

19. Nonlinear Optics

19.1 NONLINEAR OPTICAL MEDIA

19.2 SECOND-ORDER NONLINEAR OPTICS

- A. Second-Harmonic Generation and Rectification
- B. The Electro-Optic Effect
- C. Three-Wave Mixing

19.3 THIRD-ORDER NONLINEAR OPTICS

- A. Third-Harmonic Generation and Self-Phase Modulation
- B. Four-Wave Mixing
- C. Optical Phase Conjugation

*19.4 COUPLED-WAVE THEORY OF THREE-WAVE MIXING

- A. Second-Harmonic Generation
- B. Frequency Conversion
- C. Parametric Amplification and Oscillation

*19.5 COUPLED-WAVE THEORY OF FOUR-WAVE MIXING

*19.6 ANISOTROPIC NONLINEAR MEDIA

*19.7 DISPERSIVE NONLINEAR MEDIA

19.8 OPTICAL SOLITONS

Nonlinear optics

Polarization : $P = \epsilon_0 \chi E$

Susceptibility : $\chi = \chi_1 + \chi_2 E + \chi_3 E^2 + \dots$

$$D = \epsilon E = \epsilon_0 E + \epsilon_0 \chi E \rightarrow \epsilon = \epsilon_0 (1 + \chi) \rightarrow n = \frac{v}{c} = \sqrt{\frac{\epsilon}{\epsilon_0}} = \sqrt{1 + \chi}$$

$$P = P_1 + P_2 + P_3 + \dots = \epsilon_0 \chi_1 E + \epsilon_0 \chi_2 E^2 + \epsilon_0 \chi_3 E^3 + \dots$$

Linear first order: $P_1 = \epsilon_0 \chi_1 E$	Nonlinear second order: $P_2 = \epsilon_0 \chi_2 E^2$	Nonlinear third order: $P_3 = \epsilon_0 \chi_3 E^3$
Classical optics: Superposition Reflection Refraction Birefringence Absorption	Materials lacking inversion symmetry: Second harmonic generation Three-wave mixing Optical rectification Parametric amplification Pockels effect	Third harmonic generation Four-wave mixing Kerr effect Raman scattering Brillouin scattering Optical phase conjugation

Second-order Nonlinear optics $P_2 = \epsilon_0 \chi_2 E^2$

Second-harmonic generation (SHG) and rectification

$$E = E(\omega)|_{\text{optical}} \rightarrow P_2 \propto E^2(\omega) \rightarrow \begin{matrix} P_2(\omega \pm \omega) = P_2(2\omega), & \rightarrow \text{Frequency doubling} \\ P_2(0) & \rightarrow \text{Rectification} \end{matrix}$$

Electro-optic (EO) effect (Pockell's effect)

$$\begin{aligned} E &= E(0)|_{\text{electrical, DC}} + E(\omega)|_{\text{optical}} \quad \{\text{but, } |E(0)| \gg |E(\omega)|\} \\ &\rightarrow P_2 \propto E^2 \\ &\rightarrow P_2(0) \{\propto E^2(0)\}, P_2(\omega) \{\propto E(0)E(\omega)\}, P_2(2\omega) \{\propto E(\omega)E(\omega)\} \\ &\rightarrow P_2(0), P_2(\omega) \{\propto E(0)E(\omega)\} \rightarrow \Delta n \propto E(0)|_{\text{electric, DC}} \rightarrow \text{Index modulation by DC E-field} \end{aligned}$$

Three-wave mixing

$$\begin{aligned} E &= E(\omega_1)|_{\text{optical}} + E(\omega_2)|_{\text{optical}} \\ &\rightarrow P_2 \propto E^2 \\ &\rightarrow P_2(2\omega_1) \{\propto E^2(\omega_1)\}, P_2(2\omega_2) \{\propto E^2(\omega_2)\}, \rightarrow \text{SHG} \\ &\quad P_2(\omega_1 + \omega_2) \{\propto E(\omega_1)E(\omega_2)\}, \rightarrow \text{Frequency up-converter} \\ &\quad P_2(\omega_1 - \omega_2) \{\propto E(\omega_1)E(\omega_2)\} \rightarrow \text{Parametric amplifier, parametric oscillator} \end{aligned}$$

Third-order Nonlinear optics

$$P_3 = \epsilon_0 \chi_3 E^3$$

Third-harmonic generation (THG)

$$E = E(\omega)|_{\text{optical}} \rightarrow P_3 \propto E^3(\omega) \rightarrow P_3(\omega) \left\{ \propto |E(\omega)|^2 E(\omega) \right\}, P_3(3\omega) \left\{ \propto E^3(\omega) \right\}$$

→ Frequency tripling

Electro-optic (EO) Kerr effect

$$E = E(0)|_{\text{electrical, DC}} + E(\omega)|_{\text{optical}} \quad \left\{ \text{but, } |E(0)| \gg |E(\omega)| \right\}$$
$$\rightarrow P_3(\omega) \propto E(0)|_{\text{electric, DC}}^2 E(\omega) \rightarrow \Delta n \propto E(0)|_{\text{electric, DC}}^2 \rightarrow \text{Index modulation by DC } E^2$$

Optical Kerr effect

$$P_3(\omega) \propto |E(\omega)|^2 E(\omega) \propto I(\omega) E(\omega) \rightarrow \Delta n \propto I(\omega) \rightarrow \text{Index modulation by optical Intensity}$$

$$n = n_0 + \Delta n(I) \rightarrow \varphi = \varphi_0 + \Delta \varphi (= k_0 \Delta n L) \rightarrow \text{Self-phase modulation}$$

$$n = n_0 + \Delta n\{I(x)\} \rightarrow \Delta n\{I(x)\} > n_0 \rightarrow \text{Self-focusing, Self-guiding (Spatial solitons)}$$

$$n = n_0 + \Delta n\{I(x)\} \rightarrow \Delta n\{I(x)\} < n_0 \rightarrow \text{Self-defocusing}$$

Third-order Nonlinear optics

$$P_3 = \epsilon_0 \chi_3 E^3$$

Four-wave mixing

$$E = E(\omega_1)|_{\text{optical}} + E(\omega_2)|_{\text{optical}} + E(\omega_3)|_{\text{optical}}$$

$$\rightarrow P_3 \propto E^3 \rightarrow (\pm \omega_1, \pm \omega_2, \pm \omega_3)^3 \rightarrow 6^3 = 216 \text{ terms}$$

$$\rightarrow \text{One example: } P_3(\omega_1 + \omega_2 + \omega_3 \equiv \omega_4) \propto E(\omega_1)E(\omega_2)E(\omega_3) \rightarrow \text{Frequency up-converter}$$

$$\rightarrow \text{If } \omega_1 = \omega_2 = \omega_3 \rightarrow \omega_4 = 3\omega \rightarrow \text{THG}$$

$$\rightarrow \text{Another example: } P_3(\omega_1 + \omega_2 - \omega_3 \equiv \omega_4) \propto E(\omega_1)E(\omega_2)E^*(\omega_3)$$

$$\rightarrow \omega_1 + \omega_2 = \omega_3 + \omega_4$$

$$\rightarrow \text{If } \omega_1 = \omega_2 = \omega_3 = \omega_4 \rightarrow \text{Degenerate four-wave mixing}$$

\rightarrow Assume two waves among them are

plane waves traveling in opposite directions

$$\rightarrow P_3(\omega_4 = \omega) \propto |E(\omega)E(\omega)|E^*(\omega) \rightarrow \text{Optical phase conjugation}$$

24-2. Second harmonic generation (SHG)

$$P_2 = \varepsilon_0 \chi_2 E^2 \quad : \text{Only for non-centro-symmetry crystals}$$

[GaAs, CdTe, InAs, KDP, ADP, LiNbO₃, LiTaO₃, ...]

$$E = E_o \cos \omega t$$

$$P = P_1 + P_2$$

$$= \varepsilon_0 \chi_1 E_o \cos \omega t + \varepsilon_0 \chi_2 E_o^2 \cos^2 \omega t \quad \left\{ \cos^2 \theta = \frac{1}{2}(1 + \cos 2\theta) \right\}$$

$$= \varepsilon_0 \chi_1 E_o \cos \omega t + \frac{1}{2} \varepsilon_0 \chi_2 E_o^2 + \frac{1}{2} \varepsilon_0 \chi_2 E_o^2 \cos 2\omega t$$

$$P_2(t) = \left\{ \frac{1}{2} \varepsilon_0 \chi_2 E_o^2 \right\} + \left\{ \frac{1}{2} \varepsilon_0 \chi_2 E_o^2 \right\} \cos 2\omega t = P_2(0) + P_2(2\omega)$$

Constant (DC) term
→ Optical rectification

Second harmonic term
→ 2ω

SHG does not occur in isotropic, centrosymmetry crystals

It can easily be shown that the second-order term makes no contribution to polarization in an isotropic optical material, or one having a center of symmetry. A crystal having a center of symmetry is characterized by an inversion center, such that if the radial coordinate r is changed to $-r$, the crystal's atomic arrangement remains unchanged and so the crystal responds in the same way to a physical influence. In such a crystal, reversing the applied field should not—except for a change in sign—change any physical property, such as its polarization. Thus we should have both

$$P_2 = \epsilon_0 \chi_2 (+E)^2 \quad \text{and} \quad -P_2 = \epsilon_0 \chi_2 (-E)^2$$

Because the E -field is squared, $P_2 = -P_2$, which can only be true if $P_2 = 0$. The quartz crystal used by Franken, and many other crystals as well, do not possess inversion symmetry. They can, therefore, manifest second harmonic generation in addition to other second-order phenomena to be described presently.

$$P_2 = \epsilon_0 \chi_2 E^2$$

If χ_2 is isotropic or centrosymmetric,

→ both $+E$ and $-E$ give the same P_2 polarization

→ that means the molecules are not polarized by the second χ effect.

Second harmonic generation

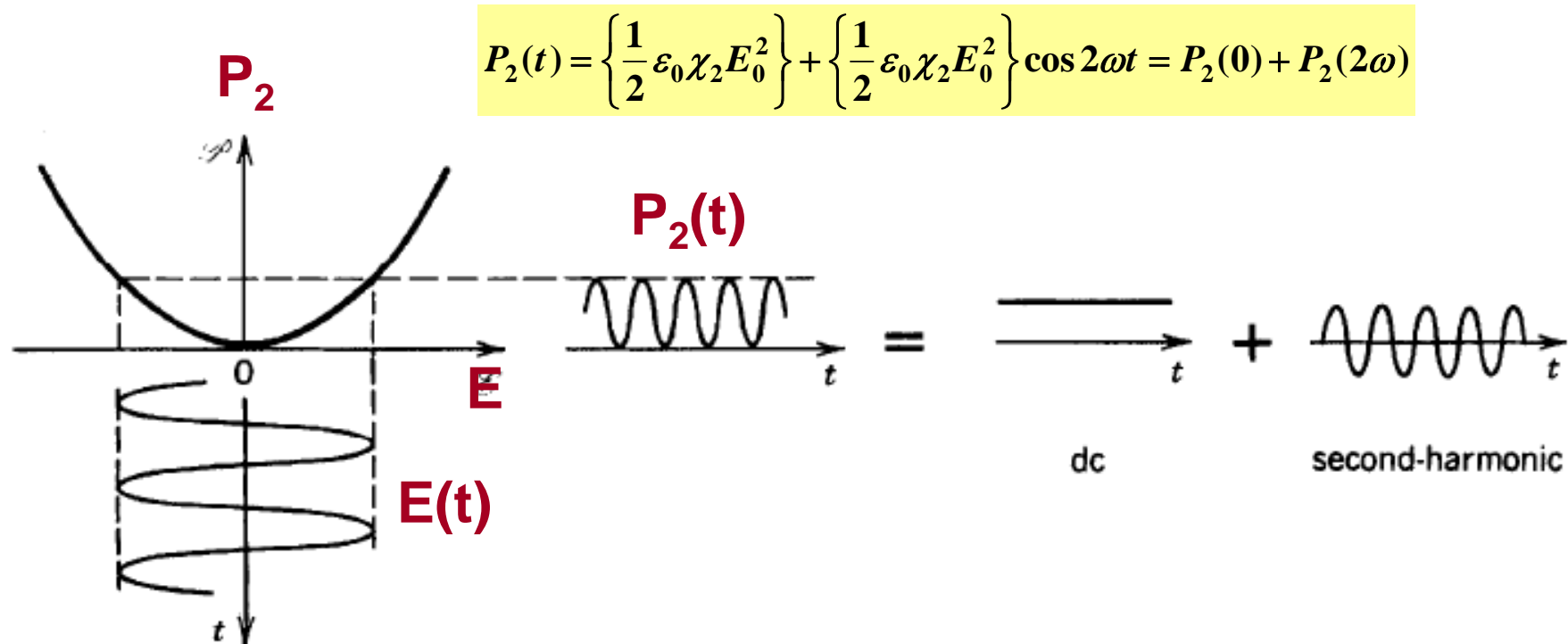
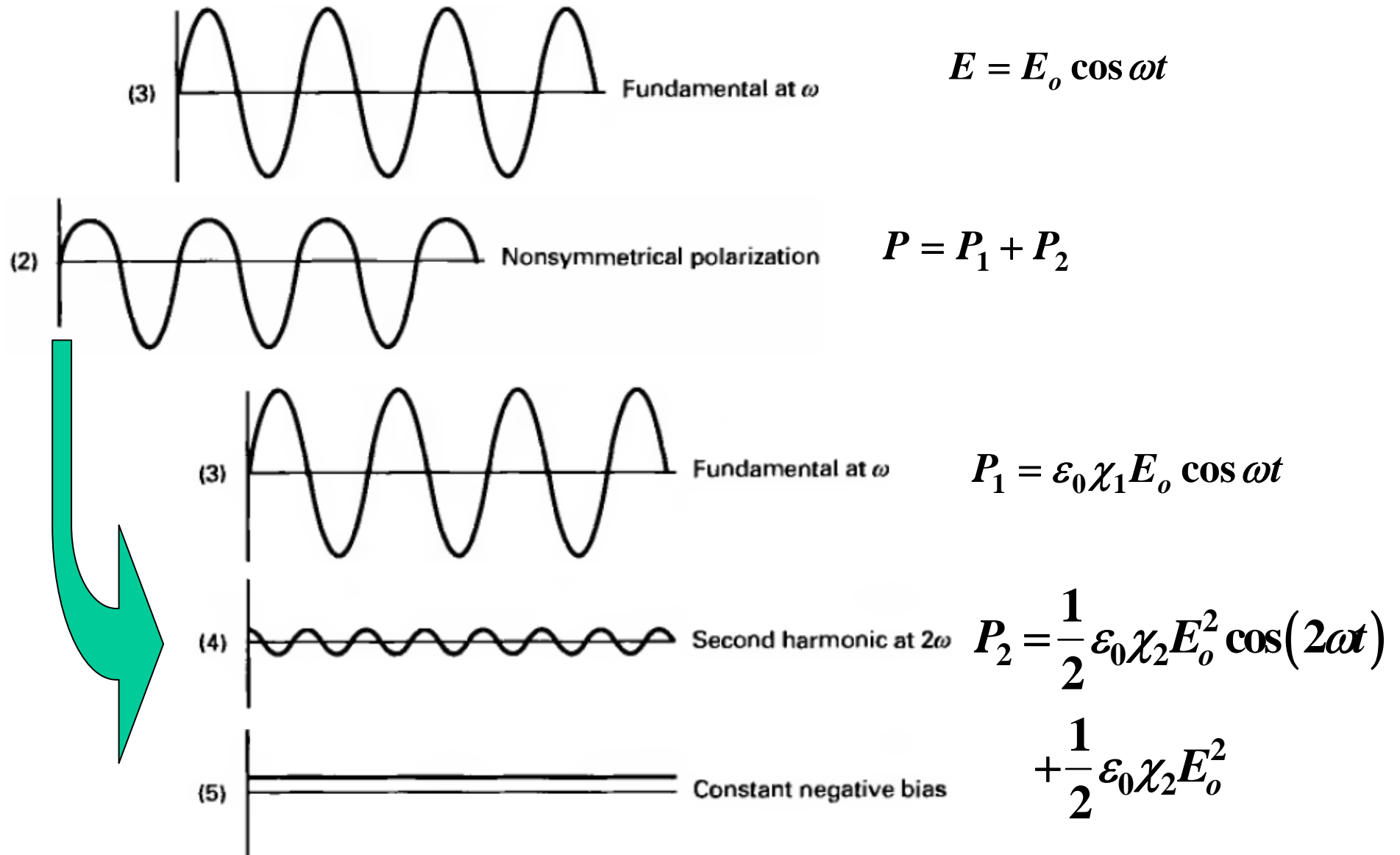


Figure 19.2-1 A sinusoidal electric field of angular frequency ω in a second-order nonlinear optical medium creates a polarization with a component at 2ω (second-harmonic) and a steady (dc) component.

Second harmonic generation



Second harmonic generation

$$\omega \rightarrow 2\omega \quad (\lambda \rightarrow \lambda / 2)$$

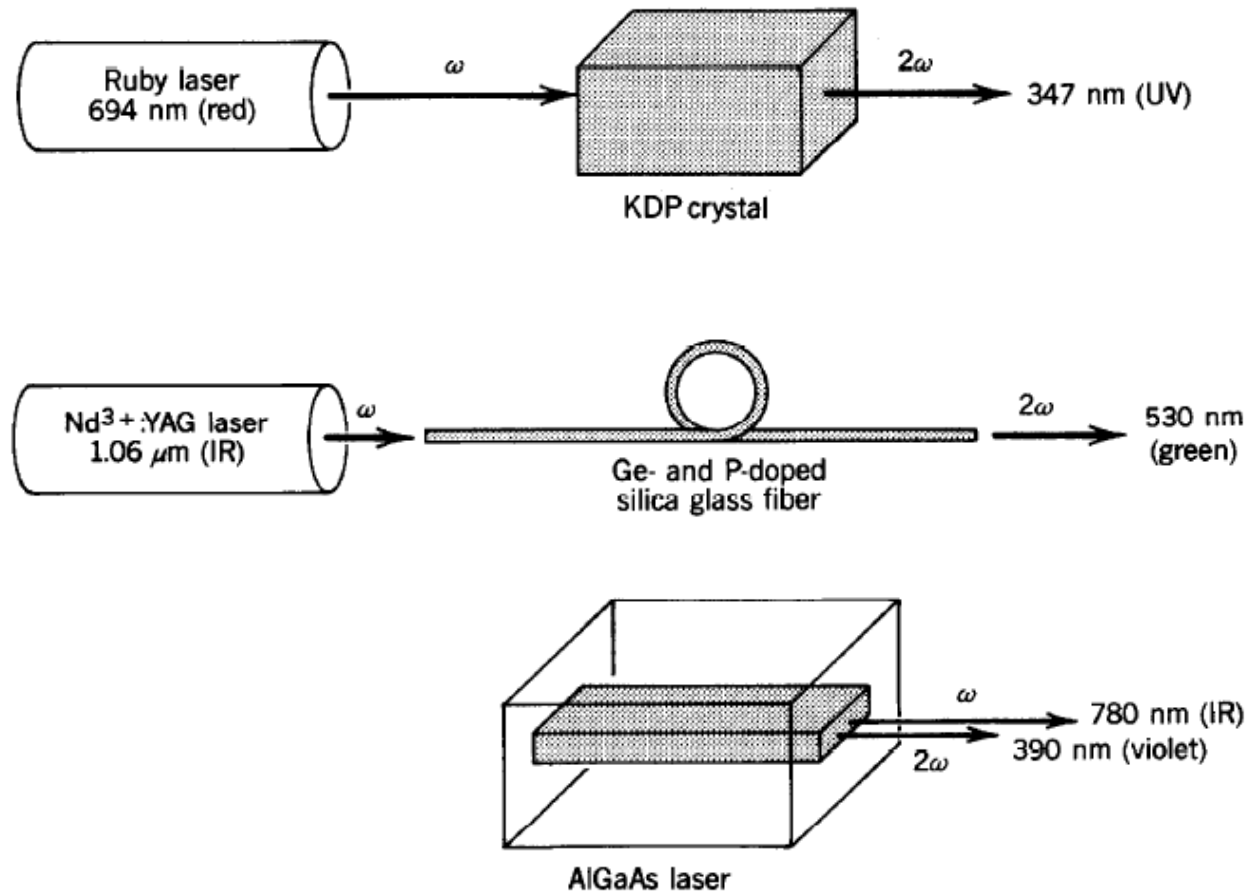
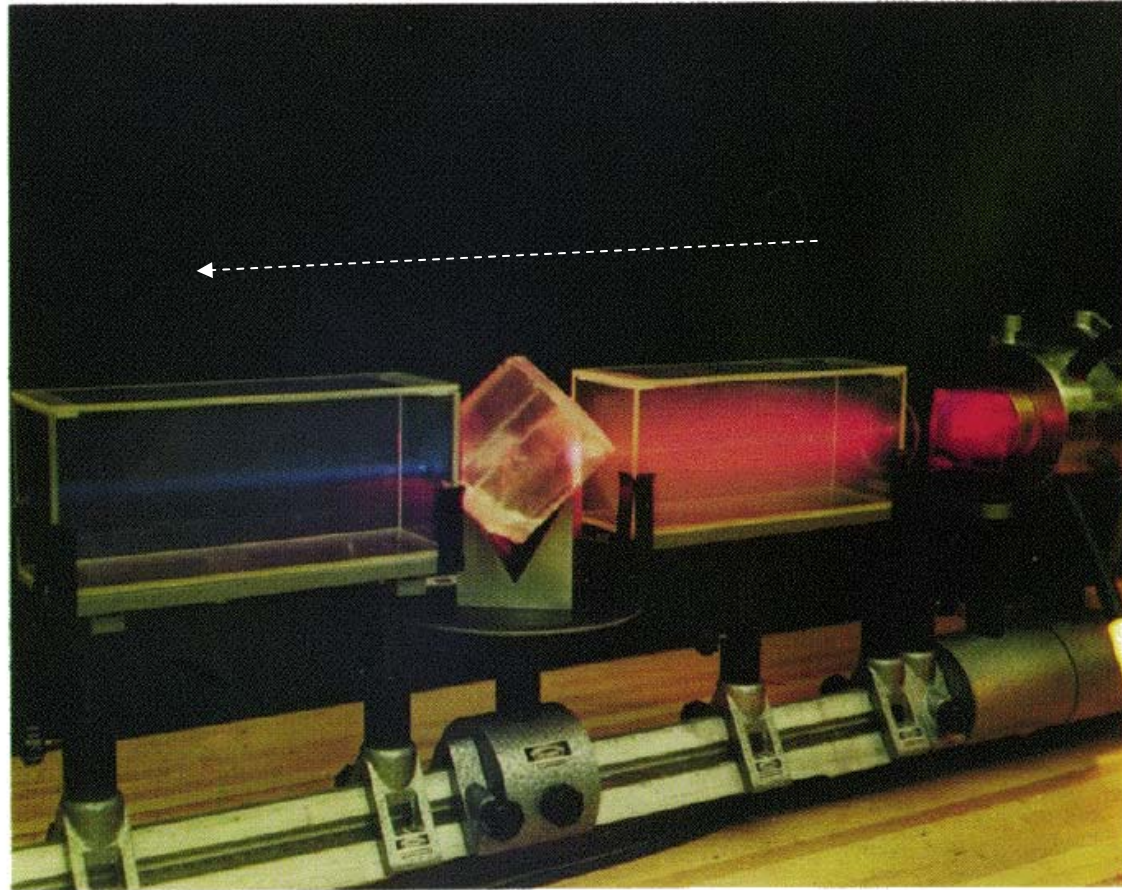


Figure 19.2-2 Optical second-harmonic generation in (a) a bulk crystal; (b) a glass fiber; (c) within the cavity of a semiconductor laser.

Second harmonic generation



Laser beam enters a crystal of ammonium dihydrogen phosphate as red light and emerges as blue—the second harmonic. Courtesy of R. W. Terhune.

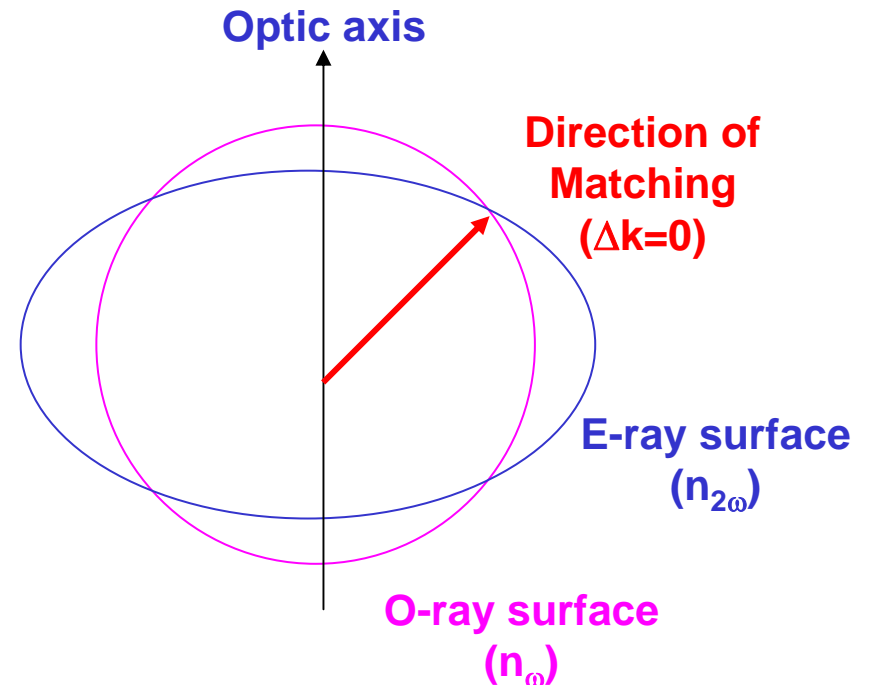
Phase matching (index matching) in SHG

Output intensity after second harmonic generation

$$I \propto \sin^2 \left(\frac{L\Delta k}{2} \right), \quad \Delta k = k_{2\omega} - 2k_{\omega}$$

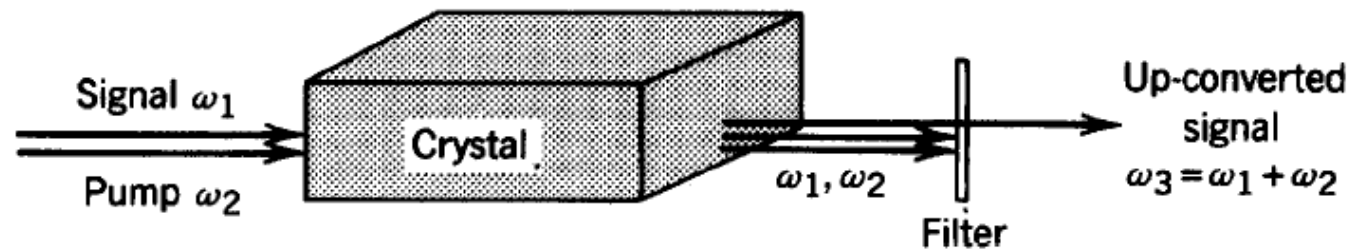
Phase matching : $\Delta k=0$

$$\begin{aligned} \Delta k &= k_{2\omega} - 2k_{\omega} \\ &= n_{2\omega} \left(\frac{2\omega}{c} \right) - 2n_{\omega} \left(\frac{\omega}{c} \right) \\ &= (n_{2\omega} - n_{\omega}) \left(2 \frac{\omega}{c} \right) = 0 \end{aligned}$$

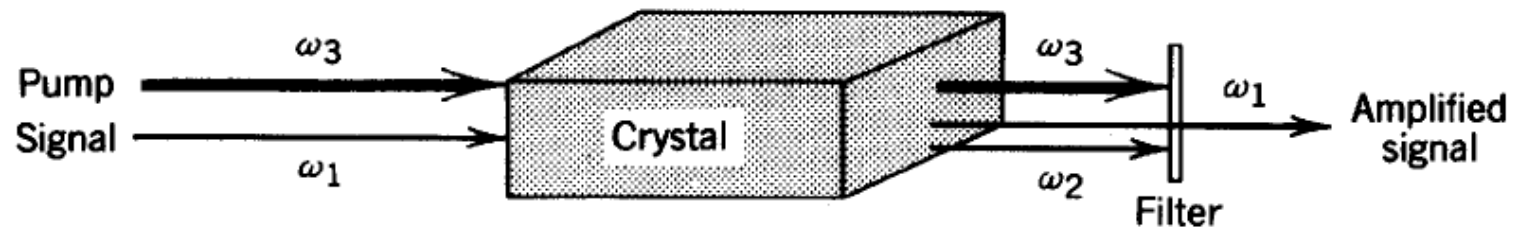


Frequency mixing by three-wave mixing

frequency up-converter ($\omega_1 + \omega_2 \Rightarrow \omega_3$)

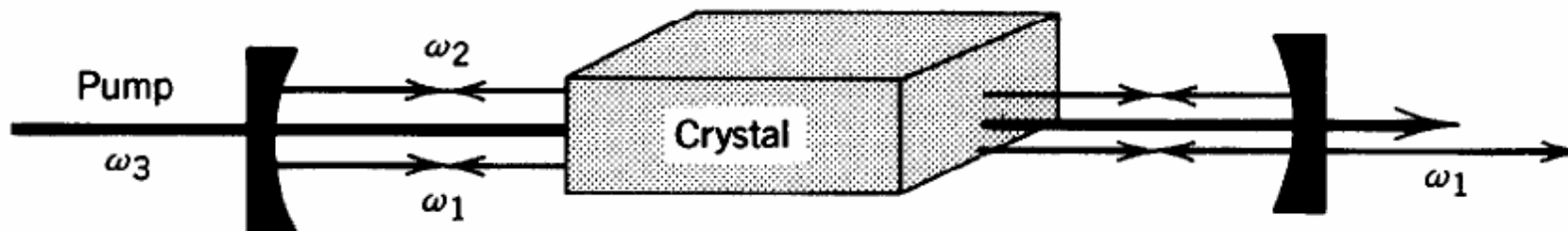


parametric amplifier ($\omega_3 - \omega_1 \Rightarrow \omega_2 \rightarrow \omega_3 - \omega_2 \Rightarrow \omega_1$)



($\omega_2 \rightarrow$ idler, or parameter, 중개자)

parametric oscillator ($\omega_3 \Rightarrow \omega_1 + \omega_2 \rightarrow \omega_3 - \omega_2 \Rightarrow \omega_1$)



Parametric interaction

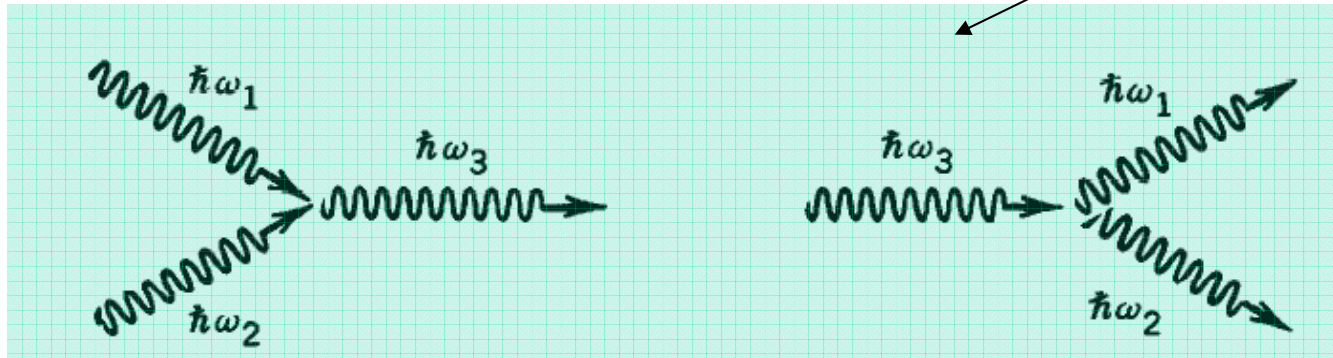
$$E = E(\omega_1) + E(\omega_2)$$

$$= E_{o1} \cos \omega_1 t + E_{o2} \cos \omega_2 t$$

$$= \frac{1}{2} E_{o1} \{ \exp(i\omega_1 t) + \exp(-i\omega_1 t) \} + \frac{1}{2} E_{o2} \{ \exp(i\omega_2 t) + \exp(-i\omega_2 t) \}$$

$$P_2 = \varepsilon_0 \chi_2 E^2$$

$$\Rightarrow (\omega_1 + \omega_1 = 2\omega_1), (\omega_2 + \omega_2 = 2\omega_2), (\omega_1 - \omega_2 = \omega_3), (\omega_1 + \omega_2 = \omega_3)$$



$$\hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2 \quad \rightarrow \text{Frequency conservation}$$

$$\hbar\mathbf{k}_3 = \hbar\mathbf{k}_1 + \hbar\mathbf{k}_2 \quad \rightarrow \text{Momentum (phase) matching}$$

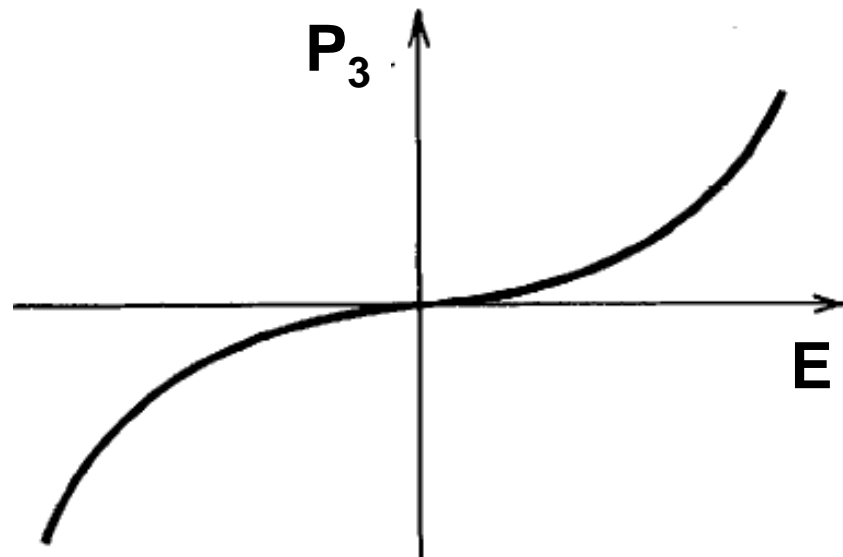
Third-order nonlinear effect

In media possessing *centrosymmetry*, the second-order nonlinear term is absent since the polarization must reverse exactly when the electric field is reversed.

The dominant nonlinearity is then of third order,

$$P_3 = \epsilon_0 \chi_3 E^3$$

The third-order nonlinear material is called a *Kerr medium*.



Optical Kerr effect

$$P_3(\omega) \propto |E(\omega)|^2 E(\omega) \propto I(\omega) E(\omega) \rightarrow \Delta n \propto I(\omega) \rightarrow \text{Index modulation by optical Intensity}$$

$$n = n_0 + \Delta n(I) \rightarrow \varphi = \varphi_0 + \Delta\varphi (= k_0 \Delta n L) \rightarrow \text{Self-phase modulation}$$

$$n = n_0 + \Delta n\{I(x)\} \rightarrow \Delta n\{I(x)\} > n_0 \rightarrow \text{Self-focusing, Self-guiding (Spatial solitons)}$$

$$n = n_0 + \Delta n\{I(x)\} \rightarrow \Delta n\{I(x)\} < n_0 \rightarrow \text{Self-defocusing}$$

$$P_{\text{NL}}(\omega) = 3\chi^{(3)} |E(\omega)|^2 E(\omega)$$

$$\epsilon_o \Delta\chi = \frac{P_{\text{NL}}(\omega)}{E(\omega)} = 3\chi^{(3)} |E(\omega)|^2 = 6\chi^{(3)} \eta I, \quad \text{where } I = |E(\omega)|^2 / 2\eta$$

Since $n^2 = 1 + \chi$,

$$\Delta n = (\partial n / \partial \chi) \Delta\chi = \Delta\chi / 2n, \longrightarrow \Delta n = \frac{3\eta}{\epsilon_o n} \chi^{(3)} I = n_2 I$$

$$\longrightarrow \boxed{n(I) = n + n_2 I} \quad \text{where } n_2 = \frac{3\eta_o}{n^2 \epsilon_o} \chi^{(3)}$$

The order of magnitude of the coefficient n_2 (in units of cm^2/W) is 10^{-16} to 10^{-14} in glasses, 10^{-14} to 10^{-7} in doped glasses, 10^{-10} to 10^{-8} in organic materials, and 10^{-10} to 10^{-2} in semiconductors. It is sensitive to the operating wavelength (see Sec. 19.7) and depends on the polarization.

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

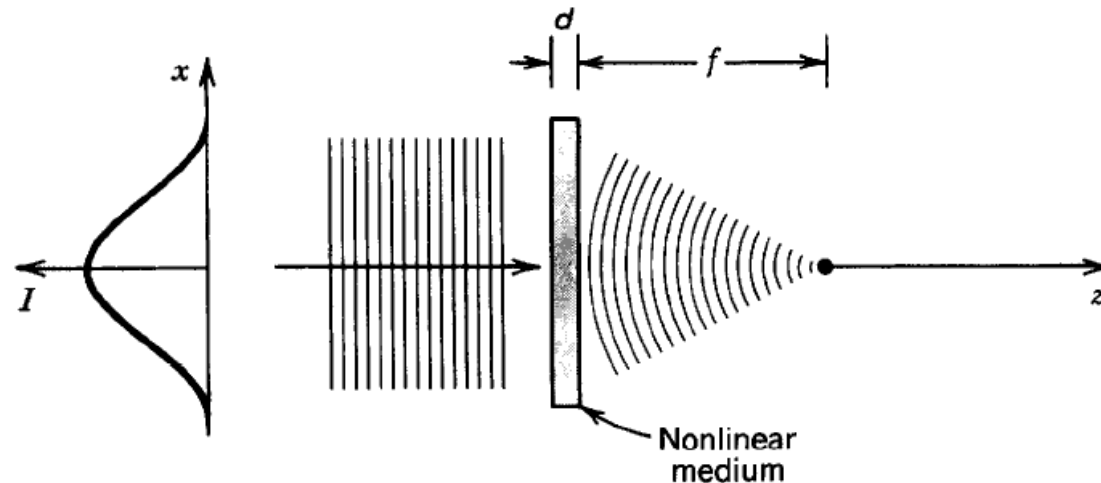
Self-phase modulation

The phase shift incurred by an optical beam of power P and cross-sectional area A , traveling a distance L in the medium,

$$\varphi = 2\pi n(I)L/\lambda_o = 2\pi(n + n_2 P/A)L/\lambda_o \longrightarrow \Delta\varphi = 2\pi n_2 \frac{L}{\lambda_o A} P$$

$$P_\pi = \lambda_o A / 2Ln_2 \text{ at which } \Delta\varphi = \pi$$

Self-focusing (Optical Kerr lens)

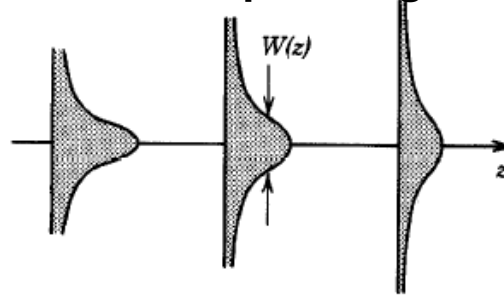


Spatial Solitons

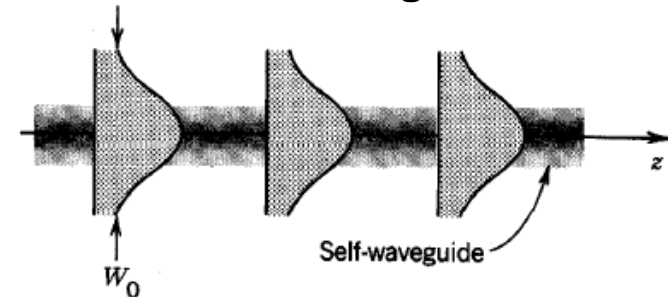
= Self-guided beam

(Also, see 19.8)

In a linear medium,
wave is spreading.



In an optical Kerr medium,
wave can be self-guided.



Helmholtz equation, $[\nabla^2 + n^2(I)k_o^2]E = 0$, where $n(I) = n + n_2 I$, $k_o = \omega/c_o$, $I = |E|^2/2\eta$

➡ nonlinear differential equation in E

$E = A \exp(-jkz)$ assuming that $A = A(x, z)$ varies slowly in the z direction

$$(\partial^2/\partial z^2)[A \exp(-jkz)] \approx (-2jk \partial A/\partial z - k^2 A) \exp(-jkz),$$

➡
$$\frac{\partial^2 A}{\partial x^2} - 2jk \frac{\partial A}{\partial z} + k_o^2 [n^2(I) - n^2] A = 0 \quad (n_2 I \ll n),$$

$$[n^2(I) - n^2] = [n(I) - n][n(I) + n] \approx [n_2 I][2n] = \frac{2n_2 n |A|^2}{2\eta} = \frac{n^2 n_2}{\eta_o} |A|^2$$


➡
$$\frac{\partial^2 A}{\partial x^2} + \frac{n_2}{\eta_o} k^2 |A|^2 A = 2jk \frac{\partial A}{\partial z} \quad \text{: Nonlinear Schrodinger equation}$$

$$A(x, z) = A_0 \operatorname{sech}\left(\frac{x}{W_0}\right) \exp\left(-j \frac{z}{4z_0}\right) \quad n_2(A_0^2/2\eta_o) = 1/k^2 W_0^2 \quad z_0 = \frac{1}{2} k W_0^2 = \pi W_0^2 / \lambda$$

Raman Gain

The nonlinear coefficient $\chi^{(3)}$ is in general complex-valued, $\chi^{(3)} = \chi_R^{(3)} + j\chi_I^{(3)}$

$$\Delta\varphi = 2\pi n_2 \frac{L}{\lambda_o A} P \quad \xrightarrow{n_2 = \frac{3\eta_o}{n^2 \epsilon_o} \chi^{(3)}} \quad = \frac{6\pi\eta_o}{\epsilon_o} \frac{\chi^{(3)}}{n^2} \frac{L}{\lambda_o A} P$$

phase factor $\exp(-j\varphi)$		phase shift	$\Delta\varphi = (6\pi\eta_o/\epsilon_o)(\chi_R^{(3)}/n^2)(L/\lambda_o A)P$
		Raman gain	$\exp(\frac{1}{2}\gamma L)$
			$\gamma = \frac{12\pi\eta_o}{\epsilon_o} \frac{\chi_I^{(3)}}{n^2} \frac{1}{\lambda_o A} P$: Raman Gain Coefficient

Coupling of light to the high-frequency vibrational modes of the medium, which act as an energy source providing the gain.

For low-loss media, the Raman gain may exceed the loss at reasonable levels of power, so that the medium can act as an optical amplifier.

→ **Fiber Raman lasers**

Four-wave mixing (third-order nonlinearity)

Superposition of three waves of angular frequencies ω_1 , ω_2 , and ω_3

$$\mathcal{E}(t) = \text{Re}\{E(\omega_1) \exp(j\omega_1 t)\} + \text{Re}\{E(\omega_2) \exp(j\omega_2 t)\} + \text{Re}\{E(\omega_3) \exp(j\omega_3 t)\}$$

$$\mathcal{E}(t) = \sum_{q=\pm 1, \pm 2, \pm 3} \frac{1}{2} E(\omega_q) \exp(j\omega_q t) \quad \text{where } \omega_{-q} = -\omega_q$$

and $E(-\omega_q) = E^*(\omega_q)$.

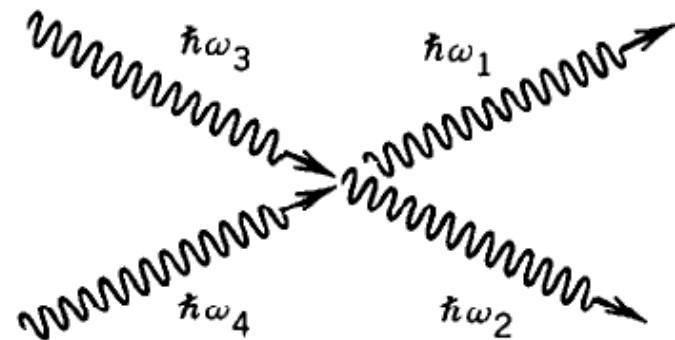
$$P_3 = \varepsilon_0 \chi_3 E^3 \quad (\text{as sum of } 6^3 = 216 \text{ terms})$$

$$= \frac{1}{2} \chi^{(3)} \sum_{q,r,l=\pm 1, \pm 2, \pm 3} E(\omega_q) E(\omega_r) E(\omega_l) \exp[j(\omega_q + \omega_r + \omega_l)t]$$

If $\omega_4 = \omega_1 + \omega_2 - \omega_3$

$$\rightarrow \{ \omega_3 + \omega_4 = \omega_1 + \omega_2 \}$$

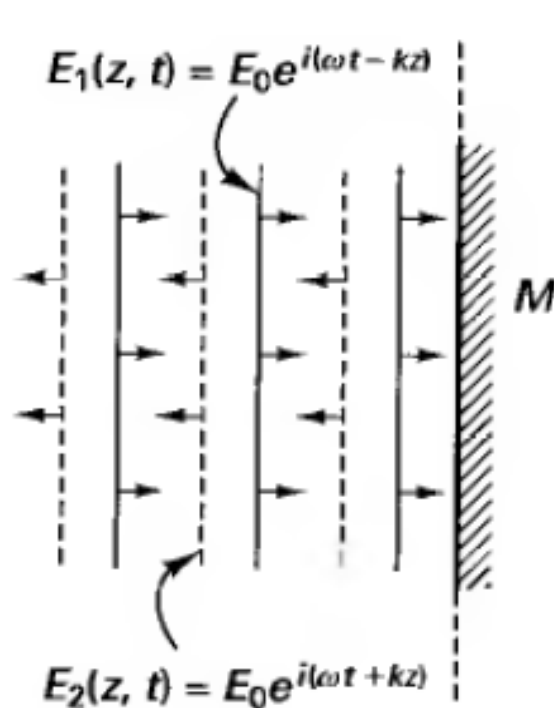
$$\{ \vec{k}_3 + \vec{k}_4 = \vec{k}_1 + \vec{k}_2 \}$$



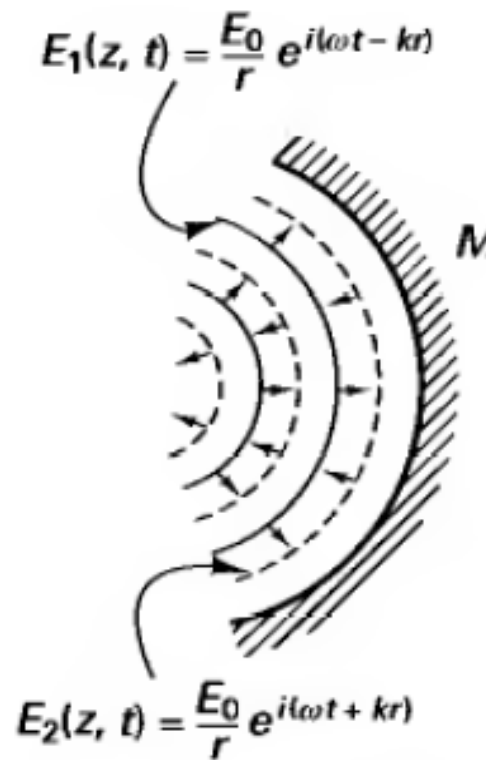
Optical phase conjugation

Optical phase conjugation (OPC)

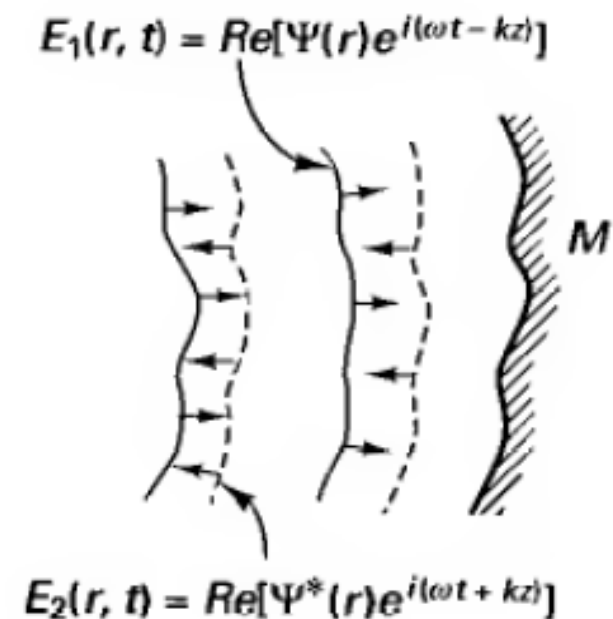
is the *spatial complex conjugate* of the incident wave. This means that a new wave is produced that exactly reverses the direction and overall phase factor of the primary beam. Thus the phase conjugate wave precisely retraces the path of the original beam and, at each position, reproduces the exact shape of the original wavefront.



(a) Plane wave



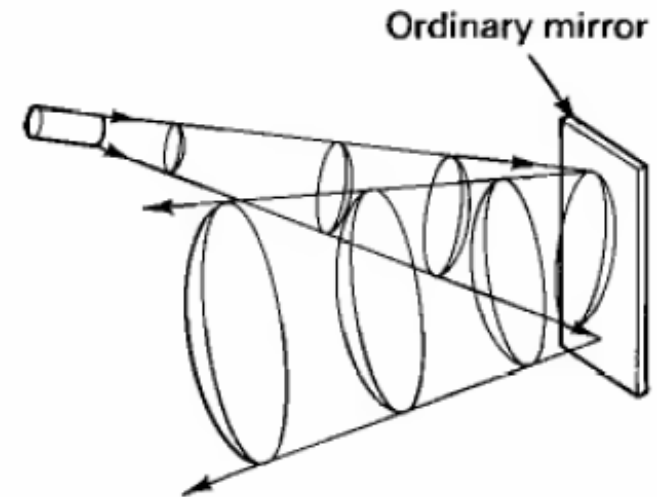
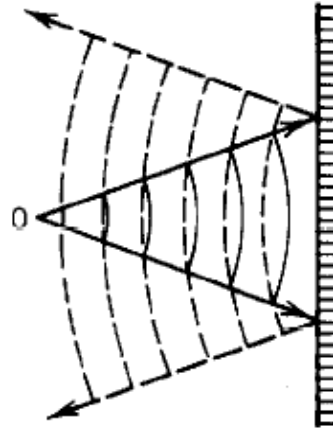
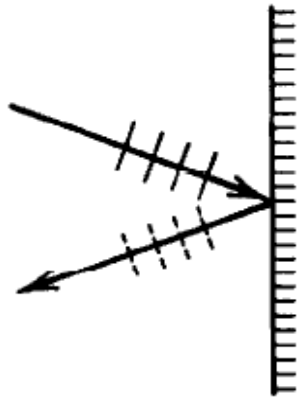
(b) Spherical wave



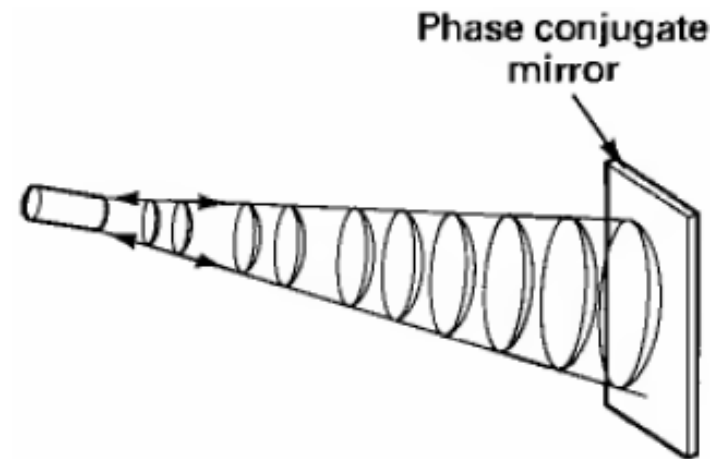
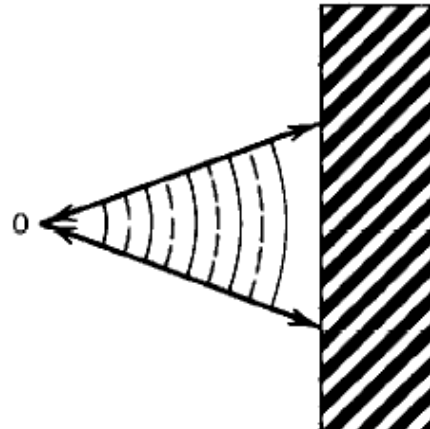
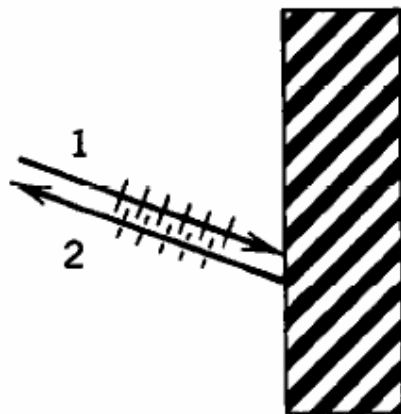
(c) "Nearly plane" wave

Phase conjugate mirror (PCM)

Conventional mirror



PCM



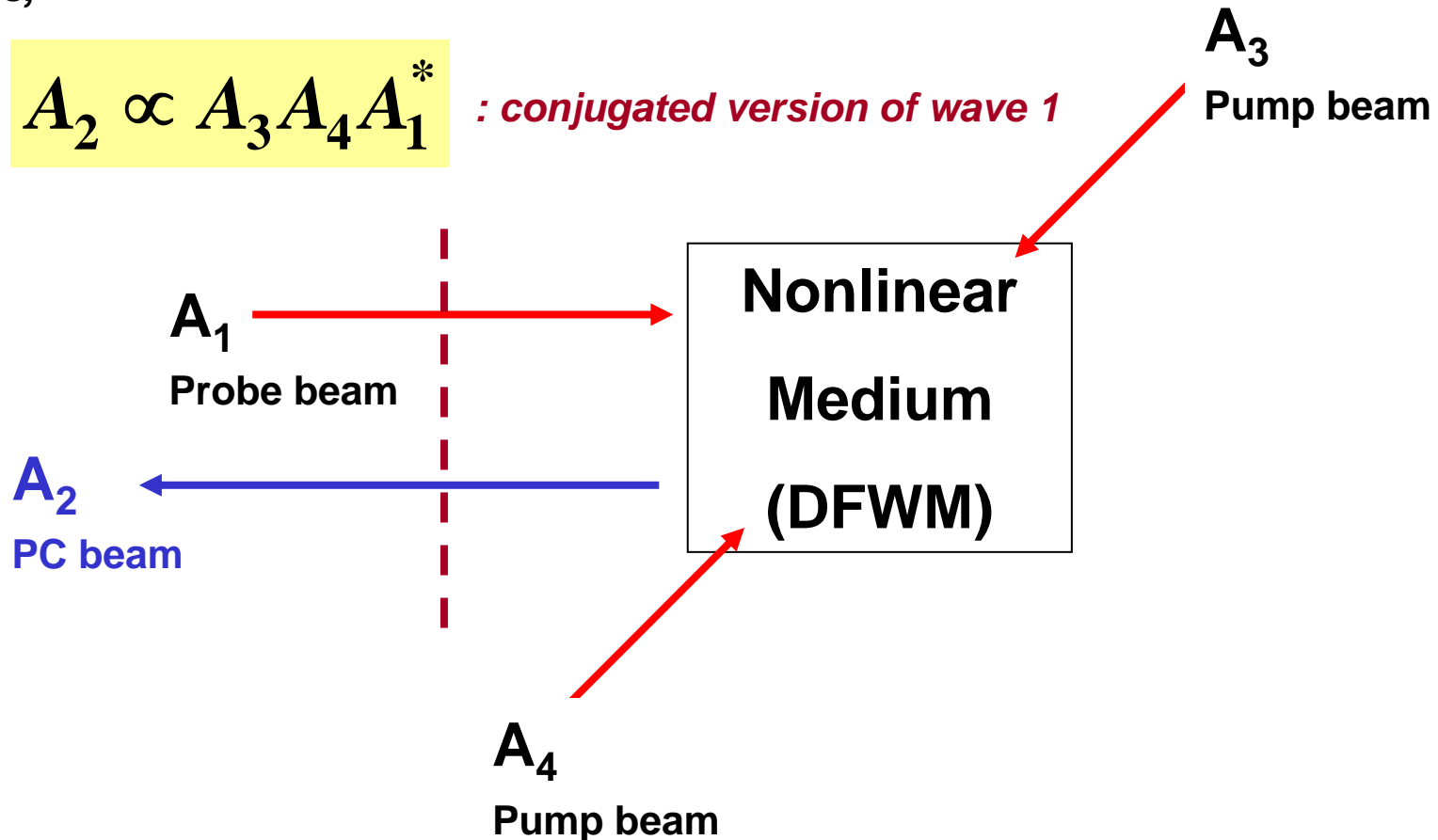
Optical phase conjugation

When all four waves are of the same frequency → *degenerated four-wave mixing (DFWM)*

$$\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega$$

Assuming further that two waves (3,4) are uniform plane waves traveling in opposite directions,

$$A_2 \propto A_3 A_4 A_1^* \quad : \text{conjugated version of wave 1}$$



Note: Phase Conjugation and Time Reversal

Incident

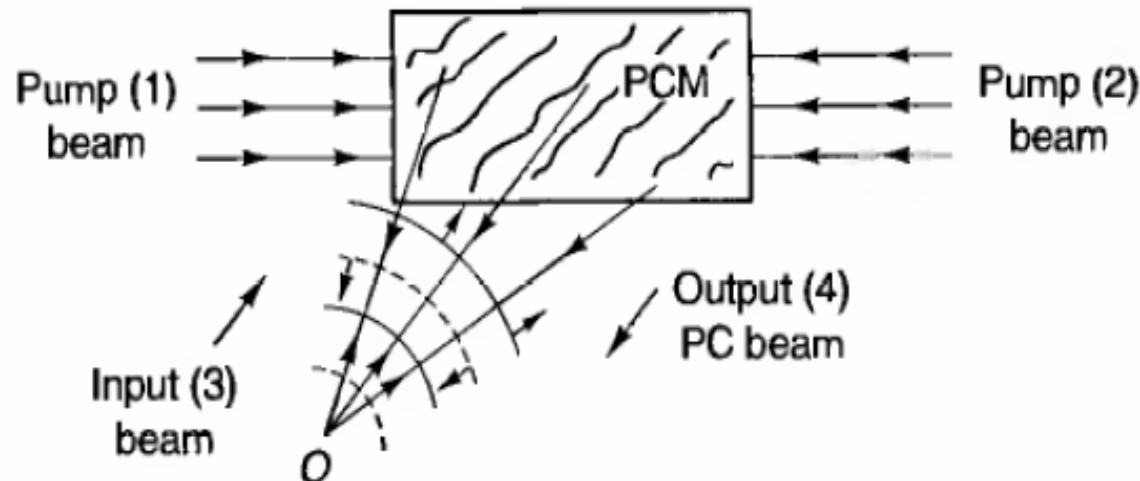
$$E_1(\mathbf{r}, t) = \text{Re}[\psi(\mathbf{r})e^{i(\omega t - kz)}]$$

Phase conjugation

$$E_2(\mathbf{r}, t) = \text{Re}[\psi^*(\mathbf{r})e^{i(\omega t + kz)}]$$

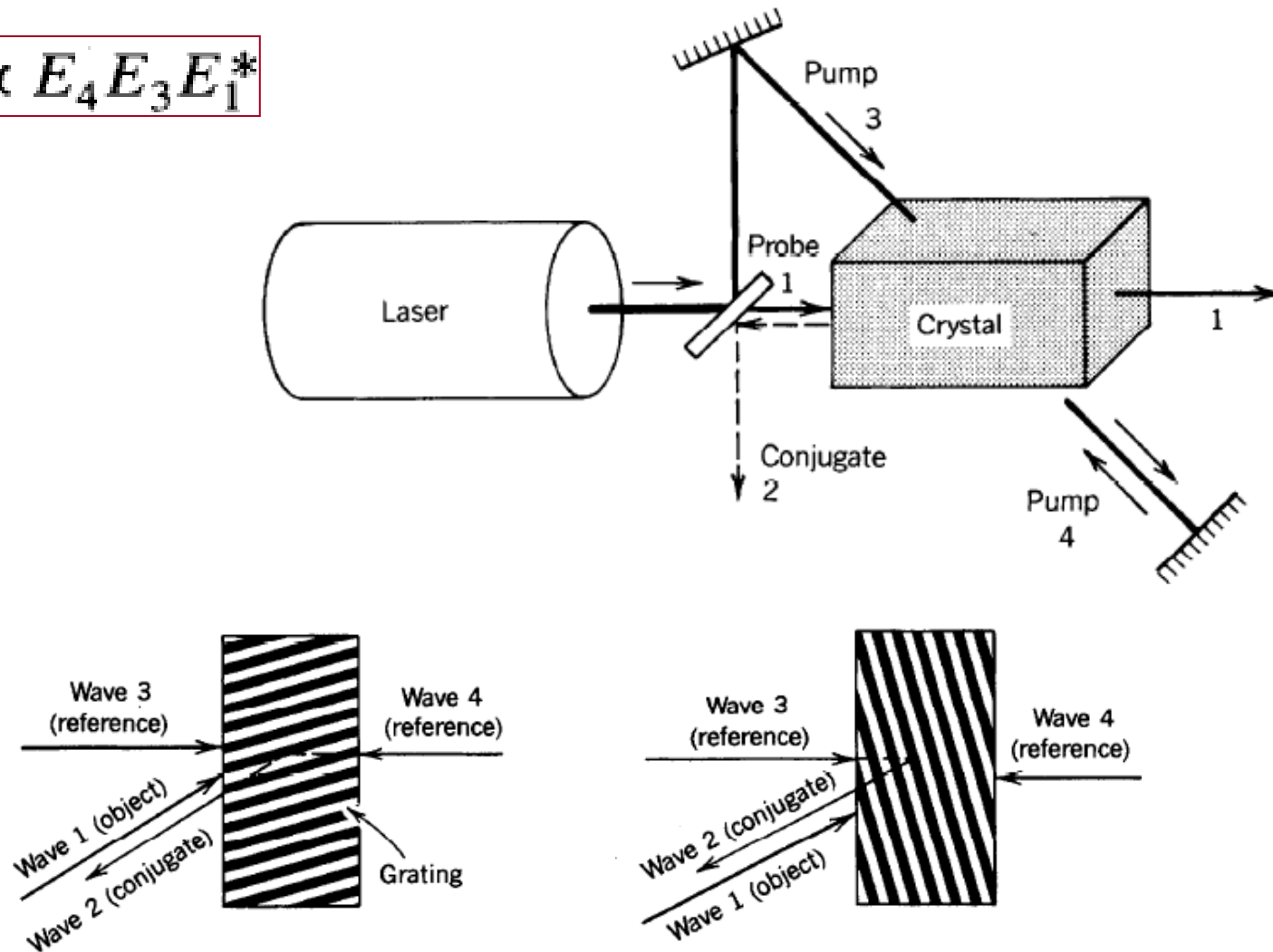
Time reversal

$$E_2(\mathbf{r}, t) = \text{Re}[\psi(\mathbf{r})e^{i[\omega(-t) - kz]}]$$



Degenerate Four-Wave Mixing as a Form of Real-Time Holography

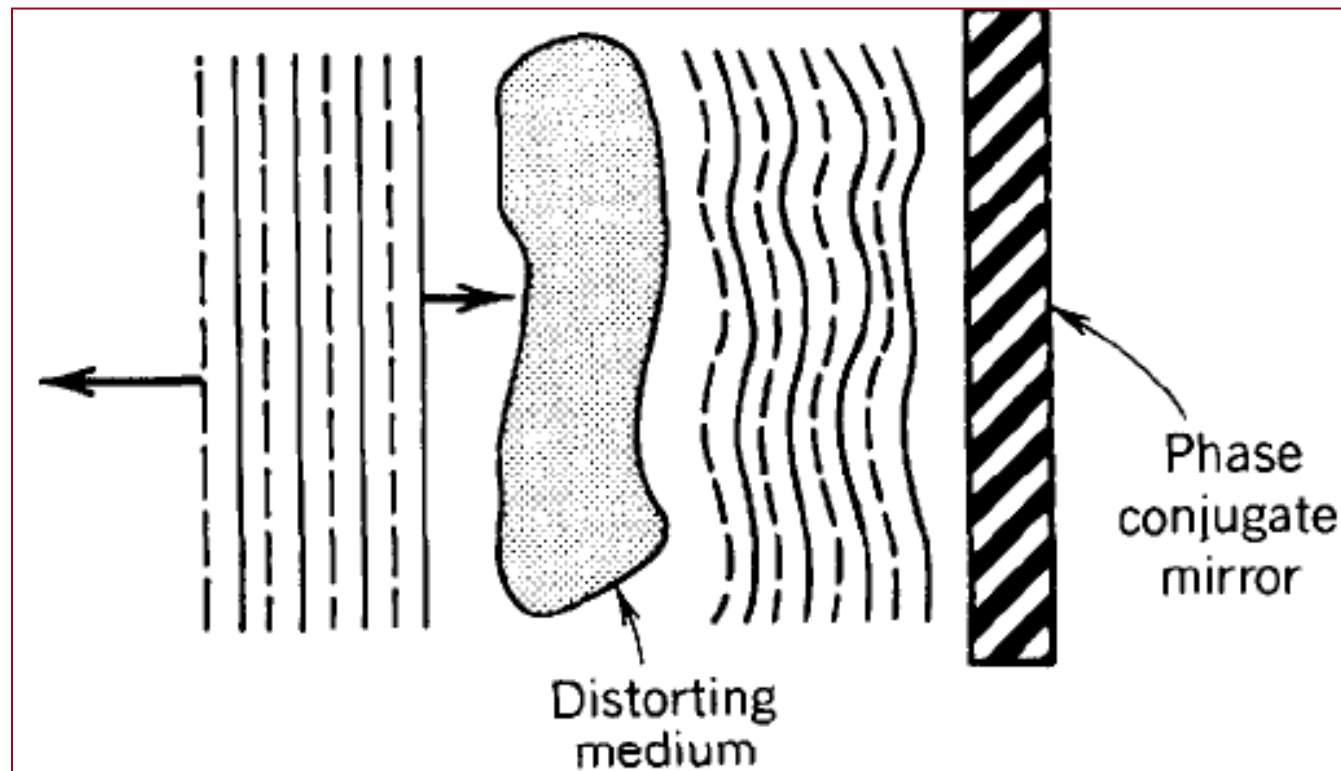
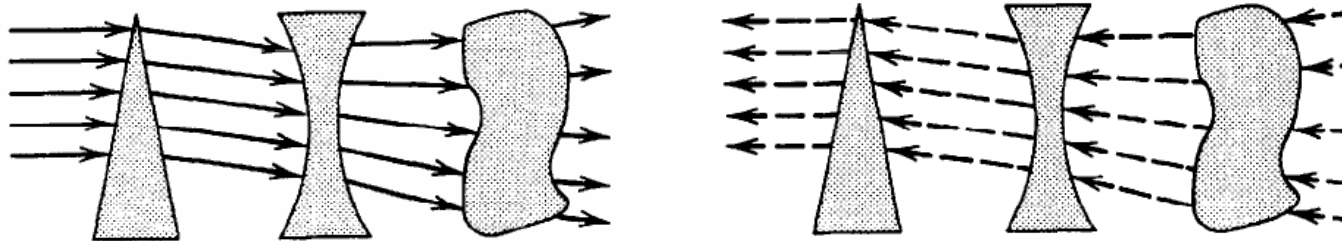
$$E_2 \propto E_4 E_3 E_1^*$$



two possibilities corresponding to (a) transmission and (b) reflection gratings.

Image restoration by phase conjugation

Optical reciprocity.



19.4. Coupled-wave theory of three-wave mixing

Coupled-Wave Equations

Wave propagation in a second-order nonlinear medium

$$\nabla^2 \mathcal{E} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = -\mathcal{J}, \quad \text{where } \mathcal{J} = -\mu_o \frac{\partial^2 \mathcal{P}_{\text{NL}}}{\partial t^2} \quad \text{and} \quad \mathcal{P}_{\text{NL}} = 2 \mathcal{d} \mathcal{E}^2$$

The field $\mathcal{E}(t)$ is a superposition of three waves of angular frequencies ω_1 , ω_2 , and ω_3

$$\begin{aligned} \mathcal{E}(t) &= \sum_{q=1,2,3} \text{Re} [E_q \exp(j\omega_q t)] = \sum_{q=1,2,3} \frac{1}{2} [E_q \exp(j\omega_q t) + E_q^* \exp(-j\omega_q t)] \\ &= \sum_{q=\pm 1, \pm 2, \pm 3} \frac{1}{2} E_q \exp(j\omega_q t), \quad \text{where } \omega_{-q} = -\omega_q \text{ and } E_{-q} = E_q^*. \end{aligned}$$

The corresponding polarization density

$$\mathcal{P}_{\text{NL}} = 2 \mathcal{d} \mathcal{E}^2 = \frac{1}{2} \mathcal{d} \sum_{q,r=\pm 1, \pm 2, \pm 3} E_q E_r \exp[j(\omega_q + \omega_r)t]$$

the corresponding radiation source

$$\mathcal{J} = -\mu_o \frac{\partial^2 \mathcal{P}_{\text{NL}}}{\partial t^2} = \frac{1}{2} \mu_o \mathcal{d} \sum_{q,r=\pm 1, \pm 2, \pm 3} (\omega_q + \omega_r)^2 E_q E_r \exp[j(\omega_q + \omega_r)t]$$

Coupled-Wave Equations

$$\nabla^2 \mathcal{E} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = -\mathcal{S}$$

by equating terms on both sides at each of the frequencies ω_1 , ω_2 , and ω_3 , separately,

$$(\nabla^2 + k_1^2)E_1 = -S_1$$

$$(\nabla^2 + k_2^2)E_2 = -S_2$$

$$(\nabla^2 + k_3^2)E_3 = -S_3$$

where S_q is the amplitude of the component of \mathcal{S} with frequency ω_q

$$S_1 = 2\mu_o\omega_1^2 \mathcal{E} E_3 E_2^* \quad (\text{since } \omega_1 = \omega_3 - \omega_2)$$

Assume, for example, $\omega_3 = \omega_1 + \omega_2$  $S_2 = 2\mu_o\omega_2^2 \mathcal{E} E_3 E_1^* \quad (\text{since } \omega_2 = \omega_3 - \omega_1)$

$$S_3 = 2\mu_o\omega_3^2 \mathcal{E} E_1 E_2. \quad (\text{since } \omega_3 = \omega_1 + \omega_2)$$



$$(\nabla^2 + k_1^2)E_1 = -2\mu_o\omega_1^2 \mathcal{E} E_3 E_2^*$$

$$(\nabla^2 + k_2^2)E_2 = -2\mu_o\omega_2^2 \mathcal{E} E_3 E_1^*$$

$$(\nabla^2 + k_3^2)E_3 = -2\mu_o\omega_3^2 \mathcal{E} E_1 E_2.$$

**Coupled-wave Equations
in three-wave mixing**

Homework : EXERCISE 19.4-1 Degenerate Three-Wave Mixing

Mixing of Three Collinear Uniform Plane Waves

$$E_q = A_q \exp(-jk_q z), \quad q = 1, 2, 3$$

$$= (2\eta\hbar\omega_q)^{1/2} a_q \exp(-jk_q z), \quad \leftarrow a_q = A_q / (2\eta\hbar\omega_q)^{1/2}$$

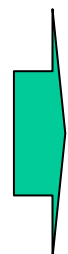
$$I_q = |E_q|^2 / 2\eta = \hbar\omega_q |a_q|^2 \quad \longrightarrow \quad |a_q|^2 = \frac{I_q}{\hbar\omega_q} \quad \begin{array}{l} \text{The photon flux} \\ \text{densities (photons/s-m}^2\text{)} \end{array}$$

slowly varying envelope approximation $(\nabla^2 + k_q^2)[a_q \exp(-jk_q z)] \approx -j2k_q \frac{da_q}{dz} \exp(-jk_q z)$

$$(\nabla^2 + k_1^2)E_1 = -2\mu_o\omega_1^2 \mathcal{A} E_3 E_2^*$$

$$(\nabla^2 + k_2^2)E_2 = -2\mu_o\omega_2^2 \mathcal{A} E_3 E_1^*$$

$$(\nabla^2 + k_3^2)E_3 = -2\mu_o\omega_3^2 \mathcal{A} E_1 E_2$$



$$\frac{da_1}{dz} = -j\mathcal{A} a_3 a_2^* \exp(-j\Delta k z)$$

$$\frac{da_2}{dz} = -j\mathcal{A} a_3 a_1^* \exp(-j\Delta k z)$$

$$\frac{da_3}{dz} = -j\mathcal{A} a_1 a_2 \exp(j\Delta k z),$$

where

$$\mathcal{A}^2 = 2\hbar\omega_1\omega_2\omega_3\eta^3 \mathcal{A}^2$$

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

Homework : EXERCISE 19.4-2 Energy Conservation.

EXERCISE 19.4-3 Photon-Number Conservation: The Manley-Rowe Relation.

Second-harmonic generation (SHG)

Assuming two collinear waves with perfect phase matching ($\Delta k = 0$),

$$\frac{da_1}{dz} = -j g a_3 a_2^* \exp(-j \Delta k z)$$

$$\frac{da_2}{dz} = -j g a_3 a_1^* \exp(-j \Delta k z)$$

$$\frac{da_3}{dz} = -j g a_1 a_2 \exp(j \Delta k z),$$

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

$$\omega_3 = 2\omega$$

$$k_3 = 2k_1$$

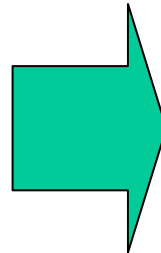
$$\frac{da_1}{dz} = -j g a_3 a_1^*$$

$$\frac{da_3}{dz} = -j \frac{g}{2} a_1 a_1$$

Coupled Equations
(Second-Harmonic
Generation)

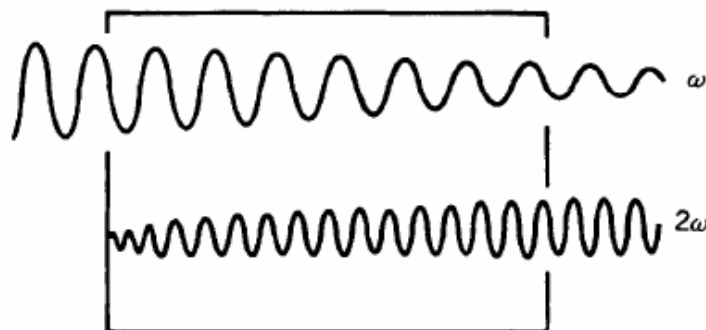
photon-number conservation

$$|a_1(z)|^2 + 2|a_3(z)|^2 = \text{constant}$$

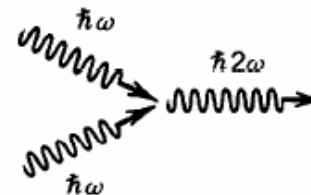


$$a_1(z) = a_1(0) \operatorname{sech} \frac{g a_1(0) z}{\sqrt{2}}$$

$$a_3(z) = -\frac{j}{\sqrt{2}} a_1(0) \tanh \frac{g a_1(0) z}{\sqrt{2}}$$



$Z = 0$



Second-harmonic generation (SHG)

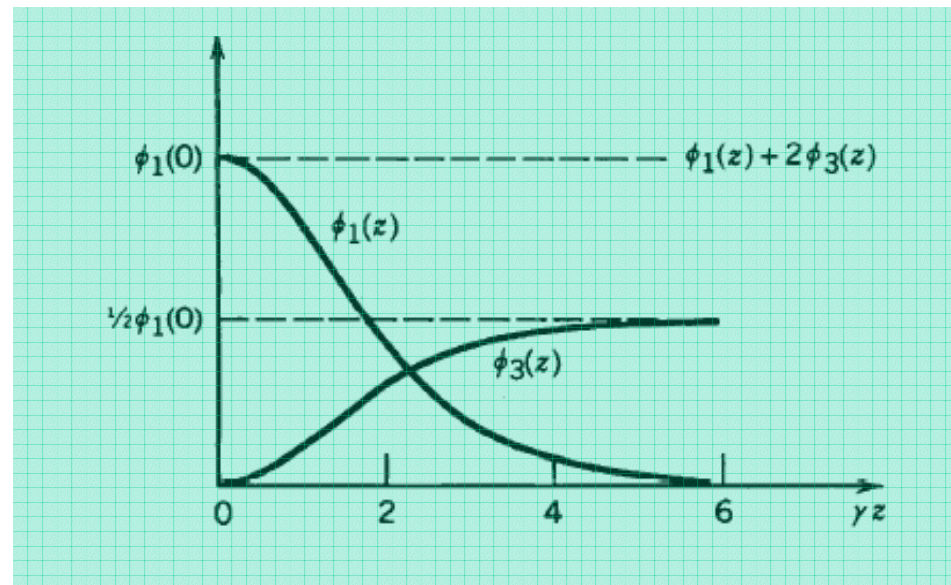
Photon flux densities

$$\phi_1(z) = |a_1(z)|^2 = \phi_1(0) \operatorname{sech}^2 \frac{\gamma z}{2}$$

$$\text{where } \gamma/2 = g a_1(0)/\sqrt{2},$$

$$\phi_3(z) = |a_3(z)|^2 = \frac{1}{2} \phi_1(0) \tanh^2 \frac{\gamma z}{2}$$

$$\begin{aligned} \gamma^2 &= 2g^2 a_1^2(0) = 2g^2 \phi_1(0) \\ &= 8d^2 \eta^3 \hbar \omega^3 \phi_1(0) = 8d^2 \eta^3 \omega^2 I_1(0) \end{aligned}$$



Since $\operatorname{sech}^2 + \tanh^2 = 1$, $\phi_1(z) + 2\phi_3(z) = \phi_1(0)$ is constant.

- photons of wave 1 are converted to half as many photons of wave 3.
- **photon numbers are conserved.**

Second-harmonic generation (SHG)

Efficiency of second-harmonic generation

$$\frac{I_3(L)}{I_1(0)} = \frac{\hbar\omega_3\phi_3(L)}{\hbar\omega_1\phi_1(0)} = \frac{2\phi_3(L)}{\phi_1(0)} = \tanh^2 \frac{\gamma L}{2} \quad \gamma^2 = 8d^2\eta^3\omega^2 I_1(0)$$

For large γL (long cell, large input intensity, or large nonlinear parameter),

all the input power (at frequency ω) has been transformed into power at frequency 2ω ;

all input photons of frequency ω are converted into half as many photons of frequency 2ω .

For small γL

$$\frac{I_3(L)}{I_1(0)} \overset{\text{tanh } x \approx x}{\approx} \frac{1}{4}\gamma^2 L^2 = \frac{1}{2}d^2 L^2 \phi_1(0) = 2d^2\eta^3\hbar\omega^3 L^2 \phi_1(0) = 2d^2\eta^3\omega^2 L^2 I_1(0) = 2\eta_o^3\omega^2 \frac{d^2}{n^3} \frac{L^2}{A} P$$

To maximize the efficiency, we must confine the wave to the smallest possible area A and the largest possible interaction length L .

This is best accomplished with waveguides (planar or channel waveguides or fibers).

Second-harmonic generation (SHG)

Effect of Phase Mismatch $\Delta k \neq 0$

$$\frac{da_1}{dz} = -j\mathcal{G}a_3a_1^* \exp(-j\Delta k z)$$

$$\frac{da_3}{dz} = -j\frac{\mathcal{G}}{2}a_1a_1 \exp(j\Delta k z),$$

For weak-coupling case, $\gamma L \ll 1$,

the fundamental wave $a_1(z)$ varies only slightly with $z \rightarrow a_1(z) \approx a_1(0)$

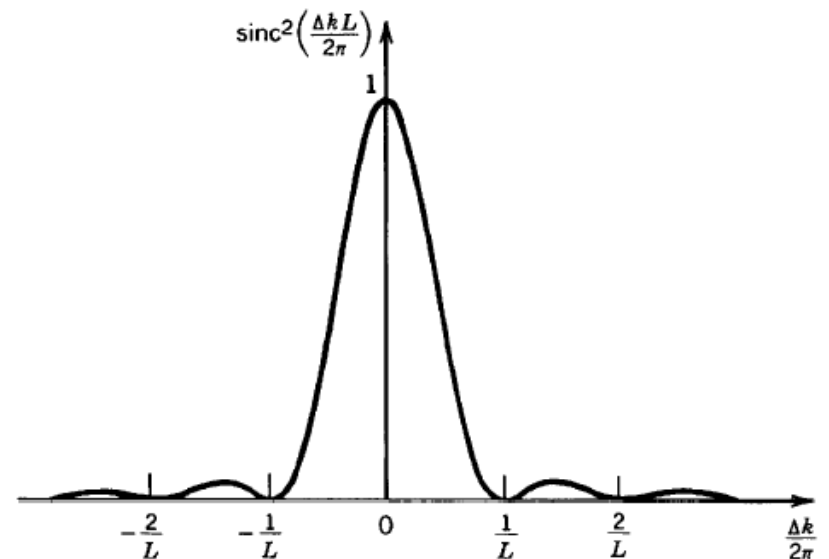
$$\begin{aligned} a_3(L) &= -j\frac{\mathcal{G}}{2}a_1^2(0) \int_0^L \exp(j\Delta k z') dz' \\ &= -\left(\frac{\mathcal{G}}{2\Delta k}\right)a_1^2(0)[\exp(j\Delta k L) - 1] \end{aligned}$$

$$\phi_3(L) = |a_3(L)|^2 = (\mathcal{G}/\Delta k)^2 \phi_1^2(0) \sin^2(\Delta k L/2)$$

The efficiency of second-harmonic generation is

$$\boxed{\frac{I_3(L)}{I_1(0)} = \frac{2\phi_3(L)}{\phi_1(0)} = \frac{1}{2}\mathcal{G}^2 L^2 \phi_1(0) \operatorname{sinc}^2 \frac{\Delta k L}{2\pi}},$$

where $\operatorname{sinc}(x) = \sin(\pi x)/(\pi x)$.



19.6. Anisotropic nonlinear media

polarization vector $\mathcal{P} = (\mathcal{P}_1, \mathcal{P}_2, \mathcal{P}_3)$

$$\mathcal{P}_i = \epsilon_o \sum_j \chi_{ij} \mathcal{E}_j + 2 \sum_{jk} d_{ijk} \mathcal{E}_j \mathcal{E}_k + 4 \sum_{jkl} \chi_{ijkl}^{(3)} \mathcal{E}_j \mathcal{E}_k \mathcal{E}_l, \quad i, j, k, l = 1, 2, 3$$

symmetries $d_{ijk} \xrightarrow{\text{dashed}} d_{Ik} = d_{iK} \xrightarrow{\text{dashed}} 6 \times 3$

$\chi_{ijkl}^{(3)} \xrightarrow{\text{dashed}} \chi_{IK}^{(3)} \xrightarrow{\text{dashed}} 6 \times 6$

Three-Wave Mixing in Anisotropic Second-Order Nonlinear Media

$$P_i(\omega_3) = 2 \sum_{jk} d_{ijk} E_j(\omega_1) E_k(\omega_2), \quad j, k = 1, 2, 3$$

where $E_j(\omega_1)$, $E_k(\omega_2)$, and $P_i(\omega_3)$ are components of these vectors
along the principal axes of the crystal.

If $E_j(\omega_1) = E(\omega_1) \cos \theta_{1j}$ and $E_k(\omega_2) = E(\omega_2) \cos \theta_{2k}$,

where θ_{1j} and θ_{2k} are the angles the vectors $\mathbf{E}(\omega_1)$ and $\mathbf{E}(\omega_2)$ make with the principal axes,

$$P_i(\omega_3) = 2 d_{\text{eff}} E(\omega_1) E(\omega_2), \quad d_{\text{eff}} = \sum_{jk} d_{ijk} \cos \theta_{1j} \cos \theta_{2k}, \quad i, j, k = 1, 2, 3.$$

Phase Matching in Three-Wave Mixing

$$\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2 \quad \text{-----} \rightarrow \quad \omega_3 n_3 \hat{u}_3 = \omega_1 n_1 \hat{u}_1 + \omega_2 n_2 \hat{u}_2$$

As an example, consider second-harmonic generation in a uniaxial crystal with waves traveling in the same direction.

$$\omega_1 = \omega_2 = \omega, \text{ and } \omega_3 = 2\omega$$

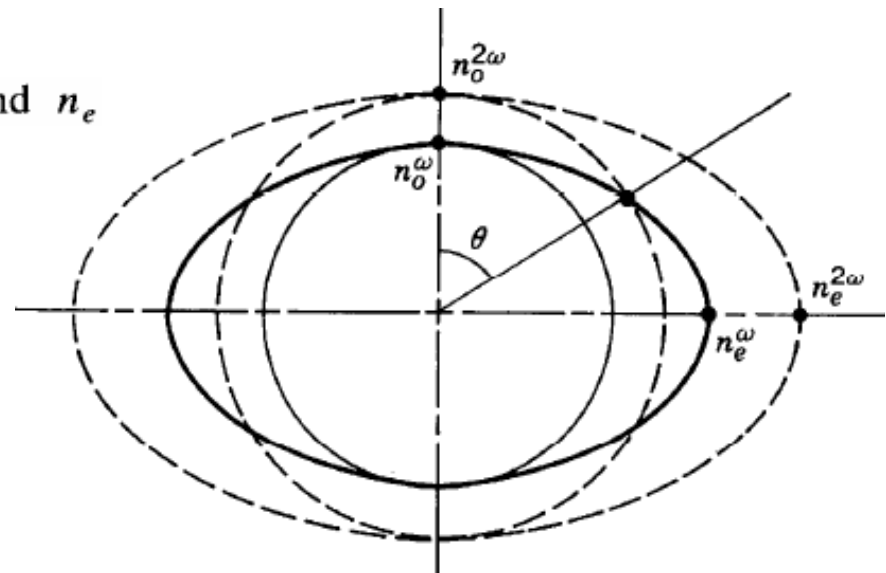
necessary to find the direction and polarizations of the two waves such that the wave of frequency ω has the same refractive index as the wave of frequency 2ω .

in a uniaxial crystal with refractive indices n_o and n_e

$$1/n^2(\theta) = \cos^2 \theta / n_o^2 + \sin^2 \theta / n_e^2$$

To match $n_a = n^\omega(\theta)$ to $n_b = n_o^{2\omega}$,

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



Thus the fundamental wave is an extraordinary wave and the second-harmonic wave is an ordinary wave.