

Chapter 21 (21.6-21.7 excluded)

# **NONLINEAR OPTICS II**

# Coupled-wave theory of three-wave mixing

Second-order nonlinear medium:  $\nabla^2 \mathcal{E} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = -\mathcal{S}$

$$\mathcal{S} = -\mu_0 \frac{\partial^2 \mathcal{P}_{\text{NL}}}{\partial t^2}, \quad \mathcal{P}_{\text{NL}}(t) = 2\mathbf{d}\mathcal{E}^2$$

$$\begin{aligned} \mathcal{E}(t) &= \sum_{q=\pm 1, \pm 2, \pm 3} \frac{1}{2} E_q \exp(j\omega_q t) \Rightarrow \mathcal{P}_{\text{NL}}(t) = 2\mathbf{d} \cdot \frac{1}{4} \sum_{q,r=\pm 1, \pm 2, \pm 3} E_q E_r \exp[j(\omega_q + \omega_r)t] \\ &\Rightarrow \mathcal{S} = \frac{1}{2} \mu_0 \mathbf{d} \sum_{q,r=\pm 1, \pm 2, \pm 3} (\omega_q + \omega_r)^2 E_q E_r \exp[j(\omega_q + \omega_r)t] \end{aligned}$$

For distinct frequencies satisfying  $\omega_1 + \omega_2 = \omega_3$ , we separate the wave equations:

$$\begin{aligned} (\nabla^2 + k_1^2) E_1 &= -S_1 = -2\mu_0 \omega_1^2 \mathbf{d} E_3 E_2^* \\ (\nabla^2 + k_2^2) E_2 &= -S_2 = -2\mu_0 \omega_2^2 \mathbf{d} E_3 E_1^* \\ (\nabla^2 + k_3^2) E_3 &= -S_3 = -2\mu_0 \omega_3^2 \mathbf{d} E_1 E_2. \end{aligned}$$

For *collinear waves* within the *slowly varying envelope* approximation, we obtain

$$\begin{aligned} \frac{da_1}{dz} &= -jg a_3 a_2^* \exp(-j\Delta k z) \\ \frac{da_2}{dz} &= -jg a_3 a_1^* \exp(-j\Delta k z) \\ \frac{da_3}{dz} &= -jg a_1 a_2 \exp(j\Delta k z) \end{aligned}$$

$$E_q = \sqrt{2\eta\hbar\omega_q} \mathbf{a}_q \exp(-jk_q z)$$

$$\phi_q = \frac{I_q}{\hbar\omega_q} = |\mathbf{a}_q|^2$$

$$g^2 = 2\hbar\omega_1\omega_2\omega_3\eta^3 \mathbf{d}^2$$

$$\Delta k = k_3 - k_2 - k_1 \leftarrow \text{mismatch}$$

# Second-harmonic generation

A degenerate case:  $\omega_1 = \omega_2 = \omega$ ,  $\omega_3 = 2\omega$  and  $\mathbf{k}_3 = 2\mathbf{k}_1$ .

For collinear waves with perfect phase matching,  $\Delta k = 0$ , we have

$$\begin{cases} \frac{da_1}{dz} = -jga_3a_2^* \exp(-j\Delta k z) \\ \frac{da_2}{dz} = -jga_3a_1^* \exp(-j\Delta k z) \\ \frac{da_3}{dz} = -jga_1a_2 \exp(j\Delta k z) \end{cases} \Rightarrow \begin{cases} \frac{da_1}{dz} = -jga_3a_1^* \\ \frac{da_3}{dz} = -j\frac{g}{2}a_1a_1 \\ \underline{a_1^2(z) + 2|a_3(z)|^2 = a_1^2(0)} \end{cases}$$

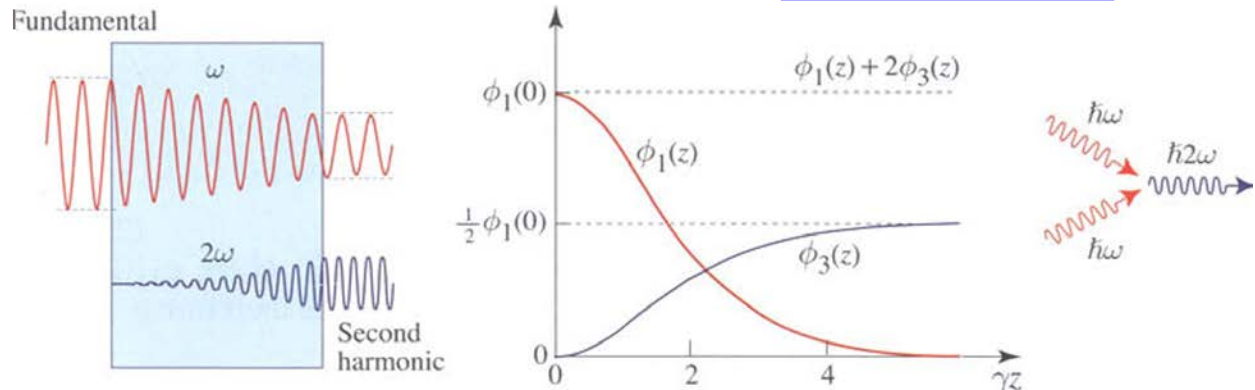
The solutions:  $\begin{cases} a_1(z) = a_1(0) \operatorname{sech}\left(\frac{1}{\sqrt{2}}ga_1(0)z\right) \\ a_3(z) = -\frac{j}{\sqrt{2}}a_1(0) \tanh\left(\frac{1}{\sqrt{2}}ga_1(0)z\right) \end{cases} \Rightarrow \begin{cases} \phi_1(z) = \phi_1(0) \operatorname{sech}^2\frac{\gamma z}{2} \\ \phi_3(z) = \frac{1}{2}\phi_1(0) \tanh^2\frac{\gamma z}{2} \end{cases}$

SHG efficiency:

$$\underline{\gamma/2 = ga_1(0)/\sqrt{2}}$$

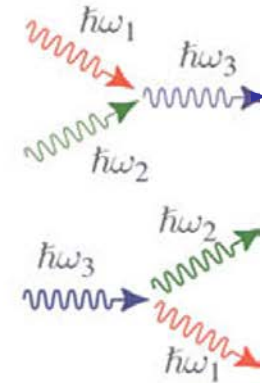
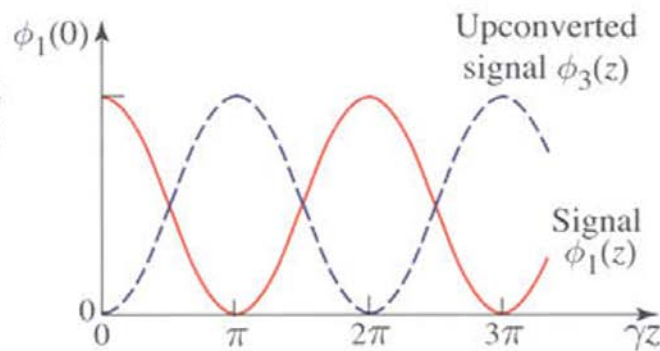
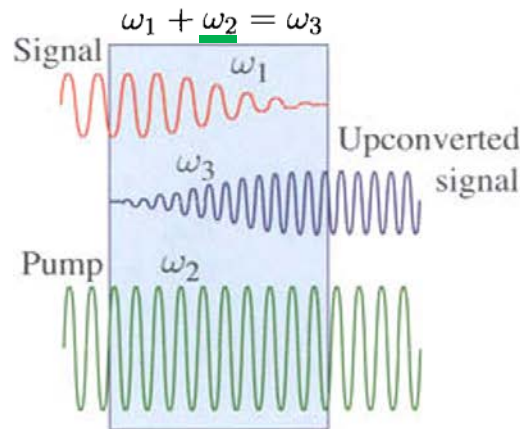
$$\eta_{\text{SHG}} \equiv \frac{I_3(L)}{I_1(0)}$$

$$\underline{\eta_{\text{SHG}} = \tanh^2\frac{\gamma L}{2}}$$



If  $\Delta k \neq 0$ ,  $\eta_{\text{SHG}}$  is reduced by  $\operatorname{sinc}^2(\Delta k L/2\pi)$ .

# Frequency conversion



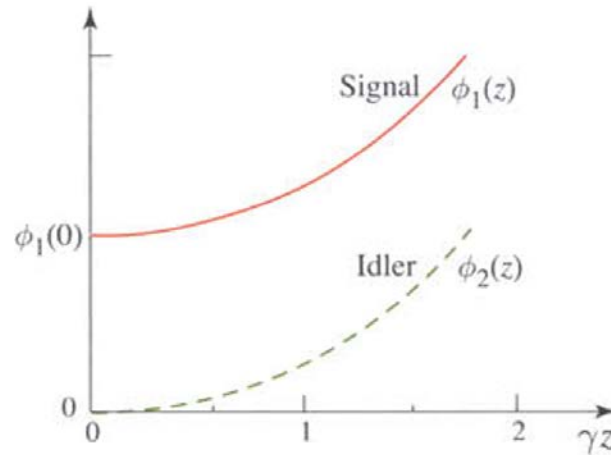
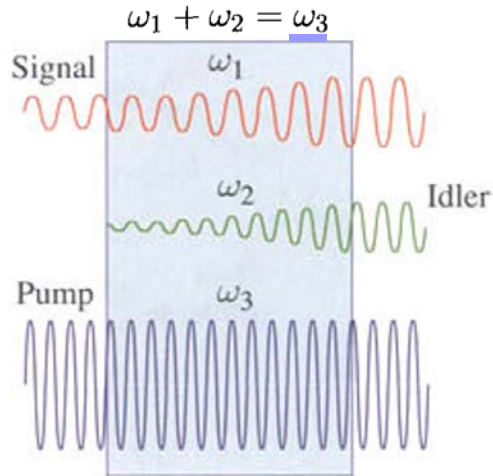
$$\begin{aligned}
 \frac{da_1}{dz} &= -jga_3a_2^* \exp(-j\Delta k z) \\
 \frac{da_2}{dz} &= -jga_3a_1^* \exp(-j\Delta k z) \\
 \frac{da_3}{dz} &= -jga_1a_2 \exp(j\Delta k z)
 \end{aligned}
 \Rightarrow
 \begin{cases}
 \frac{da_1}{dz} = -j\frac{\gamma}{2}a_3 \\
 \frac{da_3}{dz} = -j\frac{\gamma}{2}a_1 \\
 \text{when } a_2(z) \approx a_2(0)
 \end{cases}
 \Rightarrow
 \begin{cases}
 a_1(z) = a_1(0) \cos^2 \frac{\gamma z}{2} \\
 a_3(z) = -ja_1(0) \sin^2 \frac{\gamma z}{2}
 \end{cases}$$

$$\Rightarrow
 \begin{cases}
 \phi_1(z) = \phi_1(0) \cos^2 \frac{\gamma z}{2} \\
 \phi_3(z) = \phi_1(0) \sin^2 \frac{\gamma z}{2}
 \end{cases}$$

The efficiency of up-conversion:

$$\eta_{\text{OFC}} = \frac{I_3(L)}{I_1(0)} = \frac{\omega_3}{\omega_1} \sin^2 \frac{\gamma L}{2} \xrightarrow{\gamma L \ll 1} \eta_{\text{OFC}} = C^2 \frac{L^2}{A} P_2, \quad C^2 = 2\omega_3^2 \eta_o^3 \frac{d^2}{n^3}$$

# Optical parametric amplification (OPA)



$$\begin{aligned} \frac{da_1}{dz} &= -jga_3a_2^* \exp(-j\Delta k z) \\ \frac{da_2}{dz} &= -jga_3a_1^* \exp(-j\Delta k z) \\ \frac{da_3}{dz} &= -jga_1a_2 \exp(j\Delta k z) \end{aligned} \Rightarrow \begin{cases} \frac{da_1}{dz} = -j\frac{\gamma}{2}a_2^* \\ \frac{da_2}{dz} = -j\frac{\gamma}{2}a_1^* \\ \text{when } a_3(z) \approx a_3(0) \end{cases} \Rightarrow \begin{cases} a_1(z) = a_1(0) \cosh \frac{\gamma z}{2} \\ a_2(z) = -ja_1^*(0) \sinh \frac{\gamma z}{2} \end{cases}$$

$$\Rightarrow \begin{cases} \phi_1(z) = \phi_1(0) \cosh^2 \frac{\gamma z}{2} \\ \phi_2(z) = \phi_1(0) \sinh^2 \frac{\gamma z}{2} \end{cases}$$

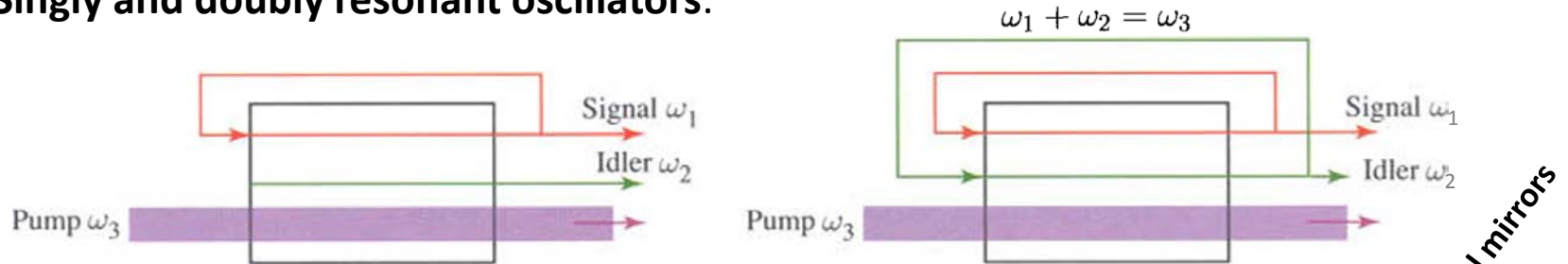
The gain coefficient:

$$G = \phi_1(L)/\phi_1(0) = \cosh^2(\gamma L/2) \approx e^{\gamma L}/4, \text{ if } \gamma L \gg 1.$$

$$\text{Here, } \gamma = 2C\sqrt{I_3(0)} = 2C\sqrt{P_3/A} \text{ and } C^2 = 2\omega_1\omega_2\eta_o^3 \frac{d^2}{n^3}.$$

# Optical parametric oscillator (OPO)

Singly and doubly resonant oscillators:



**Threshold of SRO:** One round-trip loss is equal to gain  $\Rightarrow a_1(L) r_1^2 = a_1(0)$

$$\Rightarrow r_1^2 \cosh(\gamma L/2) = 1 \Rightarrow \mathcal{R}_1^2 \cosh^2(\gamma L/2) = 1, \text{ where } \mathcal{R}_1 = r_1^2.$$

At small  $x$ ,  $\cosh^2(x) \approx 1 + x^2 \Rightarrow (\gamma L/2)^2 \approx (1 - \mathcal{R}_1^2)/\mathcal{R}_1^2$ . As  $\gamma = 2C\sqrt{P_3/A}$ ,

$$P_3|_{\text{threshold}}(0) \approx \frac{1}{C^2} \frac{A}{L^2} \frac{1 - \mathcal{R}_1^2}{\mathcal{R}_1^2},$$

$$C^2 = 2\omega_1\omega_2 \eta_o^3 d^2/n^3.$$

**Threshold of DRO:**  $a_1(L) r_1^2 = a_1(0)$  and  $a_2(L) r_2^2 = a_2(0)$ . These conditions lead to

$$\tanh^2(\gamma L/2) = (1 - \mathcal{R}_1)(1 - \mathcal{R}_2)/(\mathcal{R}_1\mathcal{R}_2) \approx (\gamma L/2)^2$$

$$\Rightarrow P_3|_{\text{threshold}}(0) \approx \frac{1}{C^2} \frac{A}{L^2} \frac{(1 - \mathcal{R}_1)(1 - \mathcal{R}_2)}{\mathcal{R}_1\mathcal{R}_2} \ll \text{than for SRO}$$

# Coupled-wave theory of four-wave mixing

In third-order nonlinear medium,  $\mathcal{P}_{NL} = 4\chi^{(3)}\mathcal{E}^3$  and  $\mathcal{E}(t) = \sum_{q=\pm 1, \pm 2, \pm 3, \pm 4} \frac{1}{2}E_q \exp(j\omega_q t)$ .

$$\Rightarrow \mathcal{S} = -\mu_o \partial^2 \mathcal{P}_{NL} / \partial t^2 = \frac{1}{2} \mu_o \chi^{(3)} \sum_{q,p,r=\pm 1, \pm 2, \pm 3, \pm 4} (\omega_q + \omega_p + \omega_r)^2 E_q E_p E_r \exp[j(\omega_q + \omega_p + \omega_r)t].$$

The three coupled Helmholtz equations are

$$(\nabla^2 + k_q^2)E_q = -S_q, \quad q = 1, 2, 3, 4$$

Example of  $\omega_1 + \omega_2 = \omega_3 + \omega_4$ :

$$\begin{cases} S_1 = \mu_o \omega_1^2 \chi^{(3)} \{6E_3 E_4 E_2^* + 3E_1[|E_1|^2 + 2|E_2|^2 + 2|E_3|^2 + 2|E_4|^2]\} \\ S_2 = \mu_o \omega_2^2 \chi^{(3)} \{6E_3 E_4 E_1^* + 3E_2[|E_2|^2 + 2|E_1|^2 + 2|E_3|^2 + 2|E_4|^2]\} \\ S_3 = \mu_o \omega_3^2 \chi^{(3)} \{6E_1 E_2 E_4^* + 3E_3[|E_3|^2 + 2|E_2|^2 + 2|E_1|^2 + 2|E_4|^2]\} \\ S_4 = \mu_o \omega_4^2 \chi^{(3)} \{6E_1 E_2 E_3^* + 3E_4[|E_4|^2 + 2|E_1|^2 + 2|E_2|^2 + 2|E_3|^2]\} \end{cases}$$

$$\Rightarrow \begin{cases} S_q = \bar{S}_q + (\omega_q/c_o)^2 \Delta \chi_q E_q \\ \bar{S}_q = 6\mu_o \omega^2 \chi^{(3)} E_l E_n E_m^* \\ \Delta \chi_q = 6 \frac{\eta}{\epsilon_o} \chi^{(3)} (2I - I_q) \end{cases}$$

↑  
total

$$\Rightarrow \begin{cases} (\nabla^2 + \bar{k}_q^2)E_q = -\bar{S}_q \\ \bar{k}_q = \bar{n}_q \frac{\omega_q}{c_o}, \quad n_2 = \frac{3\eta_o}{\epsilon_o n^2} \chi^{(3)} \\ \bar{n}_q \approx n + n_2(2I - I_q) \end{cases}$$

Optical Kerr effect

# Three-wave mixing

If  $\omega_3 = \omega_4 = \omega_0$ , then  $\omega_1 + \omega_2 = 2\omega_0$  and

$$\begin{cases} S_1 = \mu_o \omega_1^2 \chi^{(3)} \{ 3E_0^2 E_2^* + 3E_1 [ |E_1|^2 + 2|E_2|^2 + 2|E_0|^2 ] \} \\ S_2 = \mu_o \omega_2^2 \chi^{(3)} \{ 3E_0^2 E_1^* + 3E_2 [ |E_2|^2 + 2|E_1|^2 + 2|E_0|^2 ] \} \\ S_0 = \mu_o \omega_0^2 \chi^{(3)} \{ 6E_1 E_2 E_0^* + 3E_0 [ |E_0|^2 + 2|E_1|^2 + 2|E_2|^2 ] \} \end{cases}$$

$$(\nabla^2 + k_q^2)E_q = -S_q \Rightarrow (\nabla^2 + k_q^2)[A_q \exp(-jk_q z)] \approx -j2k_q(dA_q/dz) \exp(-jk_q z)$$

Introducing  $g = \hbar\omega_0(\omega_0/c_o)n_2$ , we obtain

$$\begin{cases} \frac{da_1}{dz} = -jg [a_0^2 a_2^* \exp(-j\Delta k z) + a_1 (|a_1|^2 + 2|a_2|^2 + 2|a_0|^2)] \\ \frac{da_2}{dz} = -jg [a_0^2 a_1^* \exp(-j\Delta k z) + a_2 (|a_2|^2 + 2|a_1|^2 + 2|a_0|^2)] \\ \frac{da_0}{dz} = -jg [2a_1 a_2 a_0^* \exp(j\Delta k z) + a_0 (|a_0|^2 + 2|a_1|^2 + 2|a_2|^2)] \end{cases} \quad \begin{aligned} I_q &= |a_q|^2 \\ \phi_q &= \hbar\omega_q \end{aligned}$$

Undepleted pump approximation,  $a_0(z) \approx const$ , and perfect phase matching,  $\Delta k = 0$ :

$$\begin{cases} \frac{da_1}{dz} = -j\gamma(a_2^* + 2a_1) \\ \frac{da_2}{dz} = -j\gamma(a_1^* + 2a_2) \end{cases} \Rightarrow \begin{cases} a_1(z) = [(1 - j\gamma z)a_1(0) - j\gamma z a_2^*(0)] \exp(-j\gamma z) \\ a_2(z) = [-j\gamma z a_1^*(0) + (1 - j\gamma z)a_2(0)] \exp(-j\gamma z). \end{cases}$$

If  $a_2(0) = 0$ , we have  $|a_1(z)|^2 = \phi_1(z) = (1 + \gamma^2 z^2)\phi_1(0)$ .

Otherwise, the amplification depends on  $\Delta\varphi(0) = \varphi_1 - \varphi_2$ .

$\gamma = g a_0^2$



# Third-harmonic generation

If  $\omega_1 = \omega_2 = \omega_4 = \omega$  and  $\omega_3 = \omega_1 + \omega_2 + \omega_4 = 3\omega$ , we obtain two Helmholtz equations:

$$(\nabla^2 + k_q^2)E_q = -S_q,$$

where

$$S_1 = \mu_o \omega_1^2 \chi^{(3)} \{ 3E_3 E_1^* E_1^* + 3E_1 [ |E_1|^2 + 2|E_3|^2 ] \}$$

$$S_3 = \mu_o \omega_3^2 \chi^{(3)} \{ E_1^3 + 3E_3 [ |E_3|^2 + 2|E_1|^2 ] \}.$$

The *undepleted-pump* and the *slowly varying envelope approximations* yield

$$\frac{da_3}{dz} = -jg a_1^3 \exp(-j\Delta k z),$$

where  $A_q = \sqrt{2\eta\hbar\omega_q} a_q$ ,  $g = \hbar\omega_1^{3/2}\omega_3^{1/2}\eta^3\chi^{(3)}$  and  $\Delta k = 3k_1 - k_3$ .

The solution is

$$a_3 = -g a_1^3 \frac{1 - e^{-j\Delta k z}}{\Delta k}.$$

The photon flux density,  $\phi_3 = |a_3|^2$ , is

$$\phi_3 = g^2 \phi_1^3 z^2 \text{sinc}^2 \left( \frac{\Delta k z}{2} \right).$$

For perfect phase matching, the photon flux grows quadratically:  $\phi_3 = g^2 \phi_1^3 z^2$ .

# Optical phase conjugation (OPC)

*Degenerate* four-wave mixing:  $\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega$ .

The two pump waves, 3 and 4, are counter-propagating:  $\mathbf{k}_3 = -\mathbf{k}_4$ .

Assuming that the pump intensities are much higher than those of the signal waves, we obtain

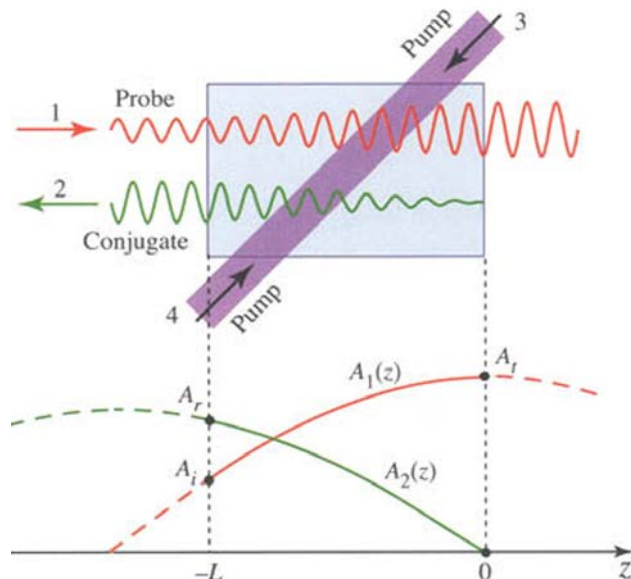
$$\begin{cases} (\nabla^2 + k^2)E_1 = -\xi E_2^* \\ (\nabla^2 + k^2)E_2 = -\xi E_1^* \end{cases}$$

where

$$\xi = 6\mu_0\omega^2\chi^{(3)}E_3E_4 = 6\mu_0\omega^2\chi^{(3)}A_3A_4$$

$$k = \bar{n}\omega/c_0, \quad \bar{n} \approx n + 2n_2I$$

**Phase conjugation:** As required by the phase-matching condition,  $\mathbf{k}_1 = -\mathbf{k}_2$ .



The slowly varying envelope approximation yields

$$\begin{cases} \frac{dA_1}{dz} = -j\gamma A_2^* \\ \frac{dA_2}{dz} = j\gamma A_1^* \end{cases} \Rightarrow \begin{cases} A_1(z) = \frac{A_i}{\cos \gamma L} \cos \gamma z \\ A_2(z) = j \frac{A_i^*}{\cos \gamma L} \sin \gamma z \end{cases}$$

$$A_r = -jA_i^* \tan \gamma L,$$

$$|\gamma| = 2C\sqrt{I_3I_4},$$

$$A_t = \frac{A_i}{\cos \gamma L}.$$

$$C = 3\omega\eta_0^2\frac{\chi^{(3)}}{\bar{n}^2}$$