$$

$$\int u_m^* u_n d^3r = \delta_{mn}$$

Método perturbativo

$$\hat{H} = \hat{H}_0 + \lambda \hat{V}(t) \quad \text{com } 0 \leq \lambda \leq 1$$

$$\psi(\mathbf{r}, t) = \psi^{(0)}(\mathbf{r}, t) + \lambda \psi^{(1)}(\mathbf{r}, t) + \lambda^2 \psi^{(2)}(\mathbf{r}, t) + \dots$$

Substituindo na equação de Schrödinger e coletando termos com a mesma dependência em λ :

$$i\hbar \frac{\partial \psi^{(0)}}{\partial t} = \hat{H}_0 \psi^{(0)} \quad \text{equação para o átomo livre}$$

$$i\hbar \frac{\partial \psi^{(N)}}{\partial t} = \hat{H}_0 \psi^{(N)} + \hat{V} \psi^{(N-1)}, \quad N = 1, 2, 3, \dots$$

$$\text{Condição inicial:} \quad \psi^{(0)}(\mathbf{r}, t) = u_g(\mathbf{r}) e^{-iE_g t/\hbar}$$

$$\text{Combinação linear:} \quad \psi^{(N)}(\mathbf{r}, t) = \sum_l a_l^{(N)}(t) u_l(\mathbf{r}) e^{-i\omega_l t}$$

Substituindo, $i\hbar \sum_l \dot{a}_l^{(N)} u_l(\mathbf{r}) e^{-i\omega_l t} = \sum_l a_l^{(N-1)} \hat{V} u_l(\mathbf{r}) e^{-i\omega_l t}$

Usando a ortonormalidade, $\dot{a}_m^{(N)} = (i\hbar)^{-1} \sum_l a_l^{(N-1)} V_{ml} e^{i\omega_{ml} t}$

onde: $\omega_{ml} \equiv \omega_m - \omega_l$ $V_{ml} \equiv \langle u_m | \hat{V} | u_l \rangle = \int u_m^* \hat{V} u_l d^3 r$

$$a_m^{(N)}(t) = (i\hbar)^{-1} \sum_l \int_{-\infty}^t dt' V_{ml}(t') a_l^{(N-1)}(t') e^{i\omega_{ml} t'}$$

Amplitude de primeira ordem: $a_l^{(0)} = \delta_{lg}$

$$V_{ml}(t') = - \sum_p \bar{\mu}_{ml} \cdot \bar{\mathbf{E}}(\omega_p) \exp\{-i\omega_p t'\} \quad \mu_{ml} = \int u_m^* \hat{\mu} u_l d^3 r$$

$$a_m^{(1)}(t) = \frac{1}{\hbar} \sum_p \frac{\mu_{mg} \cdot \mathbf{E}(\omega_p)}{\omega_{mg} - \omega_p} e^{i(\omega_{mg} - \omega_p)t}$$

Amplitude de segunda ordem:

$$a_n^{(2)}(t) = \frac{1}{\hbar^2} \sum_{pq} \sum_m \frac{[\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_q)][\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{ng} - \omega_p - \omega_q)(\omega_{mg} - \omega_p)} e^{i(\omega_{ng} - \omega_p - \omega_q)t}$$

Amplitude de terceira ordem:

$$a_v^{(3)}(t) = \frac{1}{\hbar^3} \sum_{pqr} \sum_{mn} \frac{[\boldsymbol{\mu}_{vn} \cdot \mathbf{E}(\omega_r)][\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_q)][\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{vg} - \omega_p - \omega_q - \omega_r)(\omega_{ng} - \omega_p - \omega_q)(\omega_{mg} - \omega_p)} \times e^{i(\omega_{vg} - \omega_p - \omega_q - \omega_r)t}$$

Susceptibilidade linear:

$$\langle \mathbf{p} \rangle = \langle \psi | \hat{\boldsymbol{\mu}} | \psi \rangle \quad \langle \mathbf{p}^{(1)} \rangle = \langle \psi^{(0)} | \hat{\boldsymbol{\mu}} | \psi^{(1)} \rangle + \langle \psi^{(1)} | \hat{\boldsymbol{\mu}} | \psi^{(0)} \rangle$$

$$\psi^{(0)}(\mathbf{r}, t) = u_g(\mathbf{r}) e^{-iE_g t/\hbar}$$

$$\psi^{(1)}(\mathbf{r}, t) = \sum_l a_l^{(1)}(t) u_l(\mathbf{r}) e^{-i\omega_l t}$$

$$a_m^{(1)}(t) = \frac{1}{\hbar} \sum_p \frac{\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)}{\omega_{mg} - \omega_p} e^{i(\omega_{mg} - \omega_p)t}$$

$$\langle \mathbf{p}^{(1)} \rangle = \frac{1}{\hbar} \sum_p \sum_m \left(\frac{\mu_{gm} [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{\omega_{mg} - \omega_p} e^{-i\omega_p t} + \frac{[\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]^* \mu_{mg}}{\omega_{mg}^* - \omega_p} e^{i\omega_p t} \right)$$

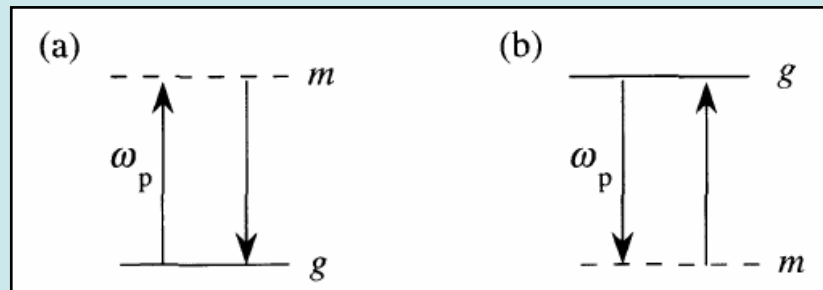
$$\omega_{mg} = \omega_{mg}^0 - i\Gamma_m/2$$

$$\langle \mathbf{p}^{(1)} \rangle = \frac{1}{\hbar} \sum_p \sum_m \left(\frac{\mu_{gm} [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{\omega_{mg} - \omega_p} + \frac{[\boldsymbol{\mu}_{gm} \cdot \mathbf{E}(\omega_p)] \mu_{mg}}{\omega_{mg}^* + \omega_p} \right) e^{-i\omega_p t}$$

$$\mathbf{P}^{(1)} = N \langle \mathbf{p}^{(1)} \rangle \quad \mathbf{P}^{(1)} = \sum_p \mathbf{P}^{(1)}(\omega_p) \exp(-i\omega_p t)$$

$$P_i^{(1)}(\omega_p) = \varepsilon_0 \sum_j \chi_{ij}^{(1)} E_j(\omega_p)$$

$$\varepsilon_0 \chi_{ij}^{(1)}(\omega_p) = \frac{N}{\hbar} \sum_m \left(\frac{\mu_{gm}^i \mu_{mg}^j}{\omega_{mg} - \omega_p} + \frac{\mu_{gm}^j \mu_{mg}^i}{\omega_{mg}^* + \omega_p} \right)$$



Susceptibilidade de segunda ordem:

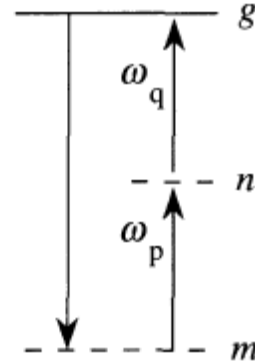
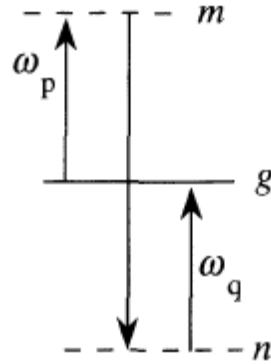
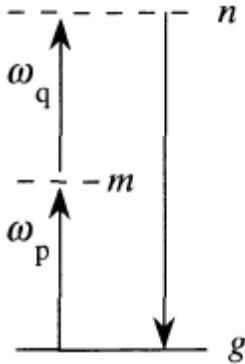
$$\langle \mathbf{p}^{(2)} \rangle = \langle \psi^{(0)} | \hat{\boldsymbol{\mu}} | \psi^{(2)} \rangle + \langle \psi^{(1)} | \hat{\boldsymbol{\mu}} | \psi^{(1)} \rangle + \langle \psi^{(2)} | \hat{\boldsymbol{\mu}} | \psi^{(0)} \rangle$$

$$\begin{aligned} \langle \mathbf{p}^{(2)} \rangle = & \frac{1}{\hbar^2} \sum_{pq} \sum_{mn} \left(\frac{\boldsymbol{\mu}_{gn} [\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_q)] [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{ng} - \omega_p - \omega_q)(\omega_{mg} - \omega_p)} \right. \\ & + \frac{[\boldsymbol{\mu}_{gn} \cdot \mathbf{E}(\omega_q)] \boldsymbol{\mu}_{nm} [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{ng}^* + \omega_q)(\omega_{mg} - \omega_p)} \\ & \left. + \frac{[\boldsymbol{\mu}_{gn} \cdot \mathbf{E}(\omega_q)] [\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_p)] \boldsymbol{\mu}_{mg}}{(\omega_{ng}^* + \omega_q)(\omega_{mg}^* + \omega_q + \omega_q)} \right) e^{-i(\omega_p + \omega_q)t} \end{aligned}$$

$$\mathbf{P}^{(2)} = N \langle \tilde{\mathbf{p}}^{(2)} \rangle \quad \mathbf{P}^{(2)} = \sum_r \mathbf{P}^{(2)}(\omega_r) \exp(-i\omega_r t)$$

$$P_i^{(2)} = \varepsilon_0 \sum_{jk} \sum_{(pq)} \chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p) E_j(\omega_q) E_k(\omega_p)$$

$$\begin{aligned} \varepsilon_0 \chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p) = \frac{N}{\hbar^2} \mathcal{P}_I \sum_{mn} \bigg(& \frac{\mu_{gn}^i \mu_{nm}^j \mu_{mg}^k}{(\omega_{ng} - \omega_p - \omega_q)(\omega_{mg} - \omega_p)} \\ & + \frac{\mu_{gn}^j \mu_{nm}^i \mu_{mg}^k}{(\omega_{ng}^* + \omega_q)(\omega_{mg} - \omega_p)} + \frac{\mu_{gn}^j \mu_{nm}^k \mu_{mg}^i}{(\omega_{ng}^* + \omega_q)(\omega_{mg}^* + \omega_p + \omega_q)} \bigg) \end{aligned}$$



Susceptibilidade de terceira ordem:

$$\langle \mathbf{p}^{(3)} \rangle = \langle \psi^{(0)} | \hat{\boldsymbol{\mu}} | \psi^{(3)} \rangle + \langle \psi^{(1)} | \hat{\boldsymbol{\mu}} | \psi^{(2)} \rangle + \langle \psi^{(2)} | \hat{\boldsymbol{\mu}} | \psi^{(1)} \rangle + \langle \psi^{(3)} | \hat{\boldsymbol{\mu}} | \psi^{(0)} \rangle$$

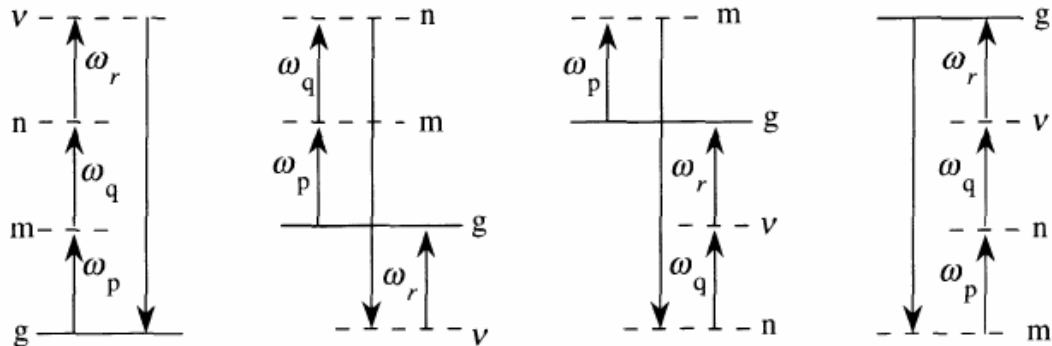
$$\begin{aligned} \langle \mathbf{p}^{(3)} \rangle = & \frac{1}{\hbar^3} \sum_{pqr} \sum_{mnv} \\ & \times \left(\frac{\boldsymbol{\mu}_{gv} [\boldsymbol{\mu}_{vn} \cdot \mathbf{E}(\omega_r)] [\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_q)] [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{vg} - \omega_r - \omega_q - \omega_p)(\omega_{ng} - \omega_q - \omega_p)(\omega_{mg} - \omega_p)} \right. \\ & + \frac{[\boldsymbol{\mu}_{gv} \cdot \mathbf{E}(\omega_r)] \boldsymbol{\mu}_{vn} [\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_q)] [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{vg}^* + \omega_r)(\omega_{ng} - \omega_q - \omega_p)(\omega_{mg} - \omega_p)} \\ & + \frac{[\boldsymbol{\mu}_{gv} \cdot \mathbf{E}(\omega_r)] [\boldsymbol{\mu}_{vn} \cdot \mathbf{E}(\omega_q)] \boldsymbol{\mu}_{nm} [\boldsymbol{\mu}_{mg} \cdot \mathbf{E}(\omega_p)]}{(\omega_{vg}^* + \omega_r)(\omega_{ng}^* + \omega_r + \omega_q)(\omega_{mg} - \omega_p)} \\ & \left. + \frac{[\boldsymbol{\mu}_{gv} \cdot \mathbf{E}(\omega_r)] [\boldsymbol{\mu}_{vn} \cdot \mathbf{E}(\omega_q)] [\boldsymbol{\mu}_{nm} \cdot \mathbf{E}(\omega_p)] \boldsymbol{\mu}_{mg}}{(\omega_{vg}^* + \omega_r)(\omega_{ng}^* + \omega_r + \omega_q)(\omega_{mg}^* + \omega_r + \omega_q + \omega_p)} \right) \\ & \times e^{-i(\omega_p + \omega_q + \omega_r)t} \end{aligned}$$

$$\mathbf{P}^{(3)} = N \langle \tilde{\mathbf{p}}^{(3)} \rangle = \sum_s \mathbf{P}^{(3)}(\omega_s) \exp(-i\omega_s t)$$

$$P_k(\omega_p + \omega_q + \omega_r) = \varepsilon_0 \sum_{hij} \sum_{(pqr)} \chi_{kjih}^{(3)}(\omega_\sigma, \omega_r, \omega_q, \omega_p) E_j(\omega_r) E_i(\omega_q) E_h(\omega_p)$$

$$\chi_{kjih}^{(3)}(\omega_\sigma, \omega_r, \omega_q, \omega_p)$$

$$= \frac{N}{\hbar^3} \mathcal{P}_I \sum_{mnv} \left[\frac{\mu_{gv}^k \mu_{vn}^j \mu_{nm}^i \mu_{mg}^h}{(\omega_{vg} - \omega_r - \omega_q - \omega_p)(\omega_{ng} - \omega_q - \omega_p)(\omega_{mg} - \omega_p)} \right. \\ + \frac{\mu_{gv}^j \mu_{vn}^k \mu_{nm}^i \mu_{mg}^h}{(\omega_{vg}^* + \omega_r)(\omega_{ng} - \omega_q - \omega_p)(\omega_{mg} - \omega_p)} \\ + \frac{\mu_{gv}^j \mu_{vn}^i \mu_{nm}^k \mu_{mg}^h}{(\omega_{vg}^* + \omega_r)(\omega_{ng}^* + \omega_r + \omega_q)(\omega_{mg} - \omega_p)} \\ \left. + \frac{\mu_{gv}^j \mu_{vn}^i \mu_{nm}^h \mu_{mg}^k}{(\omega_{vg}^* + \omega_r)(\omega_{ng}^* + \omega_r + \omega_q)(\omega_{mg}^* + \omega_r + \omega_q + \omega_p)} \right]$$



3.3) Cálculo da susceptibilidade não linear com o formalismo da matriz densidade

$$i\hbar \frac{\partial \psi_s(\mathbf{r}, t)}{\partial t} = \hat{H} \psi_s(\mathbf{r}, t)$$

$$\hat{H} = \hat{H}_0 + \hat{V}(t)$$

$$\psi_s(\mathbf{r}, t) = \sum_n C_n^s(t) u_n(\mathbf{r})$$

$$\hat{H}_0 u_n(\mathbf{r}) = E_n u_n(\mathbf{r})$$

$$\int u_m^*(\mathbf{r}) u_n(\mathbf{r}) d^3r = \delta_{mn}$$

$$i\hbar \sum_n \frac{dC_n^s(t)}{dt} u_n(\mathbf{r}) = \sum_n C_n^s(t) \hat{H} u_n(\mathbf{r})$$

$$i\hbar \frac{d}{dt} C_m^s(t) = \sum_n H_{mn} C_n^s(t)$$

$$H_{mn} = \int u_m^*(\mathbf{r}) \hat{H} u_n(\mathbf{r}) d^3r$$

$$\langle A \rangle = \int \psi_s^* \hat{A} \psi_s d^3r$$

Na notação de Dirac,

$$\langle A \rangle = \langle \psi_s | \hat{A} | \psi_s \rangle = \langle s | \hat{A} | s \rangle$$

$$\langle A \rangle = \sum_{mn} C_m^{s*} C_n^s A_{mn}$$

$$A_{mn} = \langle u_m | \hat{A} | u_n \rangle = \int u_m^* \hat{A} u_n d^3r$$

Definição da matriz densidade

$$\rho_{nm} = \sum_s p(s) C_m^{s*} C_n^s = \overline{C_m^* C_n}$$

$$\langle A \rangle = \sum_{mn} C_m^{s*} C_n^s A_{mn} \quad \overline{\langle A \rangle} = \text{tr}(\hat{\rho} \hat{A}).$$

Evolução temporal da matriz densidade

$$\dot{\rho}_{nm} = \frac{-i}{\hbar} [\hat{H}, \hat{\rho}]_{nm} - \gamma_{nm} (\rho_{nm} - \rho_{nm}^{\text{eq}}) \quad \text{Equação de Liouville}$$

onde a relaxação foi Introduzida fenomenologicamente

3.4) Teoria da perturbação para o cálculo da evolução temporal da matriz densidade

$$\dot{\rho}_{nm} = \frac{-i}{\hbar} [\hat{H}, \hat{\rho}]_{nm} - \gamma_{nm} (\rho_{nm} - \rho_{nm}^{\text{eq}}) \quad \hat{H} = \hat{H}_0 + \hat{V}(t)$$

$$\hat{V} = -\hat{\mu} \cdot \tilde{\mathbf{E}}(t) \quad \hat{\mu} = -e\hat{\mathbf{r}} \quad \hat{H}_0 u_n = E_n u_n \quad H_{0,nm} = E_n \delta_{nm}$$

$$\rho_{nm} = \rho_{nm}^{(0)} + \lambda \rho_{nm}^{(1)} + \lambda^2 \rho_{nm}^{(2)} + \dots$$

com $0 \leq \lambda \leq 1$

$$\begin{aligned} \dot{\rho}_{nm}^{(0)} &= -i\omega_{nm}\rho_{nm}^{(0)} - \gamma_{nm}(\rho_{nm}^{(0)} - \rho_{nm}^{\text{eq}}), \\ \dot{\rho}_{nm}^{(1)} &= -(i\omega_{nm} + \gamma_{nm})\rho_{nm}^{(1)} - i\hbar^{-1}[\hat{V}, \hat{\rho}^{(0)}]_{nm}, \\ \dot{\rho}_{nm}^{(2)} &= -(i\omega_{nm} + \gamma_{nm})\rho_{nm}^{(2)} - i\hbar^{-1}[\hat{V}, \hat{\rho}^{(1)}]_{nm}, \end{aligned}$$

$$\rho_{nm}^{(1)}(t) = \int_{-\infty}^t \frac{-i}{\hbar} [\hat{V}(t'), \hat{\rho}^{(0)}]_{nm} e^{(i\omega_{nm} + \gamma_{nm})(t' - t)} dt'$$

3.5) Cálculo da susceptibilidade linear pela matriz densidade

$$\rho_{nm}^{(1)} = \hbar^{-1} (\rho_{mm}^{(0)} - \rho_{nn}^{(0)}) \sum_p \frac{\mu_{nm} \cdot \mathbf{E}(\omega_p) e^{-i\omega_p t}}{(\omega_{nm} - \omega_p) - i\gamma_{nm}}$$

$$\langle \tilde{\mu}(t) \rangle = \text{tr}(\hat{\rho}^{(1)} \hat{\mu}) = \sum_{nm} \rho_{nm}^{(1)} \mu_{mn} = \sum_{nm} \hbar^{-1} (\rho_{mm}^{(0)} - \rho_{nn}^{(0)}) \sum_p \frac{\mu_{mn} [\mu_{nm} \cdot \mathbf{E}(\omega_p)] e^{-i\omega_p t}}{(\omega_{nm} - \omega_p) - i\gamma_{nm}}$$

$$\mathbf{P}(\omega_p) = N \langle \boldsymbol{\mu}(\omega_p) \rangle = \varepsilon_0 \boldsymbol{\chi}^{(1)}(\omega_p) \cdot \mathbf{E}(\omega_p)$$

$$\varepsilon_0 \boldsymbol{\chi}^{(1)}(\omega_p) = \frac{N}{\hbar} \sum_{nm} (\rho_{mm}^{(0)} - \rho_{nn}^{(0)}) \frac{\boldsymbol{\mu}_{mn} \boldsymbol{\mu}_{nm}}{(\omega_{nm} - \omega_p) - i\gamma_{nm}}$$

$$\varepsilon_0 \chi_{ij}^{(1)}(\omega_p) = \frac{N}{\hbar} \sum_n \left[\frac{\mu_{an}^i \mu_{na}^j}{(\omega_{na} - \omega_p) - i\gamma_{na}} + \frac{\mu_{na}^i \mu_{an}^j}{(\omega_{na} + \omega_p) + i\gamma_{na}} \right]$$

Ressonante

Anti-ressonante

3.6) Cálculo da susceptibilidade de segunda ordem pela matriz densidade

$$\rho_{nm}^{(2)} = e^{-(i\omega_{nm} + \gamma_{nm})t} \int_{-\infty}^t \frac{-i}{\hbar} [\hat{V}, \hat{\rho}^{(1)}]_{nm} e^{(i\omega_{nm} + \gamma_{nm})t'} dt'$$

$$\begin{aligned} \rho_{nm}^{(2)} = & \sum_v \sum_{pq} e^{-i(\omega_p + \omega_q)t} \\ & \times \left\{ \frac{\rho_{mm}^{(0)} - \rho_{vv}^{(0)}}{\hbar^2} \frac{[\boldsymbol{\mu}_{nv} \cdot \mathbf{E}(\omega_q)][\boldsymbol{\mu}_{vm} \cdot \mathbf{E}(\omega_p)]}{[(\omega_{nm} - \omega_p - \omega_q) - i\gamma_{nm}][(\omega_{vm} - \omega_p) - i\gamma_{vm}]} \right. \\ & \left. - \frac{\rho_{vv}^{(0)} - \rho_{nn}^{(0)}}{\hbar^2} \frac{[\boldsymbol{\mu}_{nv} \cdot \mathbf{E}(\omega_q)][\boldsymbol{\mu}_{vm} \cdot \mathbf{E}(\omega_q)]}{[(\omega_{nm} - \omega_p - \omega_q) - i\gamma_{nm}][(\omega_{nv} - \omega_p) - i\gamma_{nv}]} \right\} \end{aligned}$$

$$\langle \tilde{\boldsymbol{\mu}} \rangle = \sum_{nm} \rho_{nm} \boldsymbol{\mu}_{mn}$$

$$\mathbf{P}^{(2)}(\omega_p + \omega_q) = N \langle \boldsymbol{\mu}(\omega_p + \omega_q) \rangle = N \sum_{nmv} \sum_{(pq)} K_{nmv} \boldsymbol{\mu}_{mn}$$

$$P_i^{(2)}(\omega_p + \omega_q) = \epsilon_0 \sum_{jk} \sum_{(pq)} \chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p) E_j(\omega_q) E_k(\omega_p)$$

$$\varepsilon_0 \chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p)$$

$$= \frac{N}{2\hbar^2} \sum_{lmn} \rho_{ll}^{(0)} \left\{ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \right. \quad (a_1)$$

$$+ \frac{\mu_{ln}^i \mu_{nm}^k \mu_{ml}^j}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_q) - i\gamma_{ml}]} \quad (a_2)$$

$$+ \frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{[(\omega_{mn} - \omega_p - \omega_q) - i\gamma_{mn}][(\omega_{nl} + \omega_p) + i\gamma_{nl}]} \quad (a'_1)$$

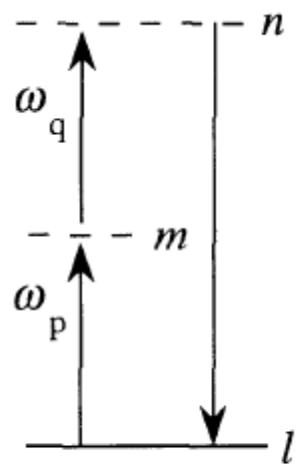
$$+ \frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{[(\omega_{mn} - \omega_p - \omega_q) - i\gamma_{mn}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \quad (a'_2)$$

$$+ \frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{[(\omega_{nm} + \omega_p + \omega_q) + i\gamma_{nm}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \quad (b_1)$$

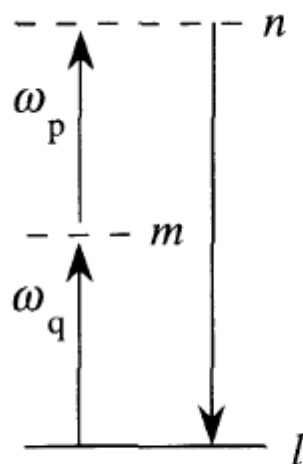
$$+ \frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{[(\omega_{nm} + \omega_p + \omega_q) + i\gamma_{nm}][(\omega_{ml} - \omega_q) - i\gamma_{ml}]} \quad (b_2)$$

$$+ \frac{\mu_{ln}^k \mu_{nm}^j \mu_{ml}^i}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{ml}][(\omega_{nl} + \omega_p) + i\gamma_{nl}]} \quad (b'_1)$$

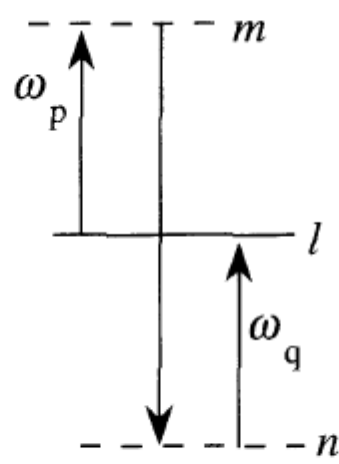
$$+ \frac{\mu_{ln}^j \mu_{nm}^k \mu_{ml}^i}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{ml}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \left. \right\}. \quad (b'_2).$$



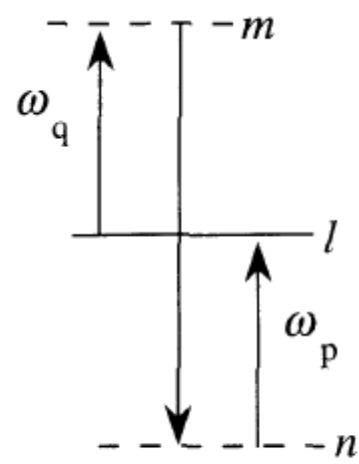
(a₁)



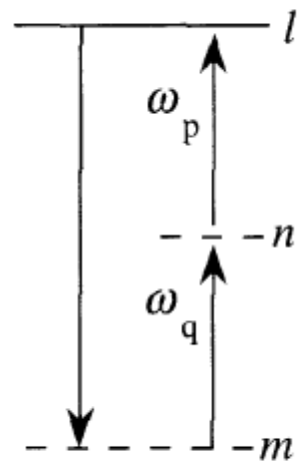
(a₂)



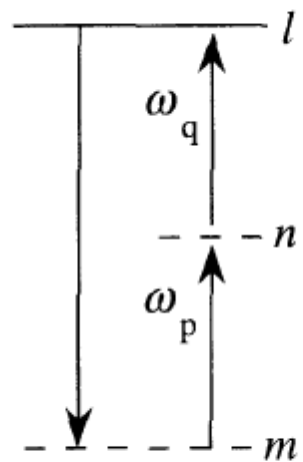
(b₁) (a'₂)



(b₂) (a'₁)



(b'₁)

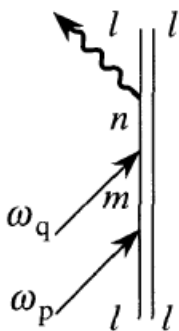


(b'₂)

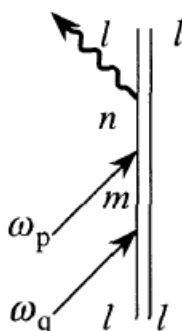
double-sided Feynman diagrams

$$\hat{\rho} = |\psi\rangle\langle\psi|$$

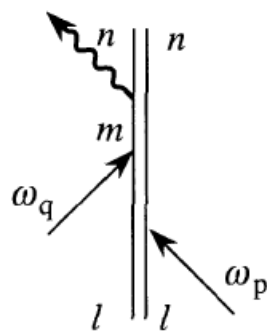
$$\rho_{nm} = \langle n|\hat{\rho}|m\rangle$$



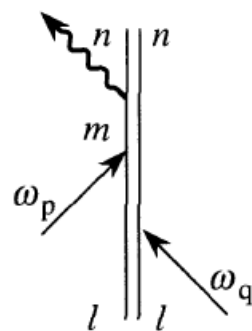
(a₁)



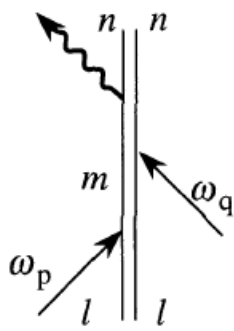
(a₂)



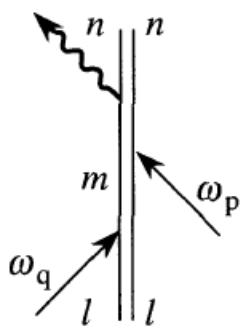
(a'₁)



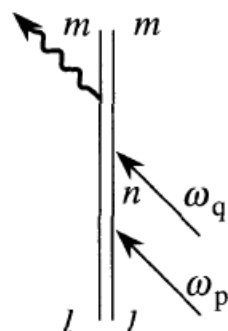
(a'₂)



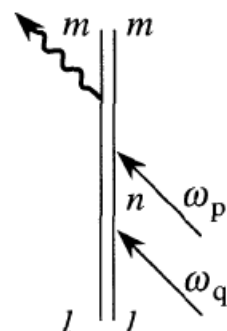
(b₁)



(b₂)

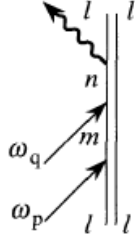
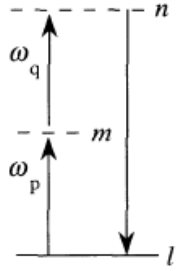


(b'₁)

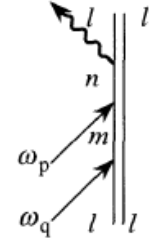
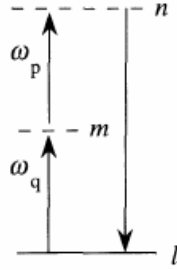


(b'₂)

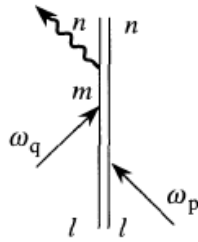
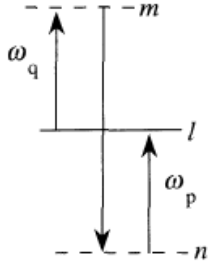
$$\omega_p \leftrightarrow k \quad \omega_q \leftrightarrow j \quad \omega_p + \omega_q \leftrightarrow i$$



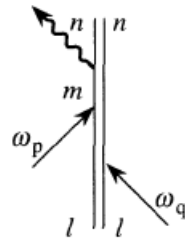
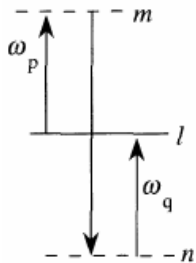
$$\begin{aligned} & \epsilon_0 \chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p) \\ &= \frac{N}{2\hbar^2} \sum_{lmn} \rho_{ll}^{(0)} \left\{ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \right. \end{aligned} \quad (a_1)$$



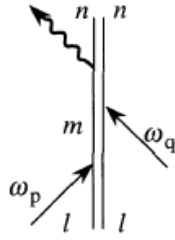
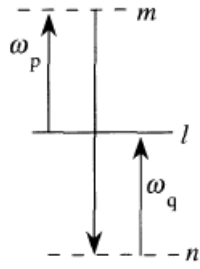
$$+ \frac{\mu_{ln}^i \mu_{nm}^k \mu_{ml}^j}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_q) - i\gamma_{ml}]} \quad (a_2)$$



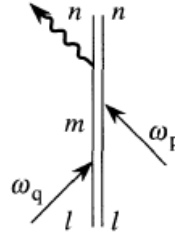
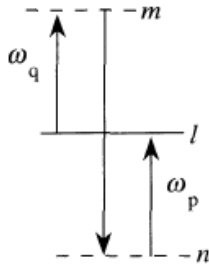
$$+ \frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{[(\omega_{mn} - \omega_p - \omega_q) - i\gamma_{mn}][(\omega_{nl} + \omega_p) + i\gamma_{nl}]} \quad (a_1')$$



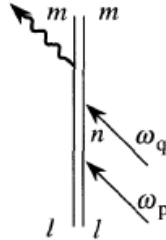
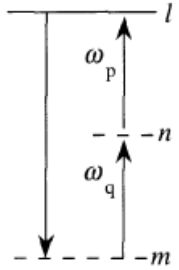
$$+ \frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{[(\omega_{mn} - \omega_p - \omega_q) - i\gamma_{mn}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \quad (a_2')$$



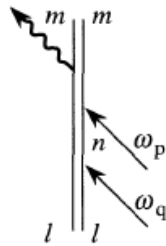
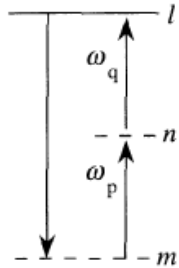
$$+ \frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{[(\omega_{nm} + \omega_p + \omega_q) + i\gamma_{nm}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \quad (b_1)$$



$$+ \frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{[(\omega_{nm} + \omega_p + \omega_q) + i\gamma_{nm}][(\omega_{ml} - \omega_q) - i\gamma_{ml}]} \quad (b_2)$$



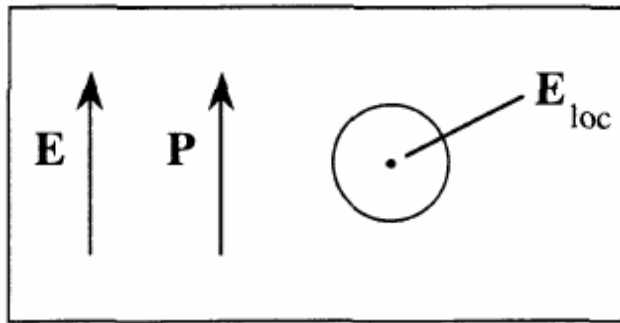
$$+ \frac{\mu_{ln}^k \mu_{nm}^j \mu_{ml}^i}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{ml}][(\omega_{nl} + \omega_p) + i\gamma_{nl}]} \quad (b'_1)$$



$$+ \frac{\mu_{ln}^j \mu_{nm}^k \mu_{ml}^i}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{ml}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \Bigg\}. \quad (b'_2)$$

3.7 – Correção de campo local para a susceptibilidade não linear

Efeito do campo local na óptica linear



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

$$\vec{E}_{\text{loc}} = \vec{E} + \frac{\vec{P}}{3\epsilon_0}$$

$$\frac{\epsilon^{(1)}}{\epsilon_0} = n^2 = 1 + \chi^{(1)}$$

$$\vec{E}_{\text{loc}} = \vec{E} + \frac{\chi^{(1)}}{3} \vec{E} = \left(1 + \frac{\chi^{(1)}}{3}\right) \vec{E} = \left(\frac{n^2 + 2}{3}\right) \vec{E}$$

Efeito do campo local na óptica não linear

$$\begin{aligned} &\mathcal{L}^{(2)}(\omega_m + \omega_n, \omega_m, \omega_n) \\ &= \left(\frac{n^2(\omega_m + \omega_n) + 2}{3}\right) \left(\frac{n^2(\omega_m) + 2}{3}\right) \left(\frac{n^2(\omega_n) + 2}{3}\right) \end{aligned}$$

$$\mathcal{L}^{(3)}(\omega_l + \omega_m + \omega_n, \omega_l, \omega_m, \omega_n) =$$

$$\left(\frac{n^2 (\omega_l + \omega_m + \omega_n) + 2}{3}\right) \left(\frac{n^2 (\omega_l) + 2}{3}\right) \left(\frac{n^2 (\omega_m) + 2}{3}\right) \left(\frac{n^2 (\omega_n) + 2}{3}\right)$$

Aula 4 – O índice de refração dependente da intensidade



Sérgio Carlos Zilio
IFSC - USP

4.1) Descrição do processo

Vamos tratar o caso: $P^{NL}(\omega) = 3\varepsilon_0\chi^{(3)}(\omega; \omega, \omega, -\omega)|E(\omega)|^2 E(\omega)$

$$P^{TOT}(\omega) = \varepsilon_0\chi^{(1)}E(\omega) + 3\varepsilon_0\chi^{(3)}|E(\omega)|^2 E(\omega) \equiv \varepsilon_0\chi_{eff}E(\omega)$$

$$\chi_{eff} = \chi^{(1)} + 3\chi^{(3)}|E(\omega)|^2 \quad n^2 = 1 + \chi_{eff} = 1 + \chi^{(1)} + 3\chi^{(3)}|E(\omega)|^2 = n_0^2 + 3\chi^{(3)}|E(\omega)|^2$$

$$n = n_0 \sqrt{1 + 3\chi^{(3)}|E(\omega)|^2 / n_0^2} \cong n_0 + \frac{3\chi^{(3)}|E(\omega)|^2}{2n_0} = n_0 + n_2 I$$

$$E(t) = E(\omega)e^{-i\omega t} + E^*(\omega)e^{i\omega t} \quad \Rightarrow \quad I = 2n_0 c \varepsilon_0 |E(\omega)|^2$$

$$n_2 \left(\frac{m^2}{W} \right) = \frac{3\chi^{(3)}}{4cn_0^2\varepsilon_0} \text{ (MKS)}$$

4.2) Natureza tensorial da susceptibilidade de terceira ordem

$\chi^{(3)}$ é um tensor de quarta ordem e para cristais com baixa simetria pode possuir 81 elementos independentes e não nulos.

Mas para um material isotrópico como vidro, líquido ou gas, no caso geral de frequências arbitrárias,

$$\chi_{ijkl} = \chi_{1122}\delta_{ij}\delta_{kl} + \chi_{1212}\delta_{ik}\delta_{jl} + \chi_{1221}\delta_{il}\delta_{jk}$$

Para efeito degenerado, $\chi_{1122} = \chi_{1212}$ (permutação intrínseca)

$$\chi_{ijkl}(\omega = \omega + \omega - \omega) = \chi_{1122}(\omega = \omega + \omega - \omega)(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + \chi_{1221}(\omega = \omega + \omega - \omega)(\delta_{il}\delta_{jk})$$

$$P_i(\omega) = 3\epsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)}(\omega; \omega, \omega, -\omega) E_j(\omega) E_k(\omega) E_l(-\omega)$$

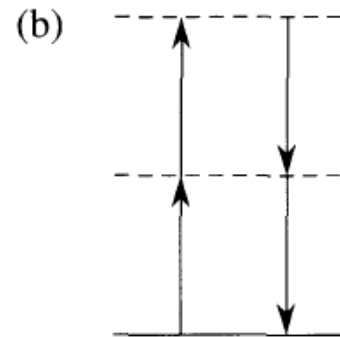
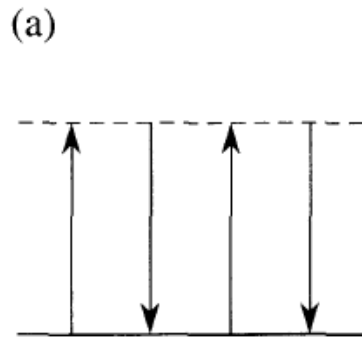
$$P_i = 6\epsilon_0 \chi_{1122}^{(3)} E_i(\omega) (\vec{E} \cdot \vec{E}^*) + 3\epsilon_0 \chi_{1221}^{(3)} E_i(\omega) (\vec{E} \cdot \vec{E})$$

$$\vec{P} = 6\epsilon_0 \chi_{1122}^{(3)} (\vec{E} \cdot \vec{E}^*) \vec{E} + 3\epsilon_0 \chi_{1221}^{(3)} (\vec{E} \cdot \vec{E}) \vec{E}^*$$

$$A = 6\chi_{1122}$$

$$B = 6\chi_{1221}$$

$$\vec{P} = \epsilon_0 A (\vec{E} \cdot \vec{E}^*) \vec{E} + \frac{1}{2} \epsilon_0 B (\vec{E} \cdot \vec{E}) \vec{E}^*$$



$$B/A = 6 \quad \text{for molecular orientation}$$

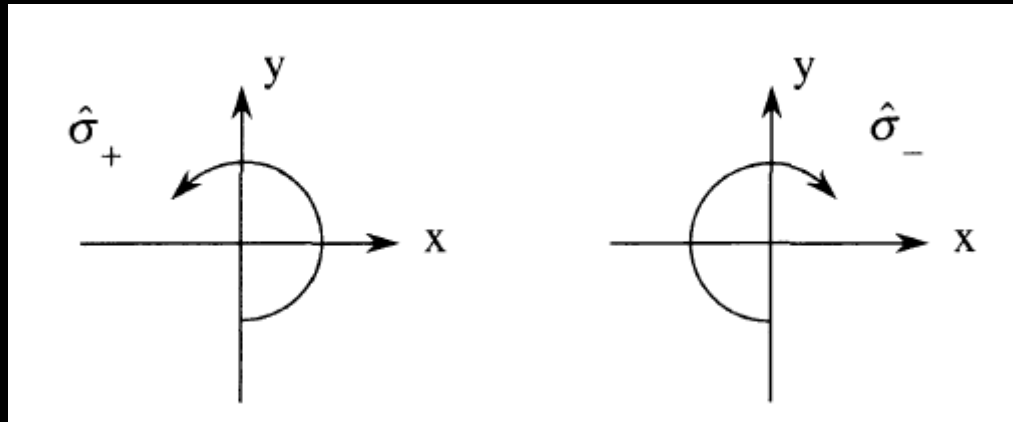
$$B/A = 1 \quad \text{for nonresonant electronic response}$$

Propagação por um meio isotrópico

$$\mathbf{P} = \epsilon_0 A (\mathbf{E} \cdot \mathbf{E}^*) \mathbf{E} + \frac{1}{2} \epsilon_0 B (\mathbf{E} \cdot \mathbf{E}) \mathbf{E}^*$$

$$\mathbf{E} = E_+ \hat{\sigma}_+ + E_- \hat{\sigma}_-$$

$$\hat{\sigma}_{\pm} = \frac{\hat{x} \pm i \hat{y}}{\sqrt{2}}$$

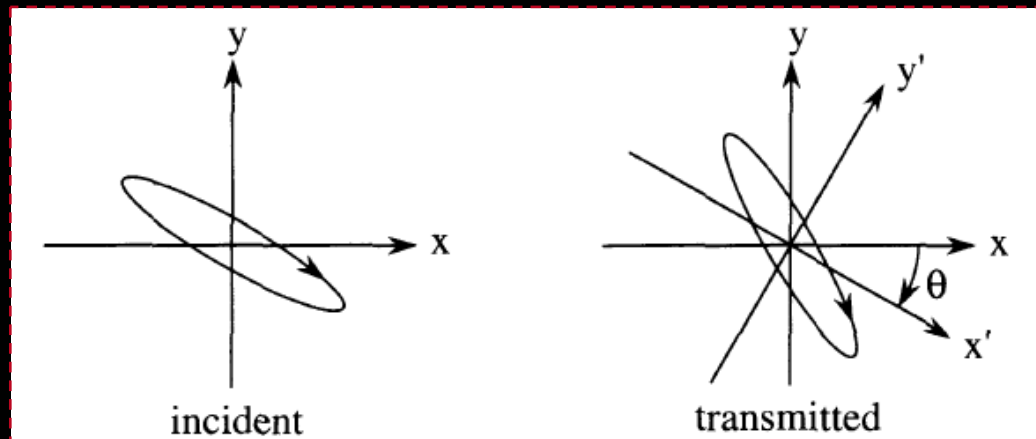


Luz circularmente polarizada

$$\delta n_{\text{circ}} = n - n_0 = \frac{1}{2n_0} A |E|^2$$

Luz linearmente polarizada

$$\delta n_{\text{linear}} = n - n_0 = \frac{1}{2n_0} \left(A + \frac{1}{2} B \right) |E|^2$$



Luz elipticamente polarizada

$$\theta = \frac{1}{2} \Delta n \frac{\omega}{c} z$$

$$\Delta n \equiv n_+ - n_- = \frac{B}{2n_0} \left[|E_-|^2 - |E_+|^2 \right]$$

4.3) Não linearidades eletrônicas não ressonantes

Tempo de resposta: $\tau = 2\pi a_0/v$

$$a_0 = 0.5 \times 10^{-8} \text{ cm} \quad v \simeq c/137 \quad \tau \simeq 10^{-16} \text{ s}$$

Modelo clássico do oscilador não harmônico

$$\omega \ll \omega_0$$

$$\chi^{(3)} \simeq \frac{Ne^4}{\epsilon_0 m^3 \omega_0^6 d^2}$$

Modelo quântico sem amortecimento

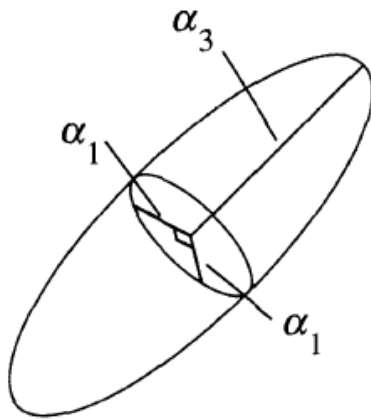
$$\chi^{(3)} \simeq \frac{8N\mu^4}{\epsilon_0 \hbar^3 \omega_0^3}$$

$$n_2 \left(\frac{\text{m}^2}{\text{W}} \right) = \frac{3\chi^{(3)}}{4cn_0^2 \epsilon_0} (\text{MKS})$$

Material	n_0	$\chi_{1111} \text{ (m}^2/\text{V}^2\text{)}$	$n_2 \text{ (m}^2/\text{W)}$
Diamond	2.42	21×10^{-22}	10×10^{-20}
Yttrium aluminum garnet	1.83	8.4×10^{-22}	8.4×10^{-20}
Sapphire	1.8	4.2×10^{-22}	3.7×10^{-20}
Borosilicate crown glass	1.5	3.5×10^{-22}	4.4×10^{-20}
Fused silica	1.47	2.8×10^{-22}	3.67×10^{-20}
CaF ₂	1.43	2.24×10^{-22}	3.1×10^{-20}
LiF	1.4	1.4×10^{-22}	2.0×10^{-20}

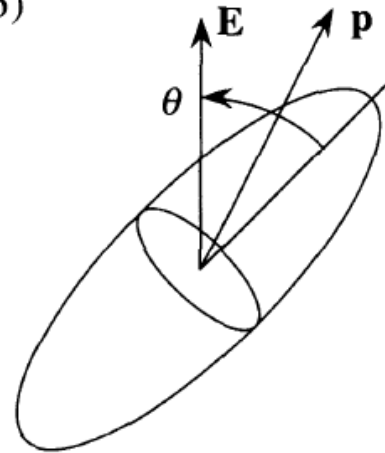
4.4) Não linearidades devidas à orientação molecular

(a)



$$\alpha_3 > \alpha_1$$

(b)



$$\boldsymbol{\tau} = \mathbf{p} \times \mathbf{E}$$

$$n_2 = \frac{N}{45n_0} \left(\frac{n_0^2 + 2}{3} \right)^4 \frac{(\alpha_3 - \alpha_1)^2}{kT}$$

Para o CS_2 , $n_2 = 3 \times 10^{-14} \text{ (cm}^2/\text{W)}$

4.5) Não linearidades térmicas

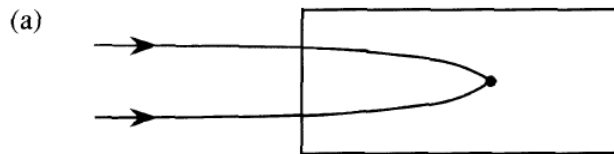
$$n = n_0 + \left(\frac{dn}{dT} \right) T_1 \quad T_1: \text{acr scimo de temperatura induzida pelo laser}$$

$$(\rho_0 C) \frac{\partial T_1}{\partial t} - \kappa \nabla^2 T_1 = \alpha I(r) \quad \tau \approx 1 \text{ ms}$$

Material	$(\rho_0 C)$ (J/cm ³) ^a	κ (W/m K)	dn/dT (K ⁻¹) ^b
Diamond	1.76	660	
Ethanol	1.91	0.168	
Fused silica	1.67	1.4	1.2×10^{-5}
Sodium chloride	1.95	6.4	-3.6×10^{-5}
Water (liquid)	4.2	0.56	
Air ^c	1.2×10^{-3}	26×10^{-3}	-1.0×10^{-6}

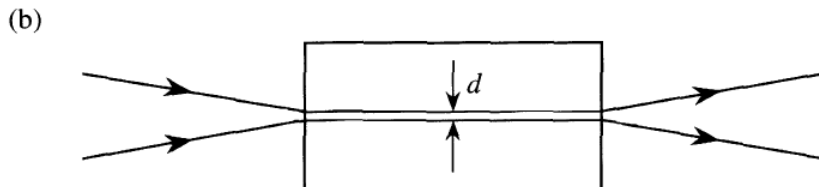
$$n_2^{(\text{th})} = 10^{-5} \text{ cm}^2/\text{W}$$

4.6) Auto-focalização e outros efeitos transversais



Auto-focalização

$$P > P_{\text{cr}}$$



Auto-aprisionamento
(sóliton espacial)

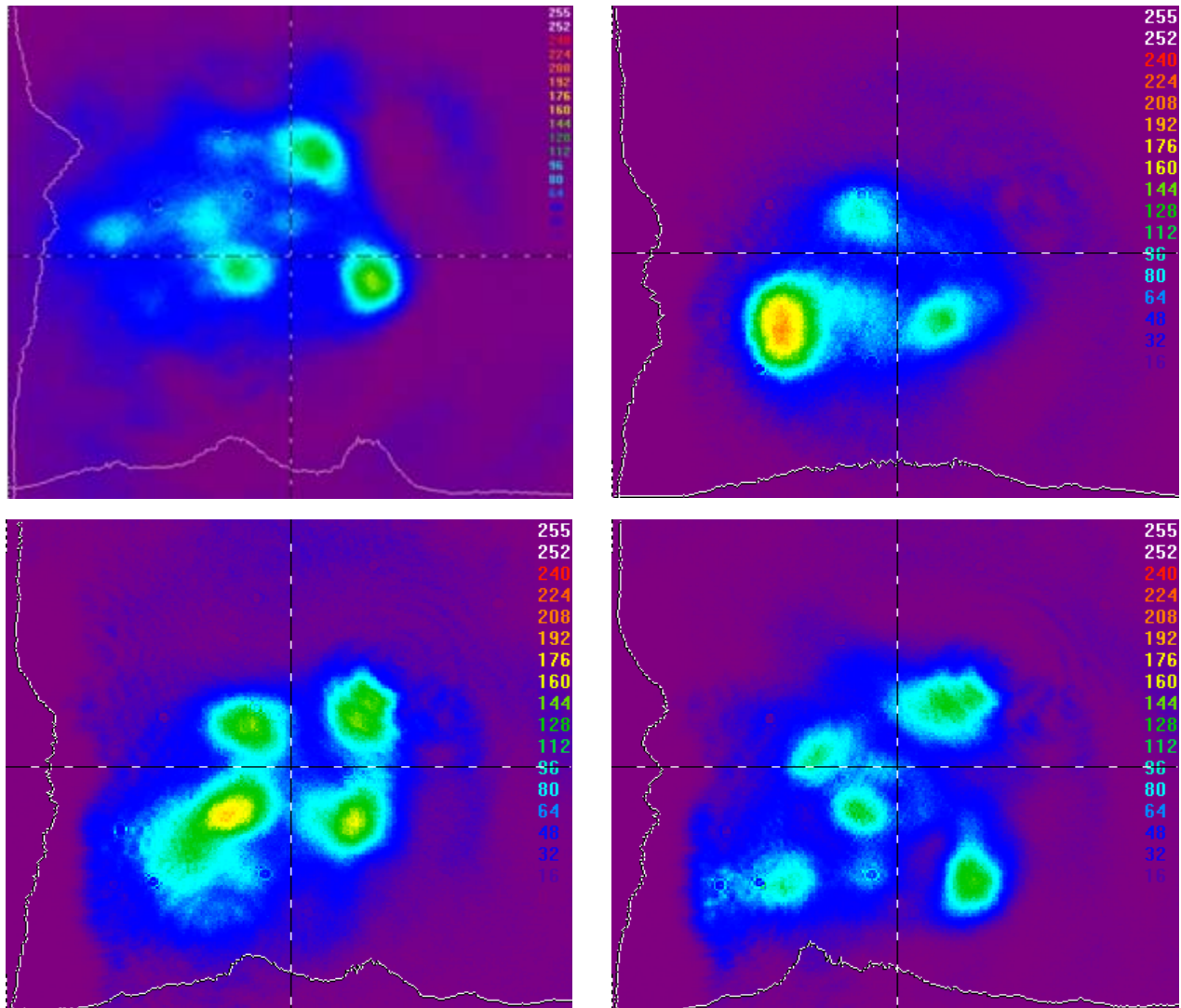
$$P_{\text{cr}} = \frac{\pi (0.61)^2 \lambda_0^2}{8 n_0 n_2}$$



Filamentação

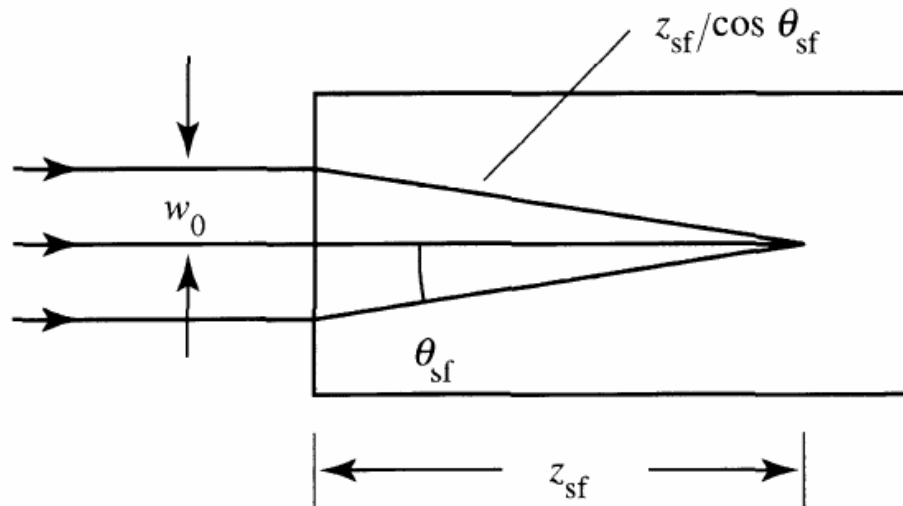
$$P \gg P_{\text{cr}}$$

Examples of beam filamentation



All peak powers are in the 15 to 35GW/cm² range. All beams began life smooth

Modelo simples do processo de auto-focalização (desprezando difração)



$$P_{cr} = \frac{\pi (0.61)^2 \lambda_0^2}{8 n_0 n_2}$$

Princípio de Fermat: todos os raios com mesmo caminho óptico

$$\Rightarrow \int n(\mathbf{r}) dl = \text{constante}$$

$$(n_0 + n_2 I) z_{sf} = n_0 z_{sf} / \cos \theta_{sf} \quad \cos \theta_{sf} \approx 1 - \frac{1}{2} \theta_{sf}^2$$

$$\theta_{sf} = \sqrt{2 n_2 I / n_0}$$

Ângulo de auto-focalização

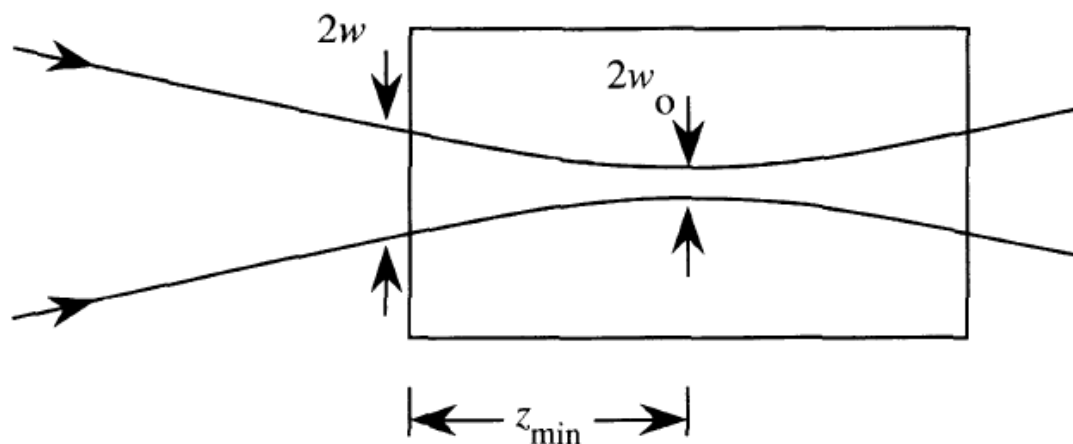
$$z_{sf} = w_0 / \theta_{sf} = w_0 \sqrt{\frac{n_0}{2 n_2 I}} = \frac{2 n_0 w_0^2}{\lambda_0} \frac{1}{\sqrt{P / P_{cr}}} \quad (P \gg P_{cr})$$

Considerando a difração,

$$\theta = (\theta_{sf}^2 - \theta_{dif}^2)^{1/2}, \quad \theta_{dif} = 0.61\lambda_0/n_0d \quad z_{sf} = w_0/\theta = \frac{2nw_0^2}{\lambda_0} \frac{1}{\sqrt{P/P_{cr} - 1}}$$

Yariv (1975)

$$z_{sf} = \frac{\frac{1}{2}kw^2}{(P/P_{cr} - 1)^{1/2} + 2z_{min}/kw_0^2}$$



Auto-aprisionamento

$$\theta_{\text{dif}} = \theta_{\text{sf}}$$

$$\theta_{\text{sf}} = \sqrt{2n_2 I / n_0}$$

$$\theta_{\text{dif}} = 0.61\lambda_0 / n_0 d$$

$$I = \frac{(0.61)^2 \lambda_0^2}{2n_2 n_0 d^2}$$

$$P = (\pi/4)d^2 I$$

$$P_{\text{cr}} = \frac{\pi (0.61)^2 \lambda_0^2}{8n_0 n_2} \approx \frac{\lambda_0^2}{8n_0 n_2}$$

$$\delta n = \frac{1}{2} n_0 (0.61\lambda_0 / d n_0)^2 \quad d = 0.61\lambda_0 (2n_0 \delta n)^{-1/2} = 0.61\lambda_0 (2n_0 n_2 I)^{-1/2}$$

Para o CS_2 , $n_2 = 3.2 \times 10^{-14} \text{ cm}^2/\text{W}$, $n_0 = 1.7$, $P_{\text{cr}}(1\mu\text{m}) = 2.7 \text{ kW}$.

Para vidros e gases típicos, $5 \times 10^{-16} \leq n_2 \leq 5 \times 10^{-15} \text{ cm}^2/\text{W}$ e $0.2 \leq P_{\text{cr}} \leq 2 \text{ MW}$.

Descrição matemática dos efeitos de auto-ação

$$2ik_0 \frac{\partial A}{\partial z} + \nabla_T^2 A = -\frac{\omega^2}{\epsilon_0 c^2} p_{\text{NL}} \quad p_{\text{NL}} = 3\epsilon_0 \chi^{(3)} |A|^2 A$$

Em apenas uma dimensão (guia de onda planar)

$$2ik_0 \frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3\chi^{(3)} \frac{\omega^2}{c^2} |A|^2 A$$

Esta equação possui uma solução do tipo: $A(x, z) = A_0 \operatorname{sech}(x/x_0) e^{i\gamma z}$

$$x_0 = \frac{1}{k_0} \sqrt{n_0 / 2\bar{n}_2 |A_0|^2} \quad \gamma = k_0 \bar{n}_2 |A_0|^2 / n_0 \quad \bar{n}_2 = 3\chi^{(3)} / 4n_0$$

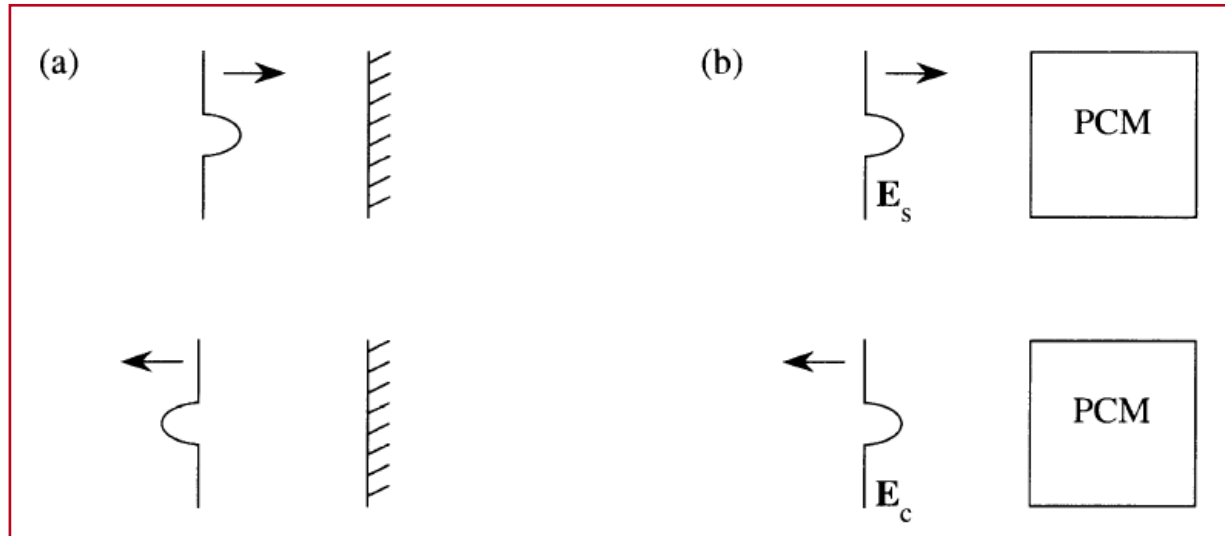
Detailed analysis shows that in two transverse dimensions spatial solitons are unstable in a pure Kerr medium (that is, one described by an $\bar{n}_2$ nonlinearity), but that they can propagate stably in a saturable nonlinear medium. Such behavior has been observed experimentally by Bjorkholm and Ashkin (1974). Higher-order solutions have been reported by Haus (1966).



Z-scan

www.optics.unm.edu/sbahaee/z-scan.htm

4.7) Conjugação de fase



signal wave

$$\mathbf{E}_s(\mathbf{r}, t) = \mathbf{E}_s(\mathbf{r})e^{-i\omega t} + \text{c.c.}$$

$$\mathbf{E}_s(\mathbf{r}) = \hat{\mathbf{e}}_s A_s(\mathbf{r})e^{i\mathbf{k}_s \cdot \mathbf{r}}$$

$$\mathbf{E}_c(\mathbf{r}, t) = r\mathbf{E}_s^*(\mathbf{r})e^{-i\omega t} + \text{c.c.}$$

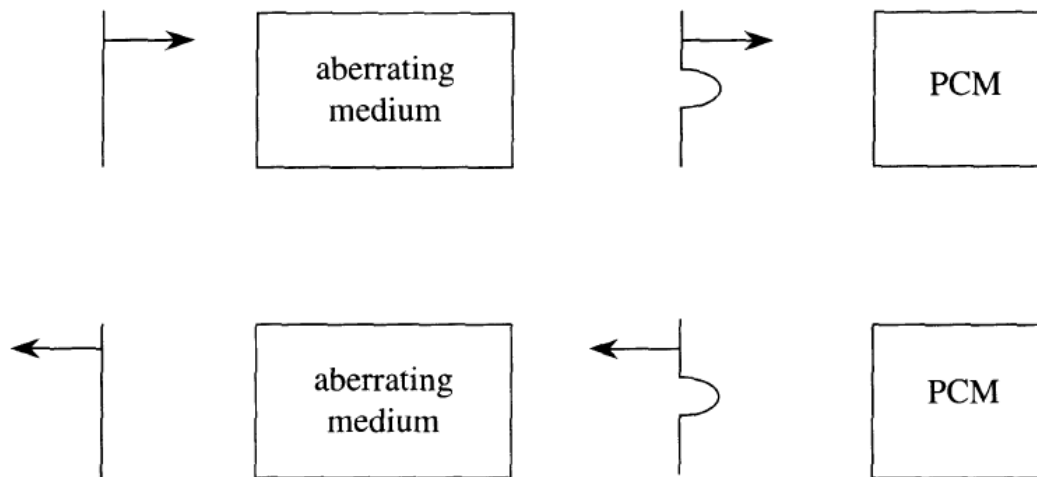
$$\mathbf{E}_s^*(\mathbf{r}) = \hat{\mathbf{e}}_s^* A_s^*(\mathbf{r})e^{-i\mathbf{k}_s \cdot \mathbf{r}}$$

$$\mathbf{E}_s(\mathbf{r}) = \hat{\mathbf{e}}_s A_s(\mathbf{r}) e^{i\mathbf{k}_s \cdot \mathbf{r}}$$

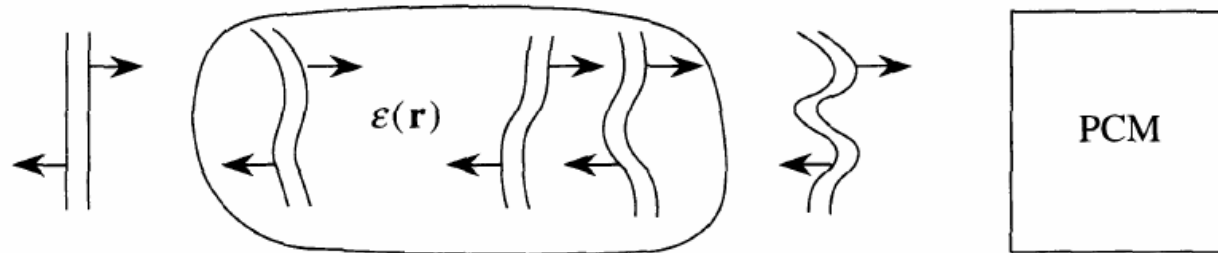
$$\mathbf{E}_s^*(\mathbf{r}) = \hat{\mathbf{e}}_s^* A_s^*(\mathbf{r}) e^{-i\mathbf{k}_s \cdot \mathbf{r}}$$

Vemos que a ação de um conjugador de fase ideal é tripla:

- 1) O vetor unitário complexo da polarização é substituído pelo seu complexo conjugado. Por exemplo, luz σ^+ continua σ^+ ao invés de ser convertida em σ^- como num espelho de alumínio.
- 2) $A_s(\mathbf{r})$ é substituído pelo seu complexo conjugado, indicando que a frente de onda é invertida, como indicado na figura.
- 3) $\mathbf{k}_s$ é substituído por $-\mathbf{k}_s$, mostrando que a onda incidente é refletida sobre si mesma (reversão temporal)



Correção de aberração por conjugação de fase



$$\nabla^2 E - \frac{\epsilon(\mathbf{r})}{c^2} \frac{\partial^2 E}{\partial t^2} = 0 \quad E(\mathbf{r}, t) = A(\mathbf{r})e^{i(kz - \omega t)} + \text{c.c.}$$

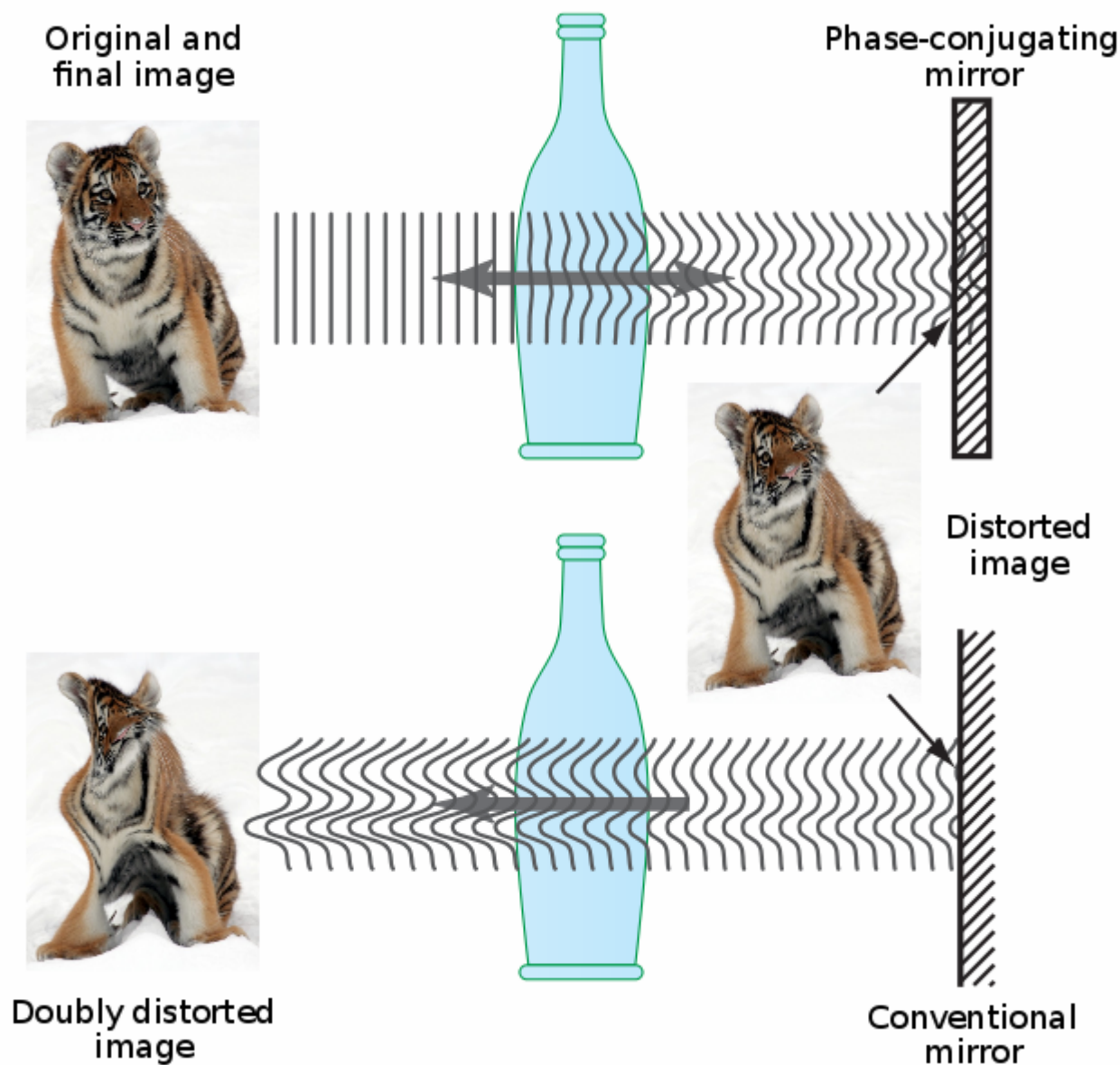
$$\nabla^2 = \frac{\partial^2}{\partial z^2} + \nabla_T^2 \quad \nabla_T^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2$$

$$\nabla_T^2 A + \left[\frac{\omega^2 \epsilon(\mathbf{r})}{c^2} - k^2 \right] A + 2ik \frac{\partial A}{\partial z} = 0.$$

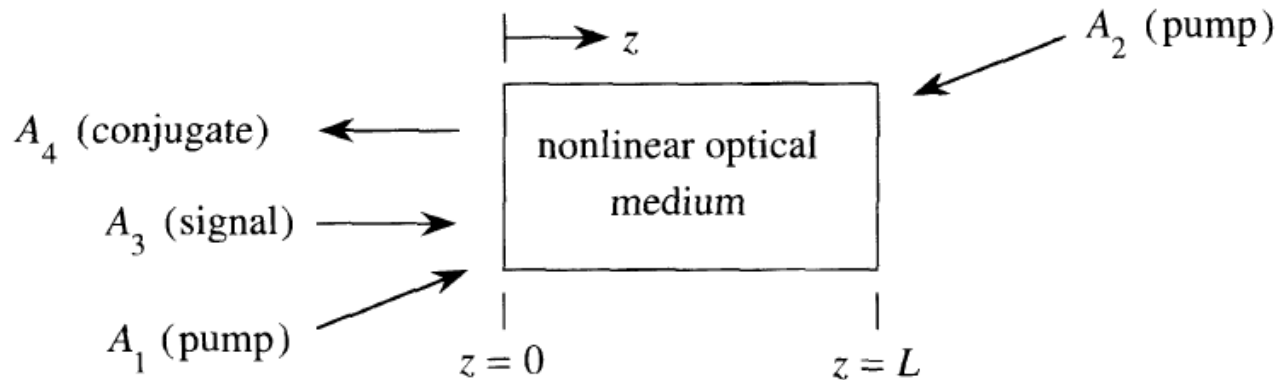
$$\nabla_T^2 A^* + \left[\frac{\omega^2 \epsilon(\mathbf{r})}{c^2} - k^2 \right] A^* - 2ik \frac{\partial A^*}{\partial z} = 0$$

$$E_c(\mathbf{r}, t) = A^*(\mathbf{r})e^{i(-kz - \omega t)} + \text{c.c.}$$

Correção de aberração por conjugação de fase



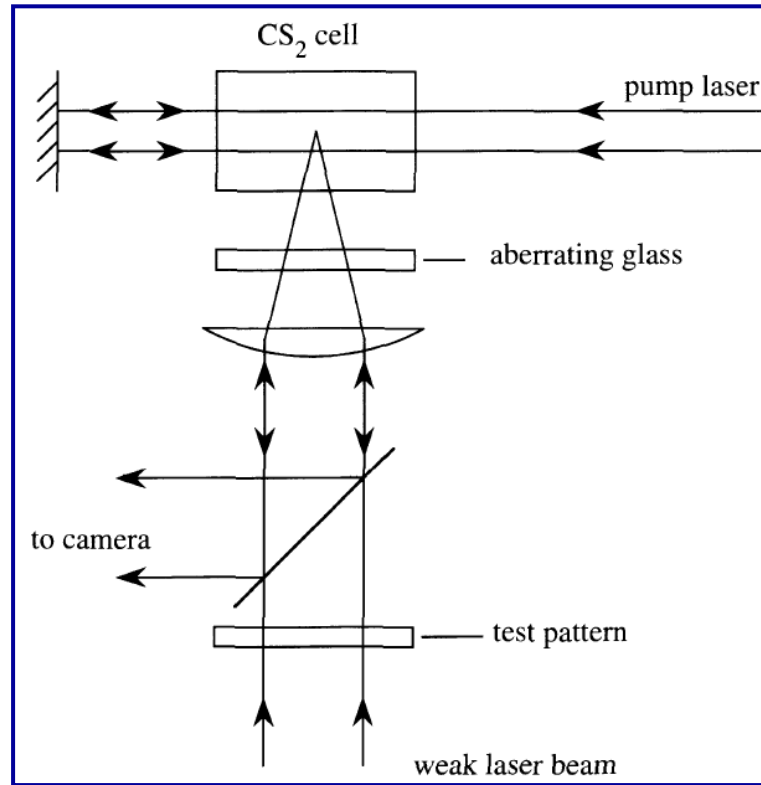
Conjugação de fase por mistura de quatro ondas degeneradas



$$E_i(\mathbf{r}, t) = E_i(\mathbf{r})e^{-i\omega t} + \text{c.c.} = A_i(\mathbf{r})E^{i(\mathbf{k}_i \cdot \mathbf{r} - \omega t)} + \text{c.c.} \quad i = 1, 2, 3, 4$$

$$P^{\text{NL}} = 6\epsilon_0 \chi^{(3)} E_1 E_2 E_3^* = 6\epsilon_0 \chi^{(3)} A_1 A_2 A_3^* e^{i(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3) \cdot \mathbf{r}} \quad \mathbf{k}_1 + \mathbf{k}_2 = 0$$

$$P^{\text{NL}} = 6\epsilon_0 \chi^{(3)} A_1 A_2 A_3^* e^{-i\mathbf{k}_3 \cdot \mathbf{r}} \quad \mathbf{k}_3 + \mathbf{k}_4 = 0$$



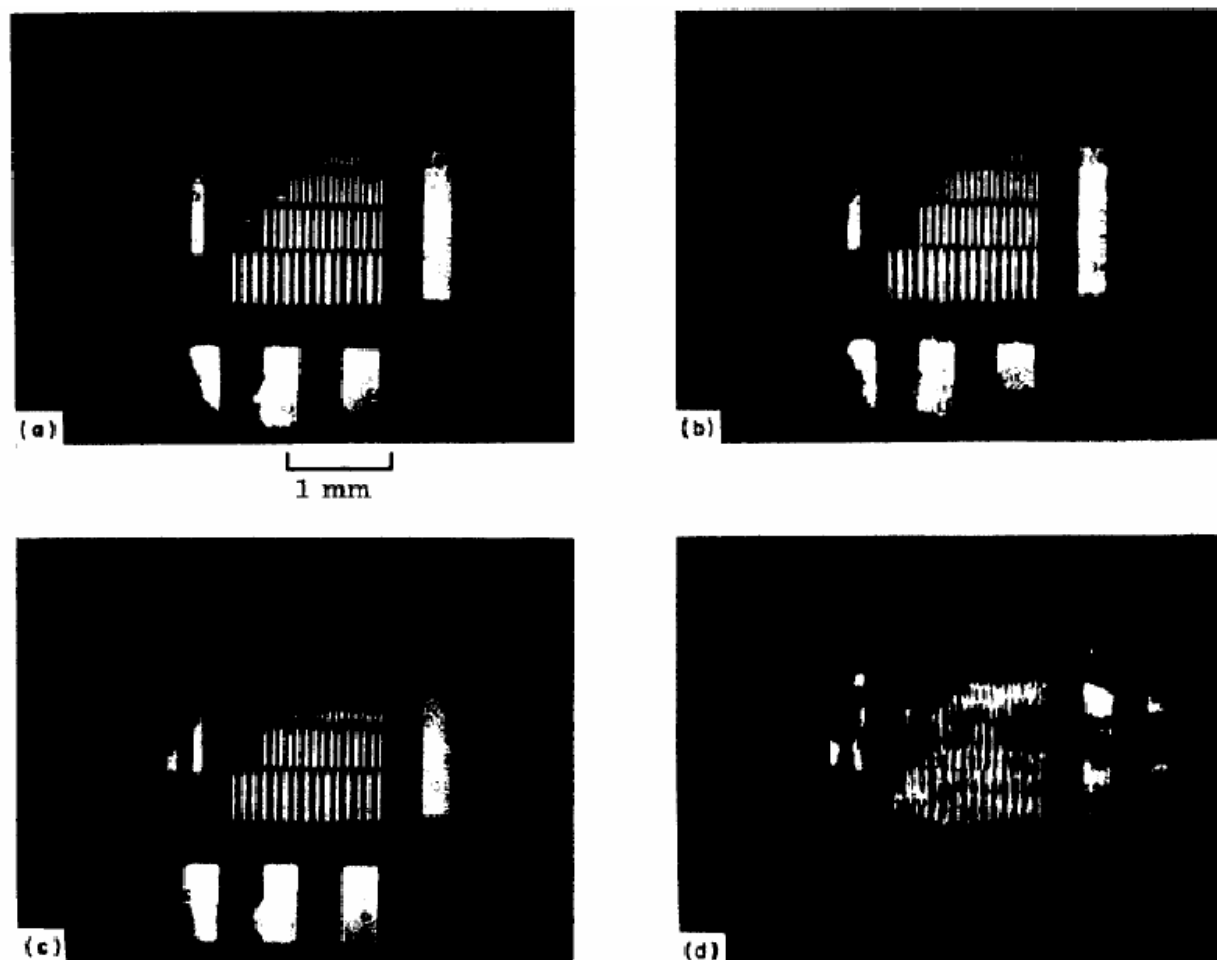
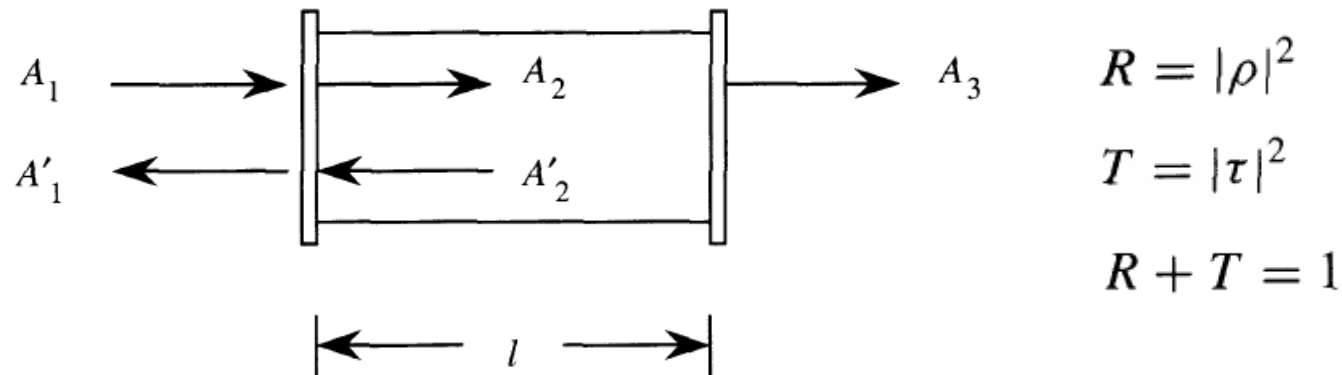


FIG. 2. Photomicrographs of reconstructed images (a) without aberrating glass, (b) with aberrating glass, (c) without aberrating glass but rear mirror misaligned by 0.25 mrad (d) with aberration but rear mirror misaligned by 0.25 mrad.

4.8) Bi-estabilidade e chaveamento óptico



$$A'_2 = \rho A_2 e^{2ikl - \alpha l}$$

$$A_2 = \frac{\tau A_1}{1 - \rho^2 e^{2ikl - \alpha l}}$$

$$A_2 = \tau A_1 + \rho A'_2$$

Bi-estabilidade absorptiva

$$A_2 = \frac{\tau A_1}{1 - \rho^2 e^{2ikl - \alpha l}}$$

Cavidade em ressonância: $kl = \pi$

$$\alpha l \ll 1$$

$$A_2 = \frac{\tau A_1}{1 - R(1 - \alpha l)}$$

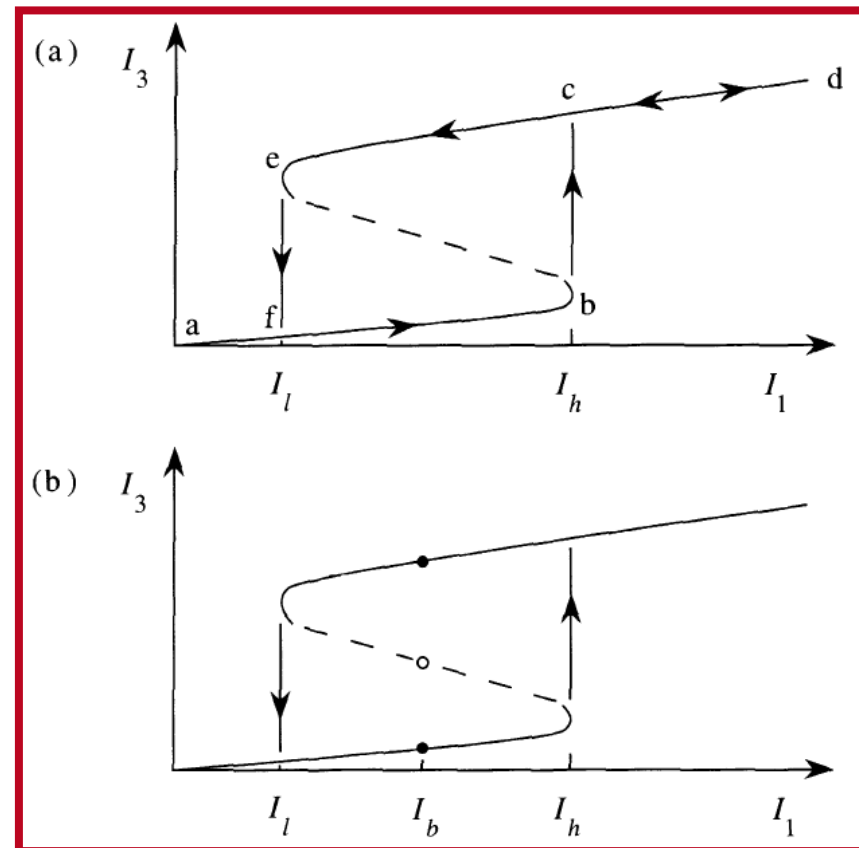
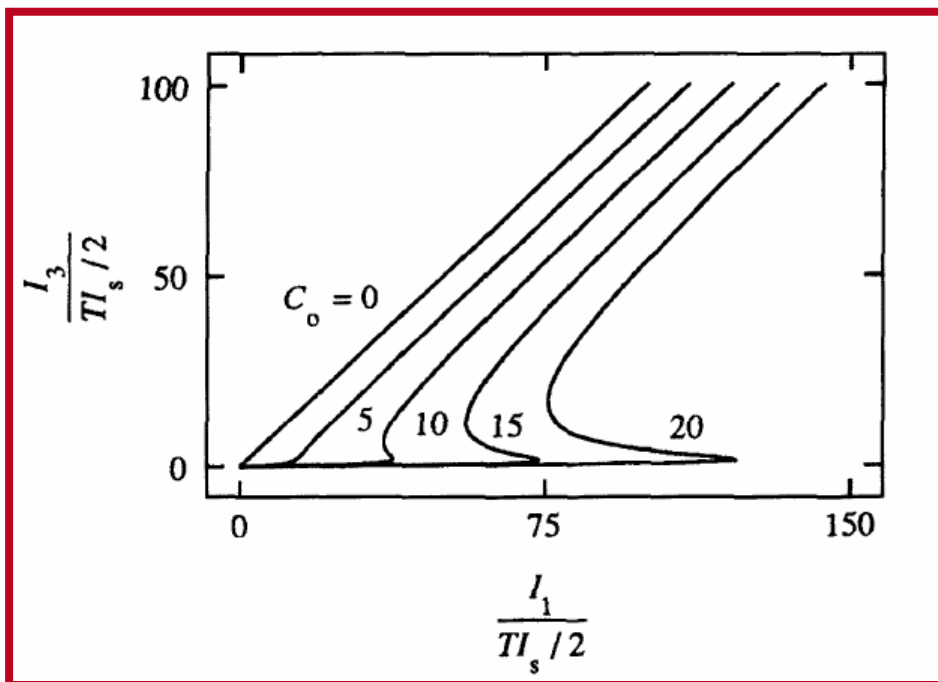
$$I_2 = \frac{TI_1}{[1 - R(1 - \alpha l)]^2}$$

$$\alpha = \frac{\alpha_0}{1 + I/I_s} \quad I_2 + I'_2 \approx 2I_2 = I$$

$$I_1 = TI_2 \left(1 + \frac{C_0}{1 + 2I_2/I_s} \right)^2$$

$$C_0 = R\alpha_0 l / (1 - R)$$

$$I_3 = TI_2$$



Bi-estabilidade refrativa

$$\alpha = 0 \quad \rho^2 = R e^{i\phi}$$

$$A_2 = \frac{\tau A_1}{1 - \rho^2 e^{2ikl}} = \frac{\tau A_1}{1 - R e^{i\delta}}$$

$$\delta = \delta_0 + \delta_2$$

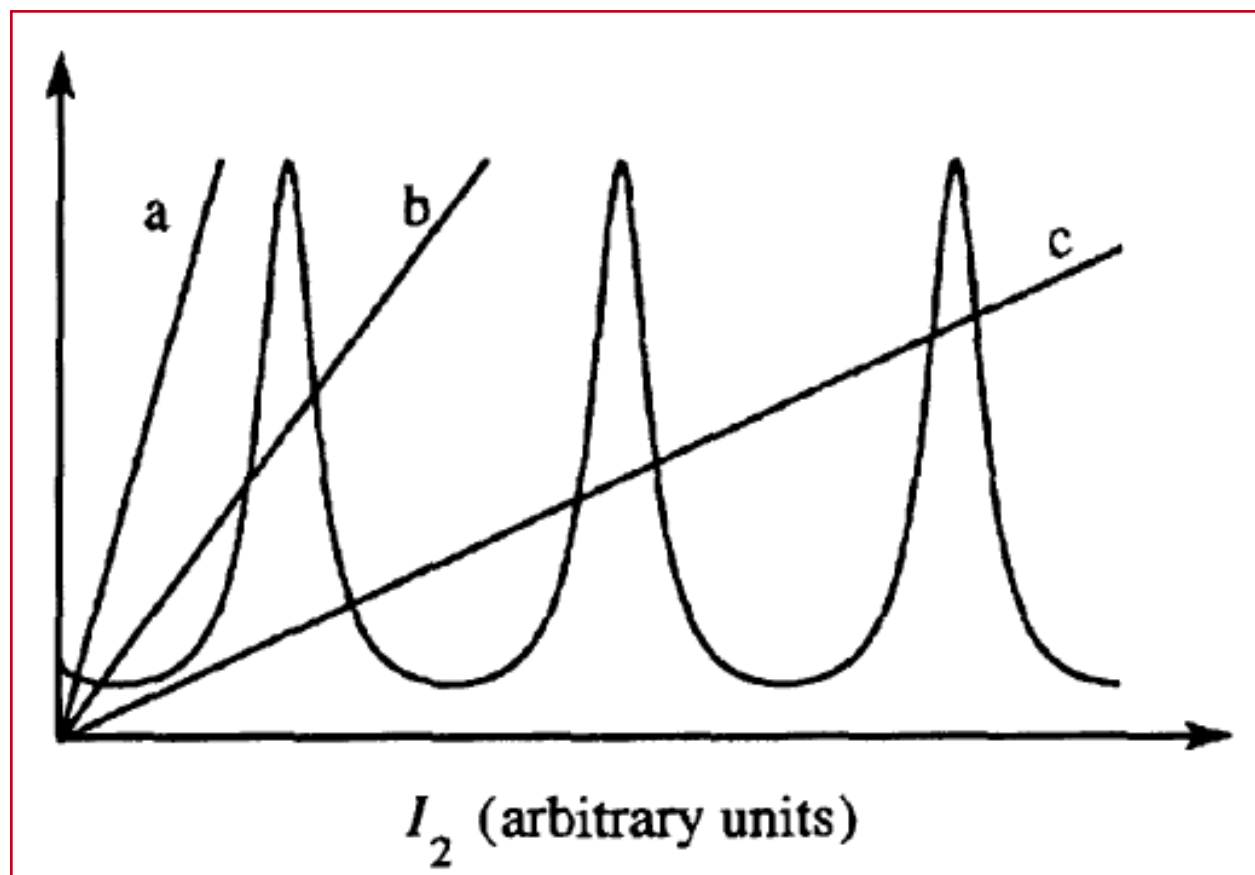
$$\delta_0 = \phi + 2n_0 \frac{\omega}{c} l \quad \delta_2 = 2n_2 l \frac{\omega}{c}$$

$$I = I_2 + I_2' \simeq 2I_2$$

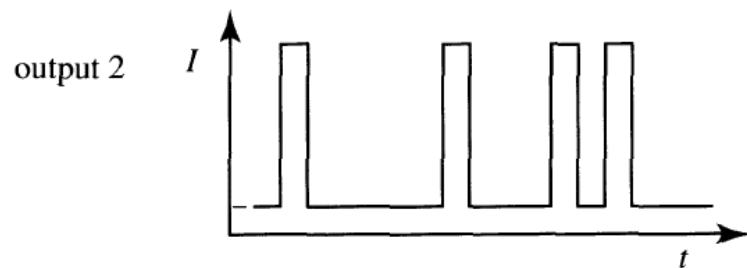
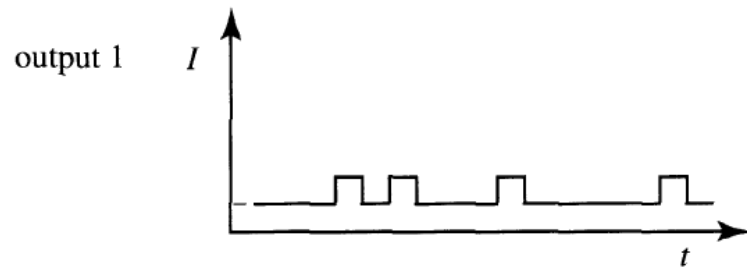
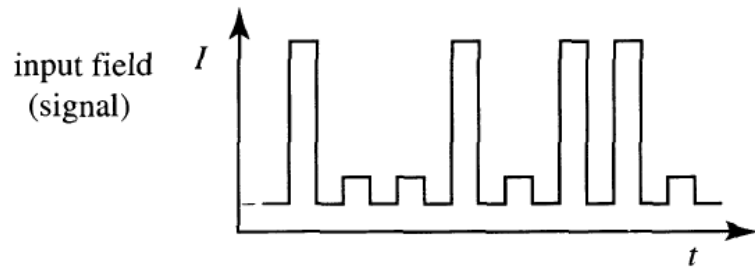
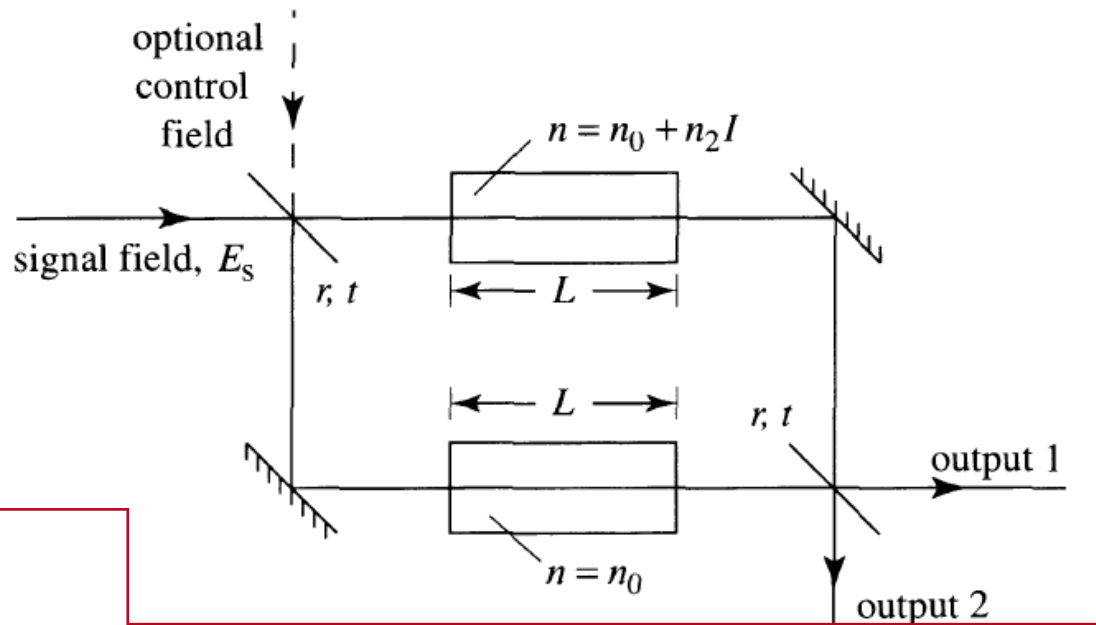
$$\begin{aligned} I_2 &= \frac{T I_1}{(1 - R e^{i\delta})(1 - R e^{-i\delta})} = \frac{T I_1}{1 + R^2 - 2R \cos \delta} \\ &= \frac{T I_1}{(1 - R)^2 + 4R \sin^2 \frac{1}{2} \delta} = \frac{T I_1}{T^2 + 4R \sin^2 \frac{1}{2} \delta} \\ &= \frac{I_1 / T}{1 + (4R / T^2) \sin^2 \frac{1}{2} \delta}, \end{aligned}$$

$$\frac{I_2}{I_1} = \frac{1/T}{1 + (4R/T^2) \sin^2 \frac{1}{2} \delta}$$

$$\delta = \delta_0 + (4n_2\omega l/c) I_2$$



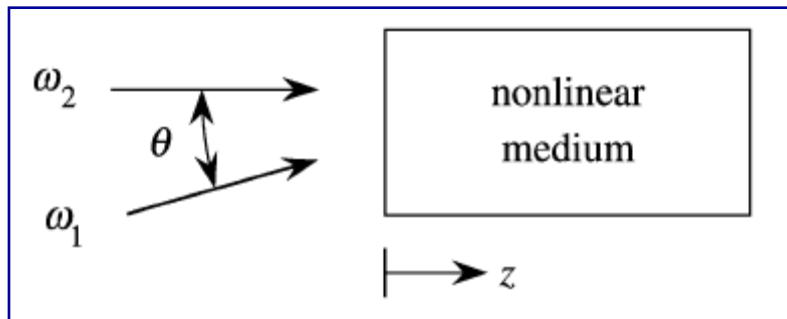
Chaveamento óptico



$$E_1 = E_s (rt + rt e^{i\phi_{NL}})$$

$$\phi_{NL} = n_2(\omega/c)IL = n_2(\omega/c)|t|^2(2n_0\epsilon_0c)|E_s|^2L$$

4.9) Mistura de dois feixes



$$E(\mathbf{r}, t) = A_1 e^{i(\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)} + A_2 e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)} + \text{c.c.}$$

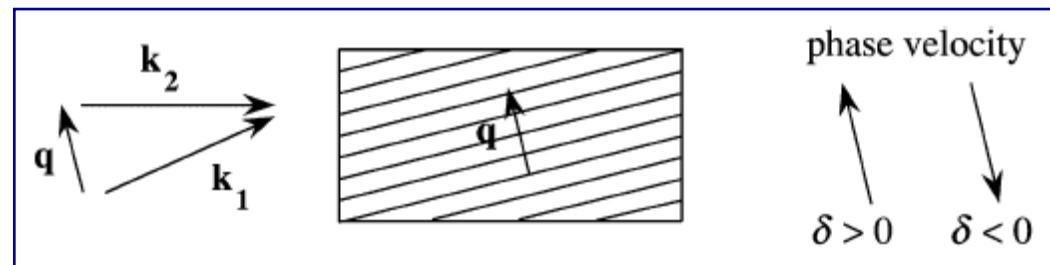
$$I = n_0 \epsilon_0 c \langle E^2 \rangle$$

$$I = 2n_0 \epsilon_0 c \{ A_1 A_1^* + A_2 A_2^* + [A_1 A_2^* e^{i(\mathbf{k}_1 - \mathbf{k}_2) \cdot \mathbf{r} - i(\omega_1 - \omega_2)t} + \text{c.c.}] \}$$

$$= 2n_0 \epsilon_0 c \{ A_1 A_1^* + A_2 A_2^* + [A_1 A_2^* e^{i(\mathbf{q} \cdot \mathbf{r} - \delta t)} + \text{c.c.}] \}$$

$$\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$$

$$\delta = \omega_1 - \omega_2$$



$$\tau \frac{dn_{\text{NL}}}{dt} + n_{\text{NL}} = n_2 I$$

$$n_{\text{NL}} = \frac{n_2}{\tau} \int_{-\infty}^t I(t') e^{(t'-t)/\tau} dt'$$

$$\int_{-\infty}^t e^{-i\delta t'} e^{(t'-t)/\tau} dt' = e^{-t/\tau} \int_{-\infty}^t e^{(-i\delta + 1/\tau)t'} dt' = \frac{e^{-i\delta t}}{-i\delta + 1/\tau}$$

$$n_{\text{NL}} = 2n_0 n_2 \epsilon_0 c \left[(A_1 A_1^* + A_2 A_2^*) + \frac{A_1 A_2^* e^{i(\mathbf{q} \cdot \mathbf{r} - \delta t)}}{1 - i\delta\tau} + \frac{A_1^* A_2 e^{-i(\mathbf{q} \cdot \mathbf{r} - \delta t)}}{1 + i\delta\tau} \right]$$

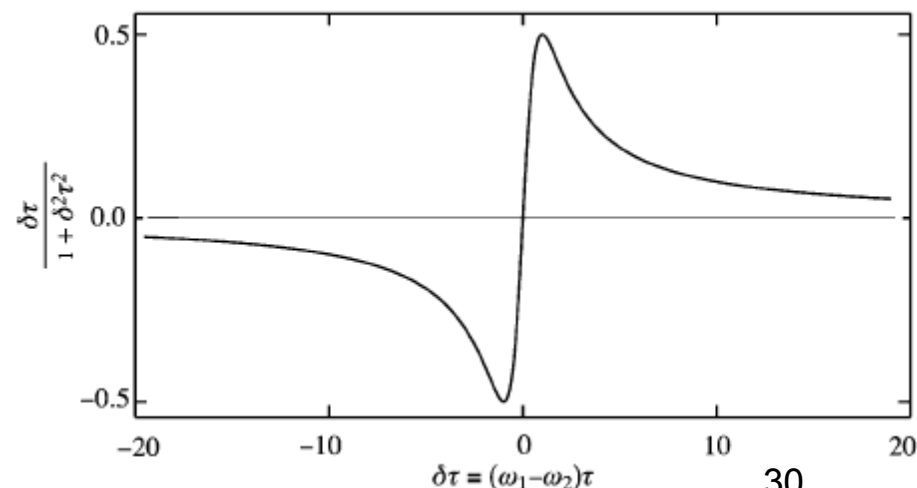
$$\nabla^2 E - \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = 0$$

$$n^2 = n_0^2 + 2n_0 n_{\text{NL}}$$

$$\frac{dA_2}{dz} = 2in_0 n_2 (\omega/c) \left[(|A_1|^2 + |A_2|^2) A_2 + \frac{|A_1|^2 A_2}{1 + i\delta\tau} \right]$$

$$\frac{dI_2}{dz} = 2n_0 \epsilon_0 c \left(A_2^* \frac{dA_2}{dz} + A_2 \frac{dA_2^*}{dz} \right)$$

$$\frac{dI_2}{dz} = \frac{2n_2 \omega}{c} \frac{\delta\tau}{1 + \delta^2 \tau^2} I_1 I_2$$



4.10) Propagação de pulsos e sólitons temporais

Automodulação de fase

$$E(z, t) = A(z, t)e^{i(k_0 z - \omega_0 t)} + \text{c.c.}$$

$$n(t) = n_0 + n_2 I(t)$$

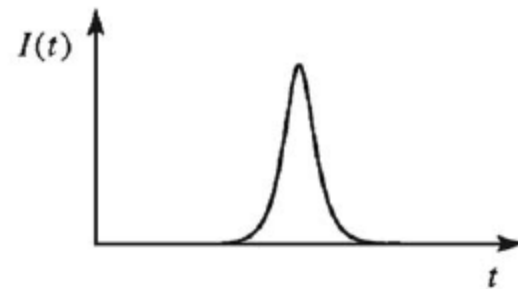
$$I(t) = 2n_0 \epsilon_0 c |A(z, t)|^2$$

$$\phi_{\text{NL}}(t) = -n_2 I(t) \omega_0 L / c$$

$$\omega(t) = \omega_0 + \delta\omega(t) \quad \delta\omega(t) = \frac{d}{dt} \phi_{\text{NL}}(t)$$

Exemplo:

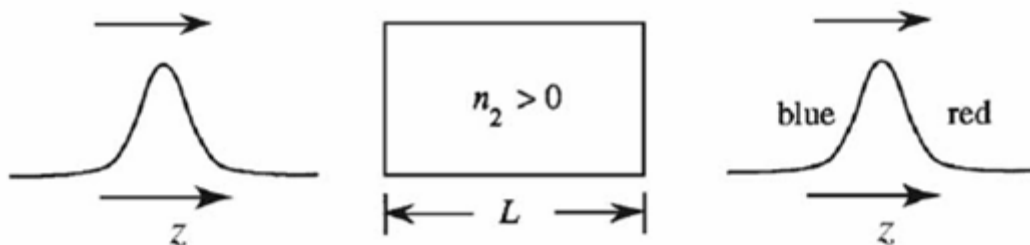
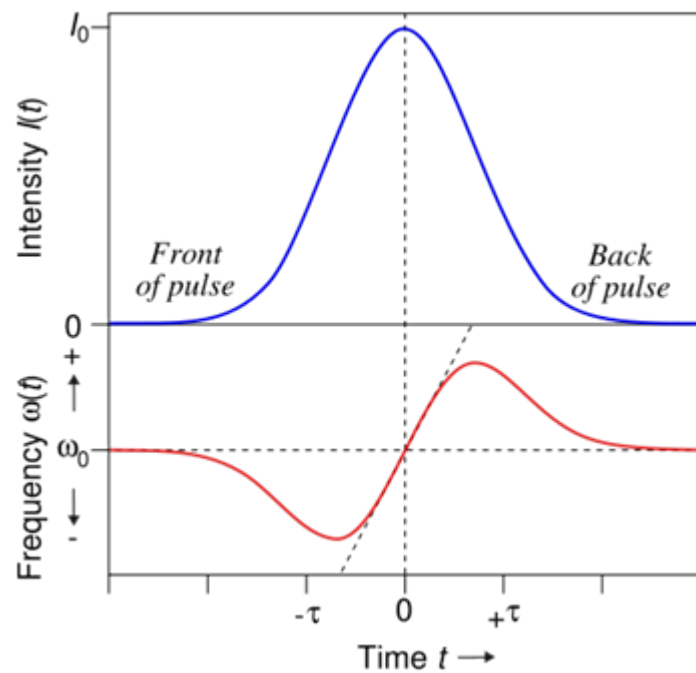
$$I(t) = I_0 \text{sech}^2(t/\tau_0)$$



$$\phi_{\text{NL}}(t) = -n_2 \frac{\omega_0}{c} L I_0 \text{sech}^2(t/\tau_0)$$

$$\delta\omega(t) = \frac{d}{dt}\phi_{\text{NL}}(t)$$

$$\delta\omega(t) = 2n_2 \frac{\omega_0}{c\tau_0} L I_0 \text{sech}^2(t/\tau_0) \tanh(t/\tau_0)$$



Equação de propagação do pulso

$$E(z, t) = A(z, t)e^{i(k_0 z - \omega_0 t)} + \text{c.c.}$$

$$E(z, t) = \int_{-\infty}^{\infty} E(z, \omega) e^{-i\omega t} \frac{d\omega}{2\pi} \quad A(z, \omega') = \int_{-\infty}^{\infty} A(z, t) e^{i\omega' t} dt$$

$$E(z, \omega) = A(z, \omega - \omega_0) e^{ik_0 z} + A^*(z, \omega + \omega_0) e^{-ik_0 z} \simeq A(z, \omega - \omega_0) e^{ik_0 z}$$

$$\frac{\partial^2 E(z, \omega)}{\partial z^2} + \epsilon(\omega) \frac{\omega^2}{c^2} E(z, \omega) = 0$$

$$2ik_0 \frac{\partial A}{\partial z} + (k^2 - k_0^2) A = 0$$

$$k(\omega) = \sqrt{\epsilon(\omega)} \omega / c$$

$$k^2 - k_0^2 \approx 2k_0(k - k_0)$$

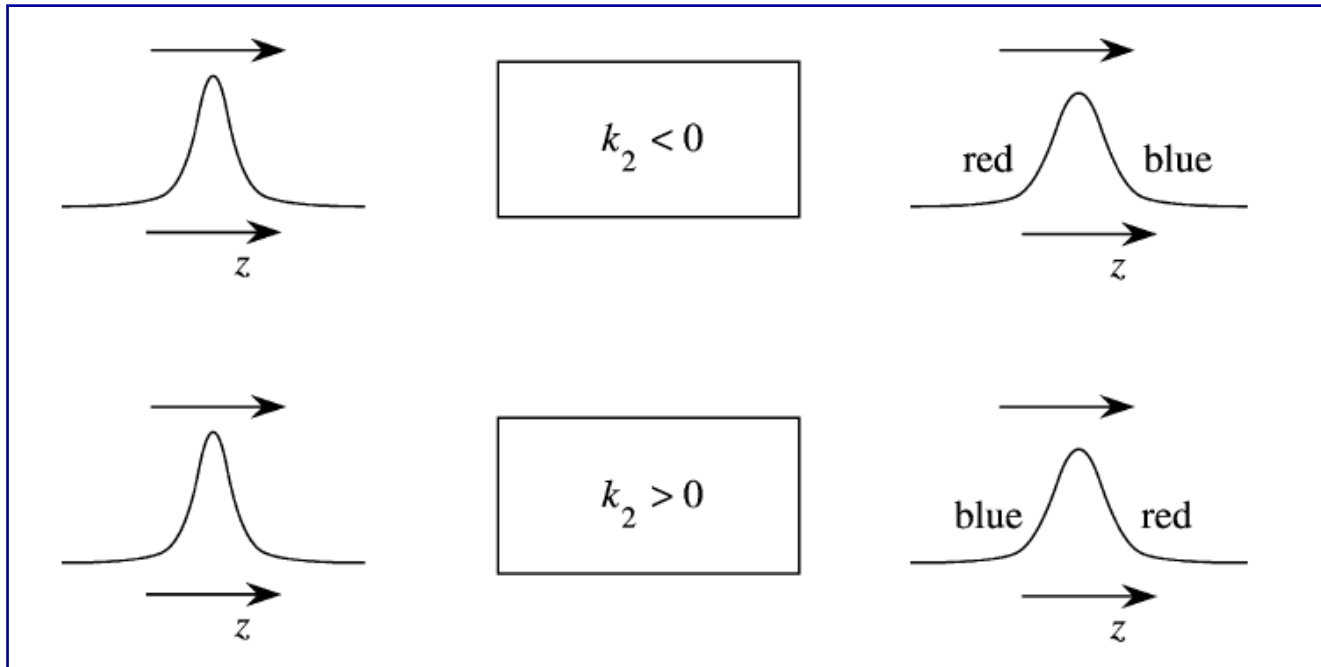
$$\frac{\partial A(z, \omega - \omega_0)}{\partial z} - i(k - k_0) A(z, \omega - \omega_0) = 0$$

$$k = k_0 + \Delta k_{\text{NL}} + k_1(\omega - \omega_0) + \frac{1}{2}k_2(\omega - \omega_0)^2$$

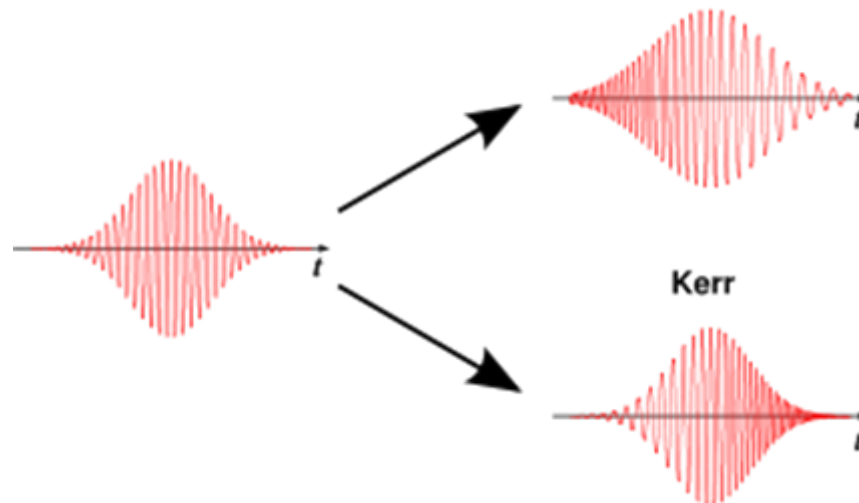
$$\Delta k_{\text{NL}} = \Delta n_{\text{NL}}\omega_0/c = n_2 I \omega_0/c$$

$$k_1 = \left(\frac{dk}{d\omega} \right)_{\omega=\omega_0} = \frac{1}{c} \left[n_{\text{lin}}(\omega) + \omega \frac{dn_{\text{lin}}(\omega)}{d\omega} \right]_{\omega=\omega_0} \equiv \frac{1}{v_g(\omega_0)}$$

$$k_2 = \left(\frac{d^2k}{d\omega^2} \right)_{\omega=\omega_0} = \frac{d}{d\omega} \left[\frac{1}{v_g(\omega)} \right]_{\omega=\omega_0} = \left(-\frac{1}{v_g^2} \frac{dv_g}{d\omega} \right)_{\omega=\omega_0}$$



GVD



$$\frac{\partial A}{\partial z} - i \Delta k_{\text{NL}} A - i k_1 (\omega - \omega_0) A - \frac{1}{2} i k_2 (\omega - \omega_0)^2 A = 0$$

we multiply each term by the factor $\exp[-i(\omega - \omega_0)t]$ and integrate

$$\int_{-\infty}^{\infty} A(z, \omega - \omega_0) e^{-i(\omega - \omega_0)t} \frac{d(\omega - \omega_0)}{2\pi} = A(z, t)$$

$$\int_{-\infty}^{\infty} (\omega - \omega_0) A(z, \omega - \omega_0) e^{-i(\omega - \omega_0)t} \frac{d(\omega - \omega_0)}{2\pi}$$

$$= \frac{1}{-i} \frac{\partial}{\partial t} \int_{-\infty}^{\infty} A(z, \omega - \omega_0) e^{-i(\omega - \omega_0)t} \frac{d(\omega - \omega_0)}{2\pi} = i \frac{\partial}{\partial t} A(z, t)$$

$$\int_{-\infty}^{\infty} (\omega - \omega_0)^2 A(z, \omega - \omega_0) e^{-i(\omega - \omega_0)t} \frac{d(\omega - \omega_0)}{2\pi} = -\frac{\partial^2}{\partial t^2} A(z, t)$$

$$\frac{\partial A}{\partial z} + k_1 \frac{\partial A}{\partial t} + \frac{1}{2} i k_2 \frac{\partial^2 A}{\partial t^2} - i \Delta k_{\text{NL}} A = 0$$

$$\tau = t - \frac{z}{v_g} = t - k_1 z \quad A_s(z, \tau) = A(z, t)$$

$$\frac{\partial A}{\partial z} = \frac{\partial A_s}{\partial z} + \frac{\partial A_s}{\partial \tau} \frac{\partial \tau}{\partial z} = \frac{\partial A_s}{\partial z} - k_1 \frac{\partial A_s}{\partial \tau} \quad \frac{\partial A}{\partial t} = \frac{\partial A_s}{\partial z} \frac{\partial z}{\partial t} + \frac{\partial A_s}{\partial \tau} \frac{\partial \tau}{\partial t} = \frac{\partial A_s}{\partial \tau}$$

$$\frac{\partial A_s}{\partial z} + \frac{1}{2} i k_2 \frac{\partial^2 A_s}{\partial \tau^2} - i \Delta k_{\text{NL}} A_s = 0 \quad \Delta k_{\text{NL}} = n_2 \frac{\omega_0}{c} I = 2 n_0 \epsilon_0 n_2 \omega_0 |A_s|^2 \equiv \gamma |A_s|^2$$

$$\frac{\partial A_s}{\partial z} + \frac{1}{2} i k_2 \frac{\partial^2 A_s}{\partial \tau^2} = i \gamma |A_s|^2 A_s$$

nonlinear Schrödinger equation

Sólitons temporais ópticos

$$\frac{\partial A_s}{\partial z} + \frac{1}{2}ik_2 \frac{\partial^2 A_s}{\partial \tau^2} = i\gamma |A_s|^2 A_s$$

$$A_s(z, \tau) = A_s^0 \operatorname{sech}(\tau/\tau_0) e^{i\kappa z}$$

$$|A_s^0|^2 = \frac{-k_2}{\gamma \tau_0^2} = \frac{-k_2}{2n_0 \epsilon_0 n_2 \omega_0 \tau_0^2}$$

$$\kappa = -k_2/2\tau_0^2 = \frac{1}{2}\gamma |A_s^0|^2$$

Note that condition (7.5.33b) shows that k_2 and n_2 must have opposite signs in order for Eq. (7.5.33a) to represent a physical pulse in which the intensity $|A_s^0|^2$ and the square of the pulse width τ_0^2 are both positive. We can see from Eq. (7.5.32) that in fact k_2 and γ must have opposite signs in order for group velocity dispersion to compensate for self-phase modulation (because $\tilde{A}_s^{-1}(\partial^2 \tilde{A}_s / \partial \tau^2)$ will be negative near the peak of the pulse, where the factor $|\tilde{A}_s|^2 \tilde{A}_s$ is most important).

One circumstance under which k_2 and γ have opposite signs occurs in fused-silica optical fibers. In this case, the nonlinearity in the refractive index occurs as the result of electronic polarization, and n_2 is consequently positive. The group velocity dispersion parameter k_2 is positive for visible light but becomes negative for wavelengths longer than approximately $1.3 \mu\text{m}$. This effect is illustrated in Fig. 7.5.3, in which the linear refractive index n_{lin} and the group index $n_g \equiv c/v_g$ are plotted as functions of the vacuum wavelength of the incident radiation. Optical solitons of the sort described by Eq. (7.5.33a) have been observed by Mollenauer *et al.* (1980) in the propagation of light pulses at a wavelength of $1.55 \mu\text{m}$ obtained from a color center laser.

