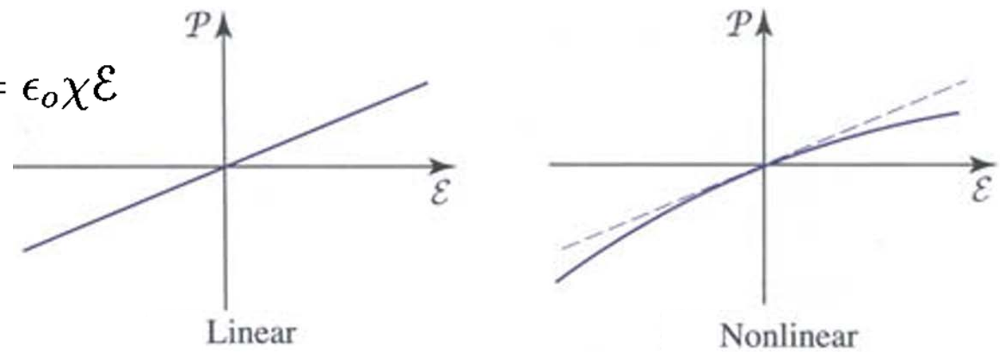
Chapter 21

NONLINEAR OPTICS I

Nonlinear optical media

$$\mathcal{P} = \epsilon_o \chi \mathcal{E}$$



Taylor series expansion of polarization $\mathcal{P}$ about $\mathcal{E}$ = 0:

$$\mathcal{P} = \epsilon_o \chi \mathcal{E} + 2\mathrm{d}\mathcal{E}^2 + 4\chi^{(3)}\mathcal{E}^3 + \cdots = \epsilon_o \chi \mathcal{E} + \mathcal{P}_{\mathrm{NL}}$$

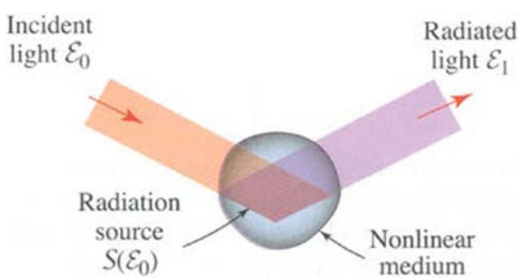
second-order **third-order**

Centrosymmetric media are third-order nonlinear, because d = 0 for them.

Nonlinear wave equation:

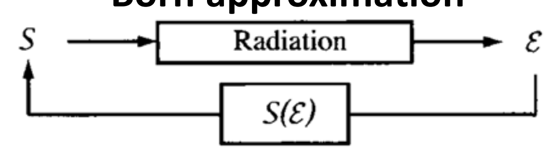
$$\nabla^2 \mathcal{E} - \frac{1}{c^2}\frac{\partial^2 \mathcal{E}}{\partial t^2} = -\mathcal{S}$$

$$\mathcal{S} = -\mu_o \frac{\partial^2 \mathcal{P}_{\mathrm{NL}}}{\partial t^2}$$ – acts as a radiation source



The first Born approximation

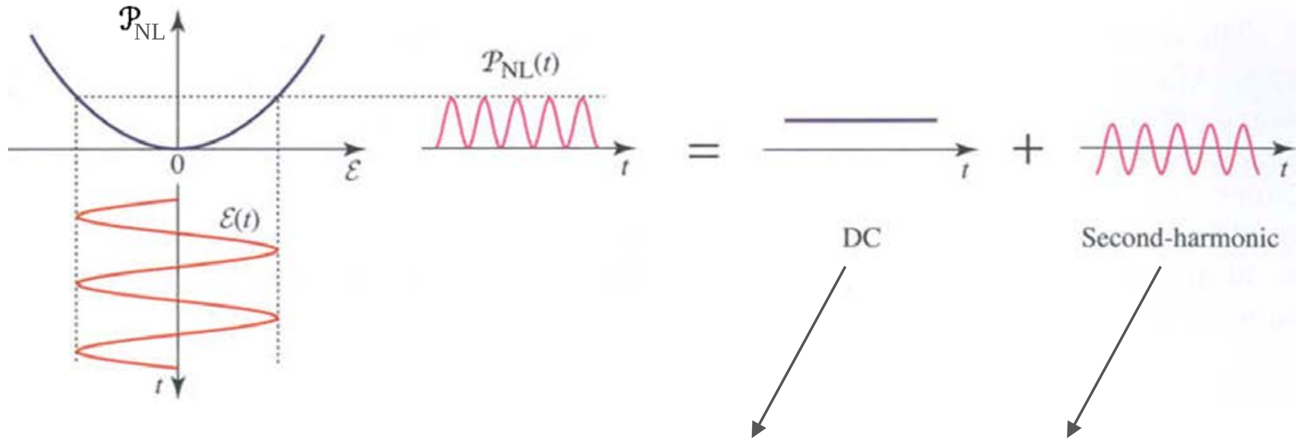
Born approximation



Second-order nonlinear optics

$$\mathcal{P} = \epsilon_o \chi \mathcal{E} + 2\mathrm{d}\mathcal{E}^2$$

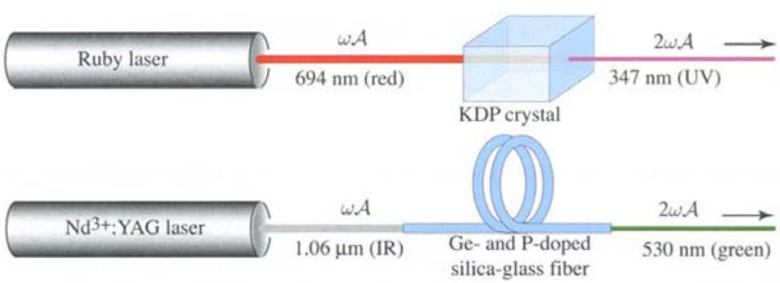
Second-harmonic generation:



$$\mathcal{E}(t) = \mathrm{Re}\{E(\omega)\exp(j\omega t)\} \Rightarrow \mathcal{P}_{NL}(t) = P_{NL}(0) + \mathrm{Re}\{P_{NL}(2\omega)\ \exp(j2\omega t)\}$$

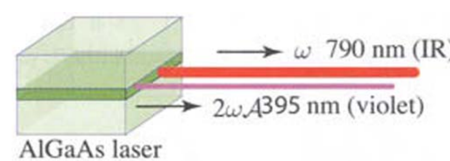
$$P_{NL}(0) = \mathrm{d}\ E(\omega)E^*(\omega)$$

$$P_{NL}(2\omega) = \mathrm{d}\ E^2(\omega)$$

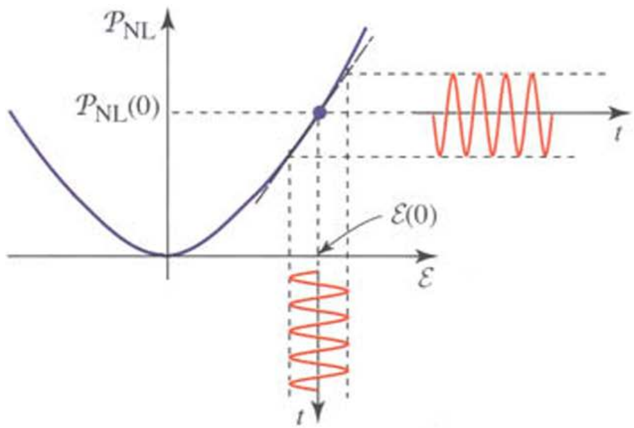


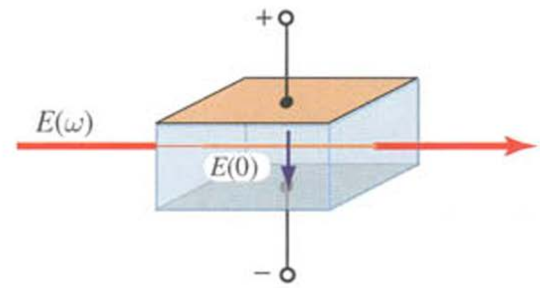
$$\eta_{SHG} = C^2 \frac{L^2}{A} P$$

efficiency (η_{SHG}); length (L); power (P); transverse area (A)



Electro-optic (Pockels) effect





$\mathcal{E}(t) = E(0) + \mathbf{Re}\{E(\omega)\ \exp(j\omega t)\}$, where usually $|E(0)| >> |E(\omega)|$.

$$\mathcal{P}_{\mathrm{NL}}(t) = P_{\mathrm{NL}}(0) + \mathbf{Re}\{P_{\mathrm{NL}}(\omega)\exp(j\omega t)\} + \mathbf{Re}\{P_{\mathrm{NL}}(2\omega)\exp(j2\omega t)\}$$

$$P_{\mathrm{NL}}(0) = \mathrm{d}\left[2E^2(0) + |E(\omega)|^2\right] \qquad P_{\mathrm{NL}}(2\omega) = \mathrm{d}\,E^2(\omega)$$

$$P_{\mathrm{NL}}(\omega) = 4\mathrm{d}\,E(0)E(\omega)$$

$$\Rightarrow\ P_{\mathrm{NL}}(\omega) = \epsilon_o \Delta\chi E(\omega),\ \text{where}\ \Delta\chi = (4\mathrm{d}/\epsilon_o)E(0)$$

$$\Rightarrow\ n^2 = 1 + \chi \ \Rightarrow\ 2n\,\Delta n = \Delta\chi$$

$$\Rightarrow\ \boxed{\Delta n = \frac{2\mathrm{d}}{n\epsilon_o}E(0)}$$

Wave mixing

Frequency conversion in three-wave mixing

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

Frequency components of $\mathcal{P}_{\mathrm{NL}}(t)$:

$$P_{\mathrm{NL}}(0) = \mathrm{d}\left[|E(\omega_1)|^2 + |E(\omega_2)|^2\right]$$

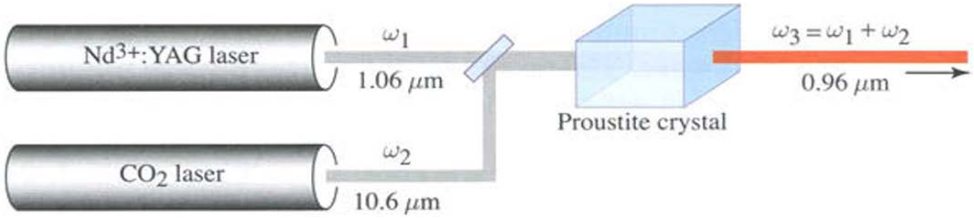
$$P_{\mathrm{NL}}(2\omega_1) = \mathrm{d}\, E(\omega_1)E(\omega_1)$$

$$P_{\mathrm{NL}}(2\omega_2) = \mathrm{d}\, E(\omega_2)E(\omega_2)$$

$$P_{\mathrm{NL}}(\omega_+) = 2\mathrm{d}\, E(\omega_1)E(\omega_2) \quad \longleftarrow \quad \omega_+ = \omega_1 + \omega_2$$

$$P_{\mathrm{NL}}(\omega_-) = 2\mathrm{d}\, E(\omega_1)E^*(\omega_2) \quad \longleftarrow \quad \omega_- = \omega_1 - \omega_2$$

Example:

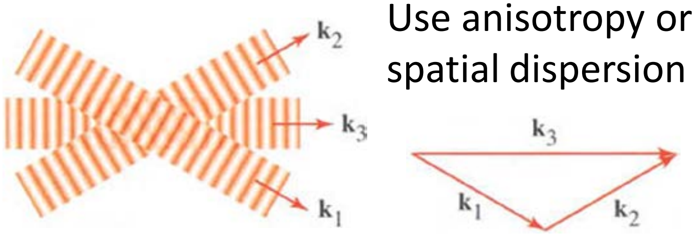


Frequency and phase matching in this case:

$\omega_1 + \omega_2 = \omega_3$ and $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3$.

If the waves co-propagate, $n_1\omega_1 + n_2\omega_2 = n_3\omega_3$.

$n_1 \neq n_2 \neq n_3$ due to dispersion



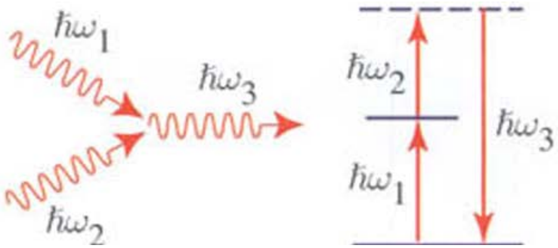
Use anisotropy or spatial dispersion

Wave mixing as a photon interaction process

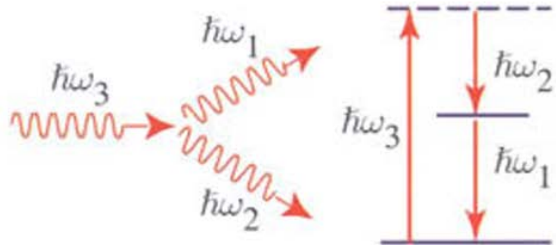
Conservation of energy and momentum:

$$\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_3$$

$$\hbar\mathbf{k}_1 + \hbar\mathbf{k}_2 = \hbar\mathbf{k}_3$$

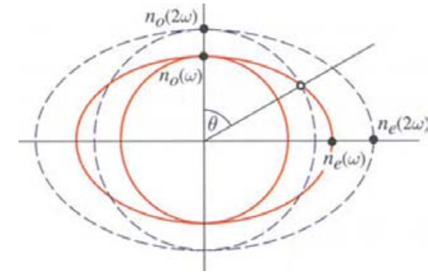


up-conversion (UC)



down-conversion (DC)

Anisotropic crystal



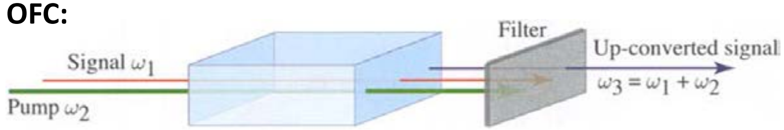
SHG phase matching (type-I)

Photon number conservation = *Manley-Rowe relation*:

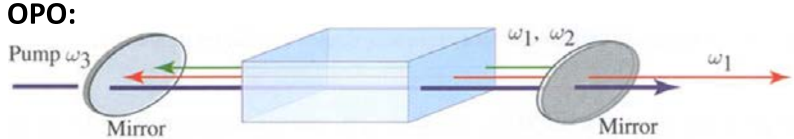
$$\frac{d\phi_1}{dz} = \frac{d\phi_2}{dz} = -\frac{d\phi_3}{dz} \Rightarrow \frac{d}{dz}\left(\frac{I_1}{\omega_1}\right) = \frac{d}{dz}\left(\frac{I_2}{\omega_2}\right) = -\frac{d}{dz}\left(\frac{I_3}{\omega_3}\right)$$

Optical parametric devices

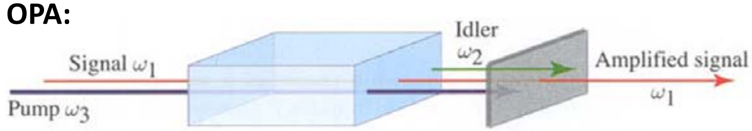
OFC:



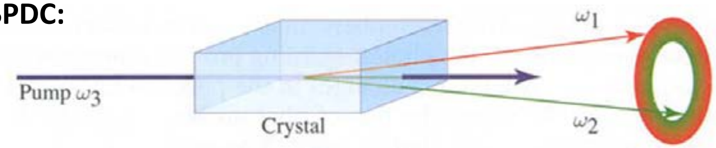
OPO:



OPA:



SPDC:



Tolerable phase mismatch:

$$\Delta\mathbf{k} = \mathbf{k}_3 - \mathbf{k}_1 - \mathbf{k}_2 \neq 0$$

$$P_{\rm NL}(\omega_3) = 2\mathrm{d}E(\omega_1)E(\omega_2) = 2\mathrm{d}A_1A_2\exp[-j(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{r}]$$

$$= \underline{2\mathrm{d}A_1A_2\exp(j\Delta\mathbf{k}\cdot\mathbf{r})}\exp(-j\mathbf{k}_3\cdot\mathbf{r}).$$

$$\Rightarrow\; I_3 \propto \left|\int_V \mathrm{d}A_1A_2\exp(j\Delta\mathbf{k}\cdot\mathbf{r})d\mathbf{r}\right|^2$$

For a plane wave: $I_3 \propto |\int_0^L \exp(j\Delta k\, z)dz|^2 \;=\; L^2\,\mathrm{sinc}^2(\Delta k\, L/2\pi)$

- maximum at $\Delta k = 0$ and vanishing at $L_c = 2\pi/\Delta k$

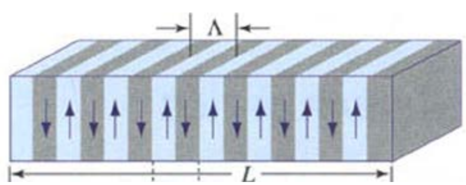
Quasi-phase matching:

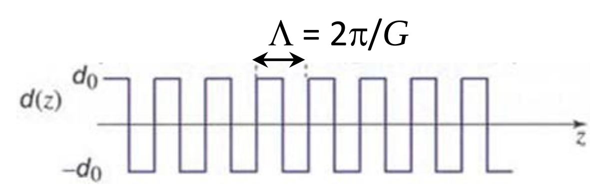
Let d be a periodic function $\mathbf{d}(\mathbf{r}) = \mathrm{d}_o\exp(-j\mathbf{G}\cdot\mathbf{r})$, with $\mathbf{G} = \Delta\mathbf{k}$.

$$\Rightarrow\; I_3 \propto \left|\int_V \mathbf{d}(\mathbf{r})\exp(j\Delta\mathbf{k}\cdot\mathbf{r})d\mathbf{r}\right|^2 \propto \left|\int_V \mathrm{d}_o\;\exp(j\Delta\mathbf{k}\cdot\mathbf{r}-j\mathbf{G}\cdot\mathbf{r})d\mathbf{r}\right|^2$$

$\Rightarrow$ A new phase-matching condition is $\mathbf{k}_1+\mathbf{k}_2+\mathbf{G} = \mathbf{k}_3$.

Example of a periodically *poled ferroelectric crystal*:

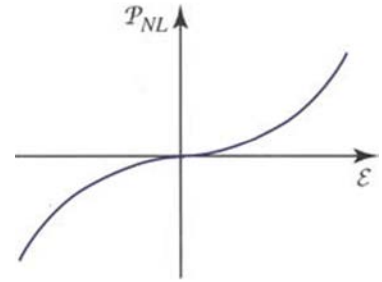




Third-order nonlinear medium

$$\mathcal{P}_{\mathrm{NL}} = 4\chi^{(3)}\mathcal{E}^3$$

Kerr medium



Kerr electro-optic effect:
A steady field $E(0)$ causes a refractive-index change $\Delta n = -\frac{1}{2}\mathfrak{s} n^3 E^2(0)$,

where $\mathfrak{s} = -\dfrac{12}{\epsilon_o n^4}\chi^{(3)}$ is the Kerr coefficient.

Third-harmonic generation:
A monochromatic field $E(\omega)$ leads to terms $P_{\mathrm{NL}}(\omega) = 3\chi^{(3)}|E(\omega)|^2 E(\omega)$ and

$$P_{\mathrm{NL}}(3\omega) = \chi^{(3)} E^3(\omega)$$

Optical Kerr effect:

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

Impedance

$$n^2 = 1 + \chi \;\Rightarrow\; \Delta n = \frac{3\eta}{\epsilon_o n}\chi^{(3)} I \equiv n_2 I$$

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

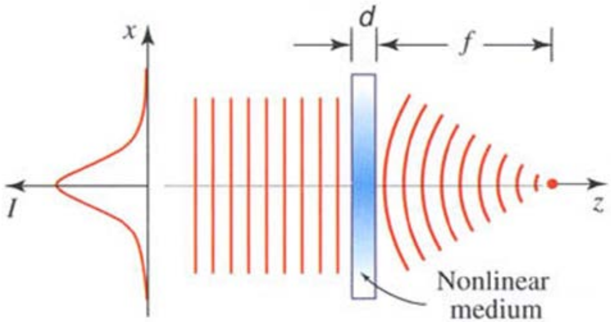
Optical Kerr coefficient

Self-phase modulation due to optical Kerr effect

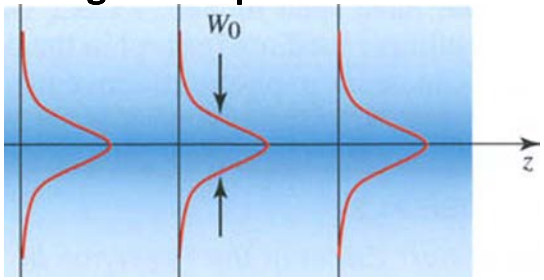
A wave of intensity I travelling over a distance L undergoes a nonlinear phase shift

$$\Delta\varphi = \frac{2\pi}{\lambda_0} n_2 I L.$$

If two waves co-propagate in the medium, a *cross-phase modulation* takes place.
An optical beam “sees” a higher refractive index in the center $\Rightarrow$ *Self-focusing*:



Self-guided spatial soliton



The Helmholtz equation, $[\nabla^2 + (n + n_2 I)^2 k_0^2]E = 0$, can be written in the slowly-varying envelope approximation for $n_2 I \ll n$ as

$$\frac{\partial^2 A}{\partial x^2} + \frac{n_2}{\eta_o} k^2 |A|^2 A = 2jk \frac{\partial A}{\partial z}$$ **- nonlinear Schrödinger equation**

One of its solutions describes a non-diverging beam (*spatial soliton*):

$$A(x,z) = A_0 \operatorname{sech}\left(\frac{x}{W_0}\right) \exp\left(-j\frac{z}{4z_0}\right) \Rightarrow I(x,z) = \frac{|A(x,z)|^2}{2\eta} = \frac{A_0^2}{2\eta} \operatorname{sech}^2\left(\frac{x}{W_0}\right).$$

Four-wave mixing

A superposition of three real-valued waves can be written in terms of their complex amplitudes as

$$\mathcal{E}(t) = \sum_{q=\pm1,\pm2,\pm3} \tfrac{1}{2} E(\omega_q) \exp(j\omega_q t),$$

where $\omega_{-q} = -\omega_q$ and $E(-\omega_q) = E^*(\omega_q)$. This leads to $6^3 = 216$ terms in

$$\mathcal{P}_{\mathrm{NL}}(t) = \tfrac{1}{2}\chi^{(3)} \sum_{q,r,l=\pm1,\pm2,\pm3} E(\omega_q)E(\omega_r)E(\omega_l) \exp[j(\omega_q + \omega_r + \omega_l)t].$$

corrected

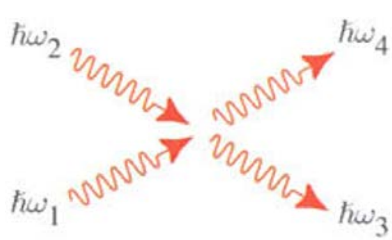
A harmonic component of frequency $\omega_3 + \omega_4 - \omega_1$ has six terms above so that

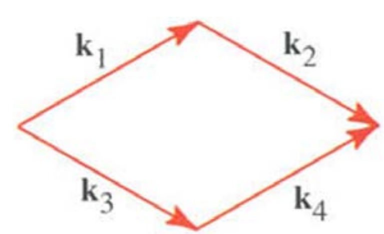
$$P_{\mathrm{NL}}(\omega_1 + \omega_2 - \omega_3) = 6\chi^{(3)} E(\omega_1)E(\omega_2)E^*(\omega_3).$$

Hence, four waves are mixed by the medium, if $\omega_4 = \omega_1 + \omega_2 - \omega_3$. The frequency- and phase-matching conditions are

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

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4.$$





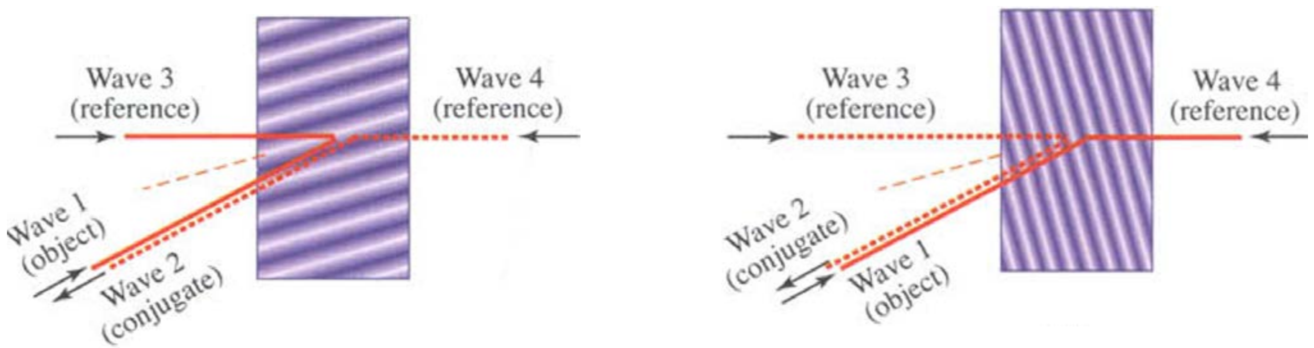
Three-wave mixing and optical phase conjugation

The condition $\omega_1 + \omega_2 = \omega_3 + \omega_4$ is satisfied, if $\omega_3 = \omega_4 \equiv \omega_0$ and therefore $\omega_1 + \omega_2 = 2\omega_0$. This results in *three-wave mixing* that still involves 4 photons.

The frequency-matching condition is satisfied also if $\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega$. This ***degenerate four-wave mixing*** leads to ***phase conjugation***. If two of the waves are counter-propagating, i.e., $\mathbf{k}_4 = -\mathbf{k}_3$, we obtain

$$E_2(\mathbf{r}) \propto A_3 A_4 E_1^*(\mathbf{r}),$$

which means that the generated wave is a conjugate of $E_1(\mathbf{r})$.



Wave restoration:
dynamic holography