Second-order nonlinear processes

Sergey A. Ponomarenko

Dalhousie University

Outline

- Conservation laws in nonlinear optics
- Coupled-wave equations for 2nd-order processes
- Second-harmonic generation (SHG)
- Phase-matching considerations
- Sum-frequency generation (SFG)
- Manley-Rowe relations
- Difference-frequency generation (DFG)

Conservation laws in nonlinear optics

Maxwell's equations for nonlinear nonmagnetic media

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t},$$

$$\nabla \times \mathbf{H} = \mathbf{J} + \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}}{\partial t},$$

$$\nabla \cdot \mathbf{D} = \rho,$$

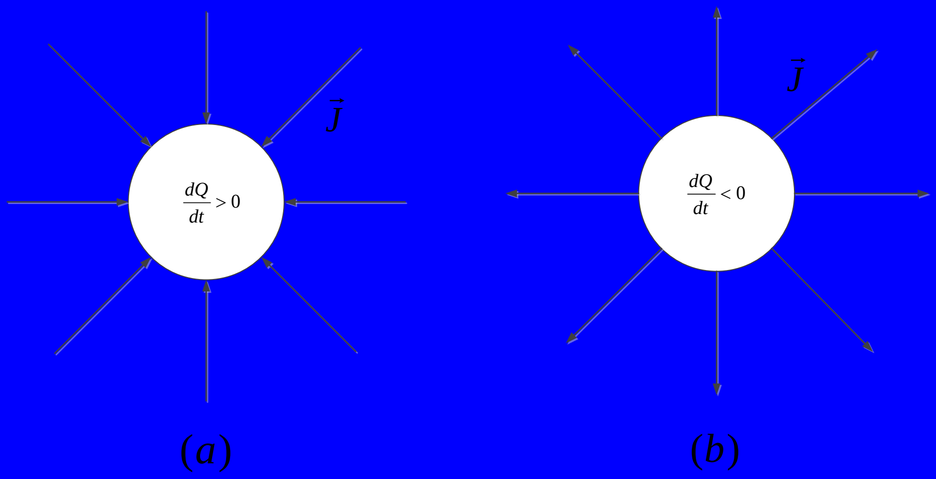
$$\nabla \cdot \mathbf{H} = 0,$$

- Maxwell's equations $\Longrightarrow$ energy conservation for EM fields
- Charge conservation $\Longrightarrow$ fundamental law of nature

Charge conservation

Given a closed volume,

$$\frac{d}{dt}\int dv\rho_v = -\oint d\mathbf{S}\cdot\mathbf{J}$$



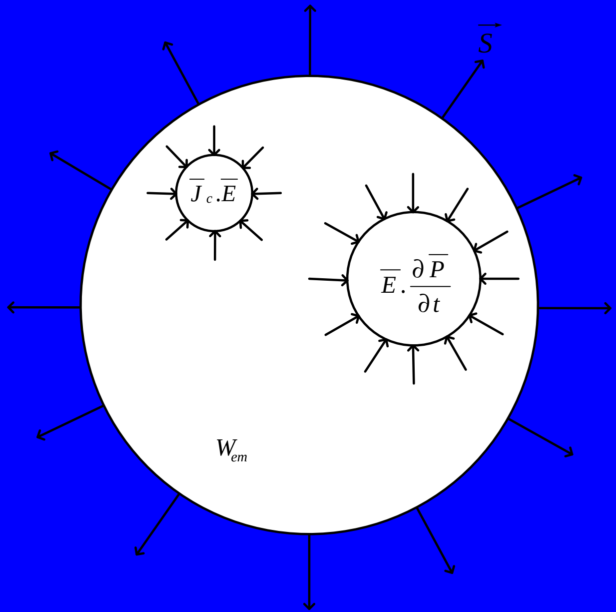
$$\frac{d}{dt}\int dv\rho_v = \int_v dv\frac{\partial \rho_v}{\partial t} = -\oint d\mathbf{S}\cdot\mathbf{J} = -\int dv\nabla\cdot\mathbf{J}$$

implying that

$$\int_v \left(\frac{\partial \rho_v}{\partial t} + \nabla\cdot\mathbf{J}\right) = 0$$

charge conservation in the differential form

$$\frac{\partial \rho_v}{\partial t} + \nabla\cdot\mathbf{J} = 0$$



Integral for of EM energy conservation

$$\frac{dW_{em}}{dt} = -\oint_{\sigma} d\boldsymbol{\sigma} \cdot \mathbf{S} - \int_{v} dv \left(\mathbf{J} \cdot \mathbf{E} + \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t} \right).$$

Differential form of EM energy conservation

$$\frac{\partial w_{em}}{\partial t} + \nabla \cdot \mathbf{S} = -\mathbf{J} \cdot \mathbf{E} - \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t}$$

electromagnetic energy density:

$$w_{em} = \tfrac{1}{2}\varepsilon_0 E^2 + \tfrac{1}{2}\mu_0 H^2$$

energy flux (Poynting vector):

$$\mathbf{S} = \mathbf{E} \times \mathbf{H}$$

Total EM energy within the volume:

$$W_{em} = \int_v dv w_{em},$$

Coupled-wave equations

- nonmagnetic medium $\mu = \mu_0$;
- isotropic medium with no spatial dispersion
- no free charges or currents, $\rho = 0, \mathbf{J} = 0$

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t},$$

$$\nabla \times \mathbf{H} = \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}}{\partial t},$$

$$\nabla \cdot \mathbf{D} = 0, \qquad \nabla \cdot \mathbf{H} = 0$$

Wave equation

$$\nabla \times (\nabla \times \mathbf{E}) = -\varepsilon_0 \mu_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} - \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2}$$

Driving source:

$$\mathbf{P} = \mathbf{P}_L + \mathbf{P}_{NL}$$

Linear electric flux density:

$$\mathbf{D}_L = \varepsilon_0 \mathbf{E} + \mathbf{P}_L = \varepsilon \otimes \mathbf{E}$$

$$\underbrace{\nabla \times (\nabla \times \mathbf{E}) + \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{D}_L}{\partial t^2}}_{\text{wavefield}} = \underbrace{-\mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}}_{\text{source}}$$

Weak guidance approximation:

$$\nabla \times (\nabla \times \mathbf{E}) = \underbrace{\nabla(\nabla \cdot \mathbf{E})}_{\approx 0} - \nabla^2 \mathbf{E}$$

$$\nabla^2 \mathbf{E} - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{D}_L}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

Fundamental field

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{2} \sum_s \tilde{\mathbf{E}}(\mathbf{r}, \omega_s) e^{-i\omega_s t} + c.\ c.$$

Nonlinear Medium Polarization

$$\mathbf{P}_{NL}(\mathbf{r}, t) = \frac{1}{2} \sum_s \tilde{\mathbf{P}}_{NL}(\mathbf{r}, \omega_s) e^{-i\omega_s t} + c.\ c.$$

Wave equation in space-frequency domain:

$$\nabla^2\tilde{\mathbf{E}} + k^2(\omega_s)\tilde{\mathbf{E}} = -\mu_0\omega_s^2\tilde{\mathbf{P}}_{NL}$$

Wave numbers:

$$k^2(\omega_s) = \varepsilon(\omega_s)\mu_0\omega_s^2$$

Electric fields:

$$\tilde{\mathbf{E}}(\mathbf{r},\omega_s) = \mathbf{e}(\omega_s)\mathscr{E}(\mathbf{r}_\perp, z, \omega_s)e^{ik_s z},$$

Induced Medium Polarization:

$$\tilde{\mathbf{P}}_{NL}(\mathbf{r},\omega_s) = \mathbf{e}(\omega_s)\mathscr{P}_{NL}(\mathbf{r}_\perp, z, \omega_s)e^{ik_s z}$$

Slowly-varying envelope approximation:

$$\frac{\partial \mathscr{E}}{\partial z} \ll k_s \mathscr{E}$$

Paraxial coupled-wave equations:

$$2ik_s \frac{\partial \mathscr{E}}{\partial z} + \nabla_\perp^2 \mathscr{E} = -\mu_0 \omega_s^2 \mathscr{P}_{NL}$$

Second-order processes:

$$\mathscr{P}^{(2)}(\mathbf{r}, \omega_s) = \varepsilon_0 c^{(2)} \sum_{ijk} \tilde{\chi}_{ijk}^{(2)}(-\omega_s; \omega_1, \omega_2) e_i(\omega_s)$$
$$\times e_j(\omega_1) e_k(\omega_2) \mathscr{E}(\mathbf{r}_\perp, \omega_1) \mathscr{E}(\mathbf{r}_\perp, \omega_2) e^{i\Delta k z}, \qquad (1)$$

Phase-mismatch

$$\Delta k \equiv k(\omega_1) + k(\omega_2) - k(\omega_s). \qquad (2)$$

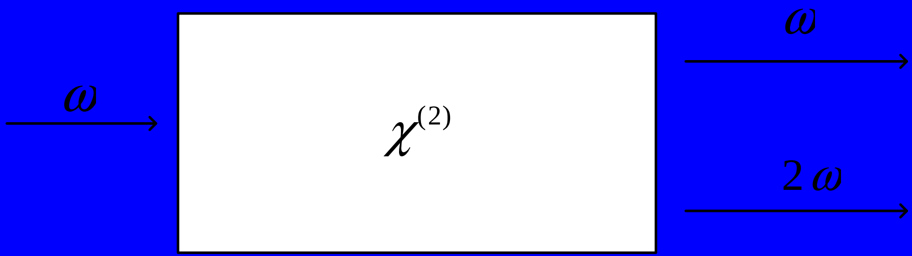
Coupled-wave equations for 2nd-order processes

$$\frac{\partial \mathscr{E}_s}{\partial z} - \frac{i}{2k(\omega_s)} \nabla_\perp^2 \mathscr{E}_s = \frac{i\omega_s^2}{2k(\omega_s)c^2} \times \chi_{eff}^{(2)}(-\omega_s;\omega_1,\omega_2)\mathscr{E}_1\mathscr{E}_2 e^{i\Delta kz} \quad (3)$$

Effective susceptibility:

$$\chi_{eff}^{(2)}(-\omega_s;\omega_1,\omega_2) \equiv c^{(2)} \sum_{ijk} \tilde{\chi}_{ijk}^{(2)}(-\omega_s;\omega_1,\omega_2) \times e_i(\omega_s)e_j(\omega_1)e_k(\omega_2) \quad (4)$$

Second-harmonic generation



Coupled-wave equations:

$$\frac{\partial \mathscr{E}_\omega}{\partial z} - \frac{i}{2k_\omega}\nabla_\perp^2 \mathscr{E}_\omega = \frac{i\omega^2}{2k_\omega c^2}\chi_{eff}^{(2)}(-\omega, 2\omega, -\omega) \times \mathscr{E}_{2\omega}\mathscr{E}_\omega^* e^{-i\Delta kz}. \quad (5)$$

$$\frac{\partial \mathscr{E}_{2\omega}}{\partial z} - \frac{i}{2k_{2\omega}}\nabla_\perp^2 \mathscr{E}_{2\omega} = \frac{i4\omega^2}{2k_{2\omega} c^2}\chi_{eff}^{(2)}(-2\omega, \omega, \omega) \times \mathscr{E}_\omega^2 e^{i\Delta kz}, \quad (6)$$

Phase mismatch:

$$\Delta k = 2k_\omega - k_{2\omega}$$

Plane-wave geometry after rearrangement

$$\frac{d\mathscr{E}_\omega}{dz} = \frac{i\omega^2}{2k_\omega c^2}\chi^{(2)}_{eff}\mathscr{E}_{2\omega}\mathscr{E}^*_\omega e^{-i\Delta kz}.$$

and

$$\frac{d\mathscr{E}_{2\omega}}{dz} = \frac{i\omega^2}{k_{2\omega} c^2}\chi^{(2)}_{eff}\mathscr{E}^2_\omega e^{i\Delta kz}.$$

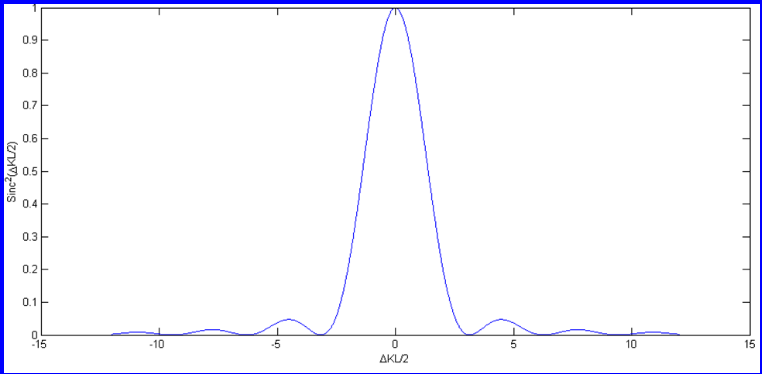
SHG efficiency for a crystal of length L:

$$\eta_{SHG} \equiv \frac{I_{2\omega}(L)}{I_\omega(0)}$$

Undepleted pump approximation

$$\eta_{SHG} \ll 1 \Longrightarrow \mathscr{E}_\omega \approx const!$$

$$I_{2\omega}(L) = \frac{\omega^2 L^2 \chi_{eff}^{(2)2} I_\omega^2}{2\varepsilon_0 n_{2\omega} n_\omega^2 c^3} \mathrm{sinc}^2\left(\frac{\Delta k L}{2}\right),$$



SHG efficiency for undepleted pump

Perfect phase matching

$$\Delta k = 0,$$

$$\eta_{SHG} = \frac{\omega^2 L^2 \chi_{eff}^{(2)2} I_\omega}{2\varepsilon_0 n_{2\omega} n_\omega^2 c^3} \ll 1$$

Pump intensity for focused beam

$$I_\omega = \frac{P}{\pi w_0^2}$$

- moderate-to-high power lasers $P \sim 1$ W,
- $\chi^2_{eff} \sim 5 \times 10^{-23}$ m^2/V^2, for $LiNbO_3$
- crystal length $L \sim 1$ cm
- beam spot size $w_0 \sim 100$ μm, such that $L_\mathrm{d} \sim 10$ cm, $L_\mathrm{d} \ll L$

$$\eta_{SHG} \sim 10^{-3} \ll 1$$

for tightly focused beams, $w_0 \sim 100$ μm say,

$$L_\mathrm{d} \ll L,$$

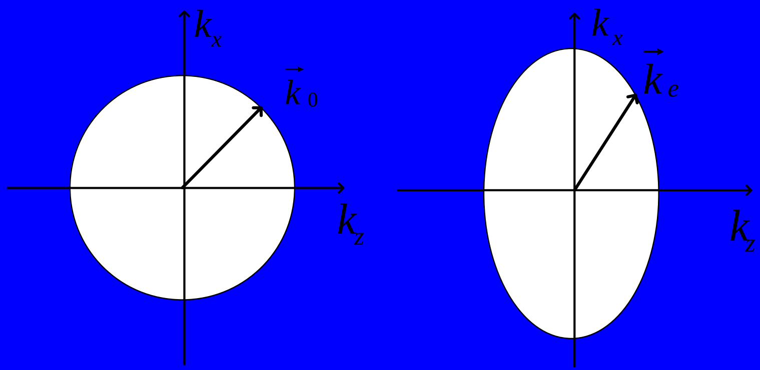
$$\eta_{SHG} \simeq 10\%$$

Phase-matching considerations

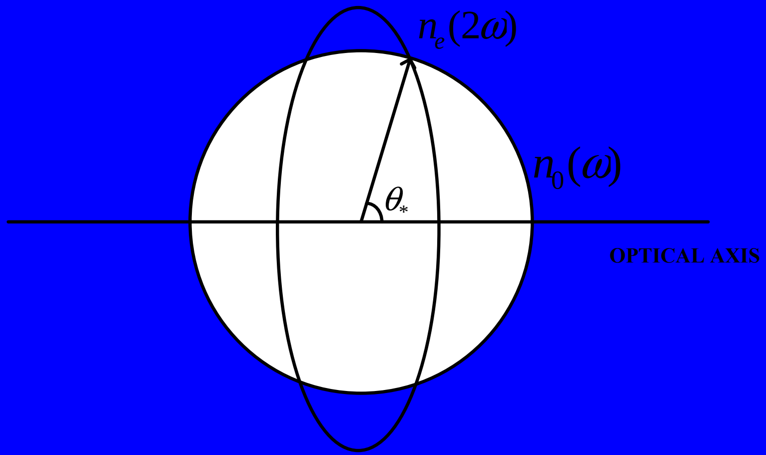
$$\Delta k = 0 \Longrightarrow n(2\omega) = n(\omega)$$

Won't work in isotropic, normally dispersive media

$\Longrightarrow$ anisotropic media



Phase matching in anisotropic media



$$\tan\theta_* = \sqrt{\frac{\frac{1}{\varepsilon_{\|}(2\omega)} - \frac{1}{\varepsilon_{\perp}(\omega)}}{\frac{1}{\varepsilon_{\perp}(\omega)} - \frac{1}{\varepsilon_{\perp}(2\omega)}}}$$

Quasi-phase-matching

- periodic poling of nonlinear susceptibility,

$$\chi^{(2)}(z) = \sum_{m=-\infty}^{\infty} \chi_m^{(2)} e^{i2\pi mz/\Lambda},$$

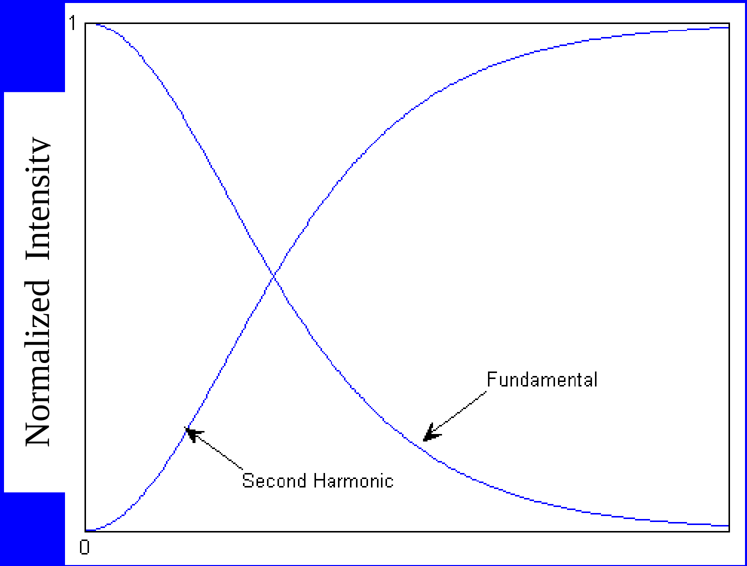
Modified phase mismatch, for $m = 1$

$$\Delta k_{eff} = \Delta k - 2\pi/\Lambda$$

Phase matching is achieved provided:

$$\Lambda = 2\pi/\Delta k$$

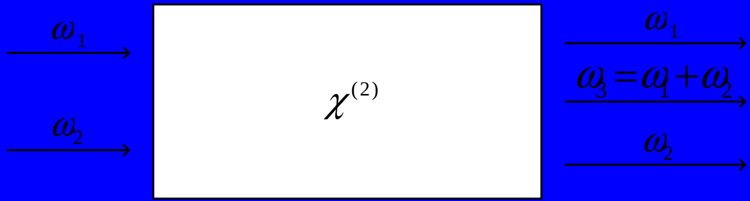
SHG beyond the undepleted pump approximation



$$\zeta = {}^{z}/_{l}$$

Perfect phase matching, $\Delta k = 0$

Sum-frequency generation



$$\frac{\partial \mathscr{E}_1}{\partial z} - \frac{i}{2k_1}\nabla_\perp^2 \mathscr{E}_1 = \frac{i\omega_1^2}{2k_1c^2}\chi_{eff}^{(2)}(-\omega_1;\omega_3,-\omega_2) \times \mathscr{E}_3\mathscr{E}_2^* e^{-i\Delta kz} \qquad (7)$$

$$\frac{\partial \mathscr{E}_2}{\partial z} - \frac{i}{2k_2}\nabla_\perp^2 \mathscr{E}_2 = \frac{i\omega_2^2}{2k_2c^2}\chi_{eff}^{(2)}(-\omega_2;\omega_3,-\omega_1) \times \mathscr{E}_3\mathscr{E}_1^* e^{-i\Delta kz} \qquad (8)$$

Back
Close

$$\frac{\partial \mathscr{E}_3}{\partial z} - \frac{i}{2k_3}\nabla_\perp^2 \mathscr{E}_3 = \frac{i\omega_3^2}{2k_3 c^2}\chi_{eff}^{(2)}(-\omega_3;\omega_1,\omega_2) \times \mathscr{E}_1 \mathscr{E}_2 e^{i\Delta kz} \tag{9}$$

$$\mathscr{E}_j = \mathscr{E}(\rho, z, \omega_j)$$

$$k_j = k(\omega_j), \quad j = 1,2,3$$

Phase mismatch

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

Plane-wave geometry

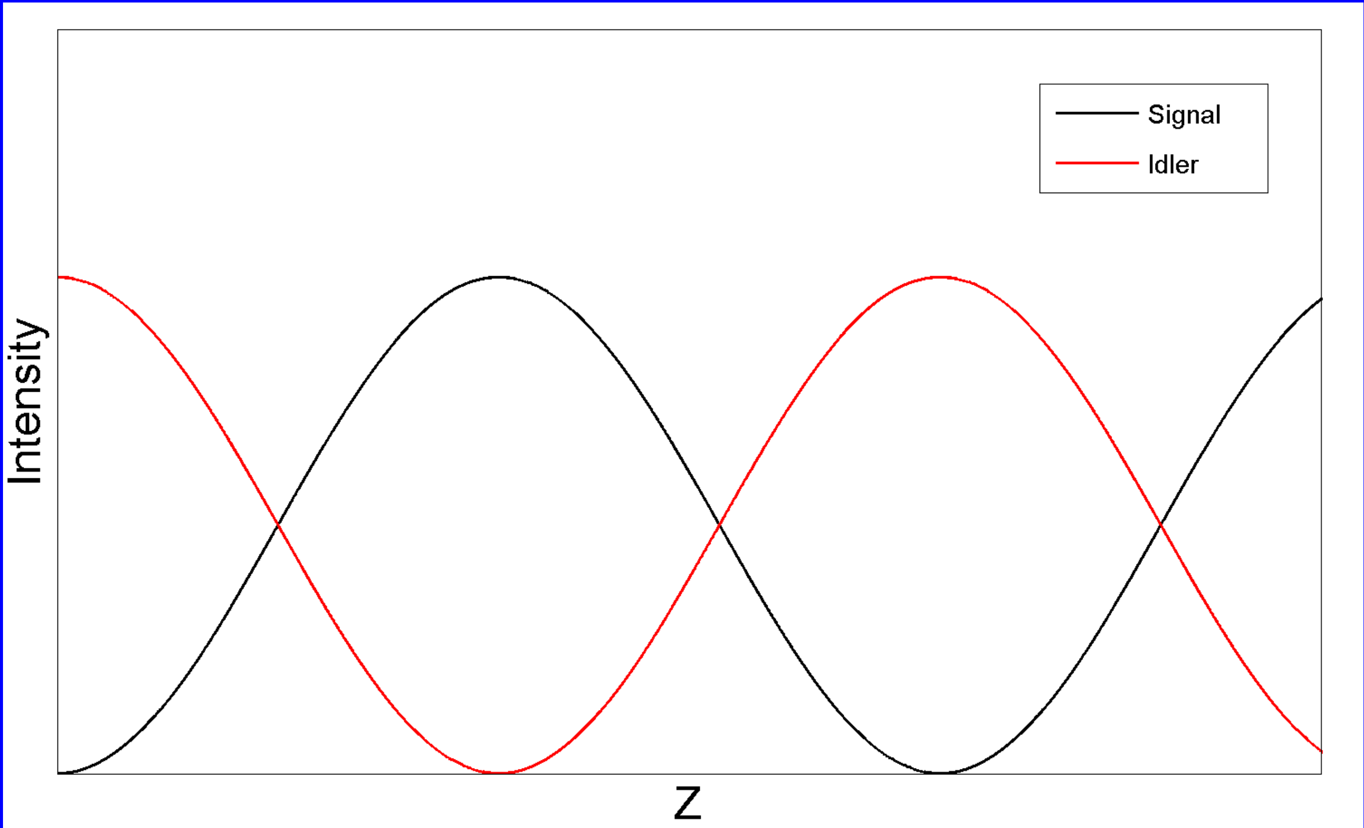
$$\frac{d\mathcal{E}_1}{dz} = \frac{i\omega_1^2}{2k_1c^2}\chi_{eff}^{(2)*}\mathcal{E}_3\mathcal{E}_2^*e^{-i\Delta kz}$$

$$\frac{d\mathcal{E}_2}{dz} = \frac{i\omega_2^2}{2k_2c^2}\chi_{eff}^{(2)*}\mathcal{E}_3\mathcal{E}_1^*e^{-i\Delta kz}$$

$$\frac{d\mathcal{E}_3}{dz} = \frac{i\omega_3^2}{2k_3c^2}\chi_{eff}^{(2)}\mathcal{E}_1\mathcal{E}_2e^{i\Delta kz}$$

Undepleted pump approximation $\Longleftrightarrow$ either pump wave is constant,

$$\mathcal{E}_2 = const$$


Signal
Idler
Intensity
Z

Manley-Rowe relations

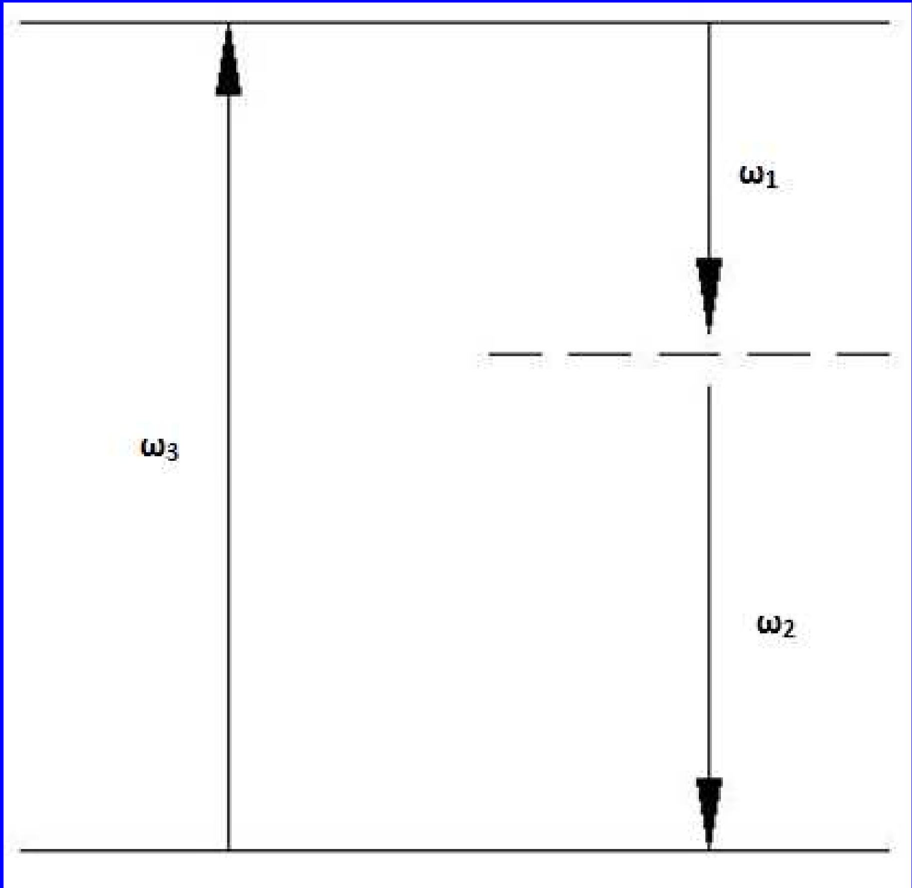
Photon flux conservation:

$$\frac{I_1}{\omega_1} - \frac{I_2}{\omega_2} = \mathscr{M}_1 = const\,,$$

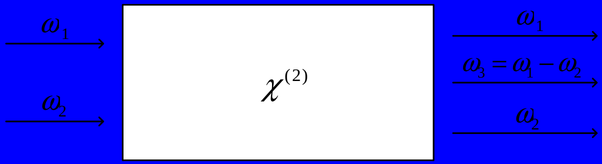
$$\frac{I_1}{\omega_1} + \frac{I_3}{\omega_3} = \mathscr{M}_2 = const\,,$$

$$\frac{I_2}{\omega_2} + \frac{I_3}{\omega_3} = \mathscr{M}_3 = const\,.$$

$$I_j = \frac{\varepsilon_0 n_j c}{2} |\mathscr{E}_j|^2, \qquad j = 1, 2, 3$$


ω1
ω3
ω2

DFG in a plane-wave geometry

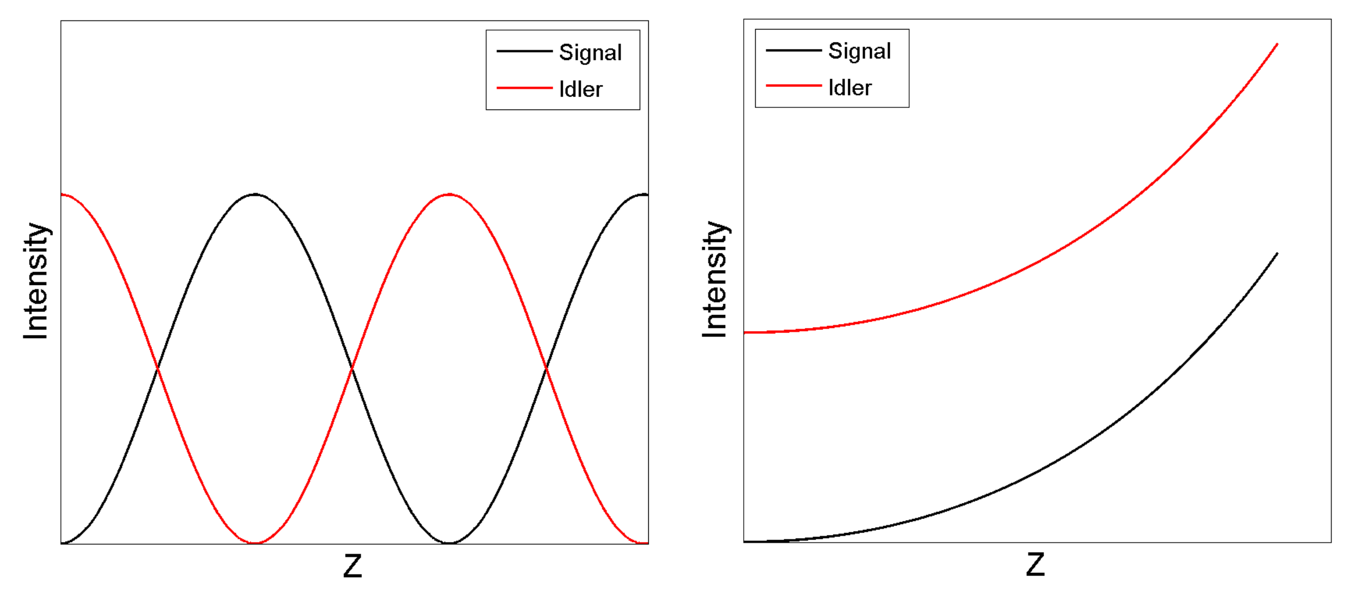


$$\frac{d\mathcal{E}_1}{dz} = \frac{i\omega_1^2}{2k_1c^2}\chi_{eff}^{(2)}\mathcal{E}_2\mathcal{E}_3e^{i\Delta kz}$$

$$\frac{d\mathcal{E}_2}{dz} = \frac{i\omega_2^2}{2k_2c^2}\chi_{eff}^{(2)*}\mathcal{E}_1\mathcal{E}_3^*e^{-i\Delta kz}$$

$$\frac{d\mathcal{E}_3}{dz} = \frac{i\omega_3^2}{2k_3c^2}\chi_{eff}^{(2)*}\mathcal{E}_1\mathcal{E}_2^*e^{-i\Delta kz}$$

Undepleted pump, $\mathscr{E}_1 = const$

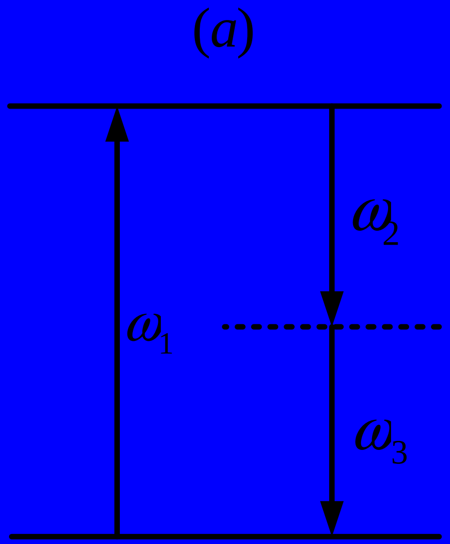


Manley-Rowe relations for DFG

$$\frac{I_1}{\omega_1} + \frac{I_2}{\omega_2} = \mathscr{M}_1 = const\,,$$

$$\frac{I_1}{\omega_1} + \frac{I_3}{\omega_3} = \mathscr{M}_2 = const\,,$$

$$\frac{I_2}{\omega_2} - \frac{I_3}{\omega_3} = \mathscr{M}_3 = const\,.$$



Presence of an idler photon enhances the chances of generating a signal photon $\Longleftrightarrow$ parametric amplification