Nonlinear Optics (WiSe 2019/20)

Lecture 4: November 8, 2019

4 Frequency doubling

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4.4.3 Frequency doubling of pulses

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4.4.3 Frequency doubling of pulses

$$P(z,\omega) = \epsilon_0 d_{eff} \int_{-\infty}^{\infty} E_1(\omega-\omega_1)\, E_1(\omega_1)\, e^{-j(k(\omega-\omega_1)+k(\omega_1))z} d\omega_1.$$

$$k(\omega-\omega_1) = k_0 + \left(\frac{\partial k}{\partial \omega}\right)_{\omega_0} (\omega-\omega_1-\omega_0), \qquad (4.63)$$

$$k(\omega_1) = k_0 + \left(\frac{\partial k}{\partial \omega}\right)_{\omega_0} (\omega_1-\omega_0). \qquad (4.64)$$

With Eqs. (4.63) and (4.64)

$$\Rightarrow k(\omega-\omega_1) + k(\omega_1) = 2k_0 + \frac{1}{\upsilon_{g1}} (\omega - 2\omega_0) \qquad (4.65)$$

where

$$\frac{1}{\upsilon_{g1}} = \left.\frac{1}{\upsilon_g}\right|_{\omega_0} = \left(\frac{\partial k}{\partial \omega}\right)_{\omega_0} \qquad (4.66)$$

is the inverse group velocity. Then the polarization at the sum-frequency is

$$P\left(z,\omega\right)=\epsilon_0 d_{eff} e^{-j\left(2k_0+\frac{1}{\upsilon_{g1}}\left(\omega-2\omega_0\right)\right)z}\int_{-\infty}^{\infty}E_1\left(\omega-\omega_1\right)E_1\left(\omega_1\right)d\omega_1. \tag{4.67}$$

The electric field at frequency ω grows according to Eq. (3.8)

$$\begin{aligned}\frac{\partial E_2\left(z,\omega\right)}{\partial z} &= -\frac{j\omega_0 d_{eff}}{nc_0}e^{-j\left(2k_0+\frac{1}{\upsilon_{g1}}\left(\omega-2\omega_0\right)-k(\omega)\right)z}\times \\ & \int_{-\infty}^{\infty}E_1\left(\omega-\omega_1\right)E_1\left(\omega_1\right)d\omega_{1.}\end{aligned} \tag{4.68}$$

If $E_1(\omega)$ is the spectrum of the pulse centered around ω_0, then the integral will only be non-zero around $\omega \approx 2\omega_0$ The wave number $k\left(\omega\right)$ around $2\omega_0$ is

$$k\left(\omega\right)=k_2+\frac{1}{\upsilon_{g2}}\left(\omega-2\omega_0\right), \tag{4.69}$$

with

$$\frac{1}{\upsilon_{g2}}=\left.\frac{1}{\upsilon_g}\right|_{2\omega_0}=\left(\frac{\partial k}{\partial\omega}\right)_{2\omega_0}. \tag{4.70}$$

For the case of phase matching $(k_2=2k_0)$ and low conversion

$$E_2\left(\ell,\omega\right)=G(\ell,\omega)\cdot F\left(\omega\right) \tag{4.71}$$

where

$$G(\ell,\omega) = -\frac{j\omega_0 d_{eff}}{nc_0} e^{j(\Delta k\ell/2)} \cdot \ell \cdot \left\{ \frac{\sin\frac{\Delta k\ell}{2}}{\Delta k\ell/2} \right\}, \tag{4.72}$$

$$\Delta k = \left(\frac{1}{v_{g1}} - \frac{1}{v_{g2}}\right)(\omega - 2\omega_0), \tag{4.73}$$

and

$$F(\omega) = \int_{-\infty}^{\infty} E_1(\omega-\omega_1) E_1(\omega_1)\, d\omega_1. \tag{4.74}$$

The electric field at the second harmonic can then be written as a Fourier transform. In the time domain we obtain with the convolution theorem

$$\frac{1}{2\pi}\int_{-\infty}^{\infty} G(\omega) F(\omega) e^{j\omega t} d\omega = \int_{-\infty}^{\infty} g(t') f(t-t')\, dt' \tag{4.75}$$

where

$$g(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} G(\omega) e^{j\omega t} d\omega \qquad f(t)=E_1(t)^2 \tag{4.76}$$

$$= \left\{ \begin{array}{ll} \underbrace{e^{j2\omega_0 t}}_{\text{phase}} \frac{\omega_0 d_{eff}}{4nc_0} \frac{1}{\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)}, & 0 < t < \left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell \\ 0, & \text{elsewhere} \end{array} \right\}$$

For a fundamental wave $E_1(t) = A_1(t)\cos(\omega_0 t - k_0 z)$ we obtain a second harmonic wave $E_2(\ell, t) = A_2(\ell, t)\cos(2\omega_0 t - 2k_0 z)$

$$A_2(\ell, t) = \frac{\omega_0 d_{eff}}{4nc_0} \frac{1}{\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)} \int_0^{\ell/v_{g2} - \ell/v_{g1}} A_1^2(t - t')\, dt', \tag{4.77}$$

where $A_2(\ell, t)$ is the envelope of the generated second-harmonic pulse obtained by a convolution of a squared input field and a rectangularly shaped pulse of duration $\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell$. In the limit $\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell \to 0$, we obtain

large doubling bandwidth

$$A_2(\ell, t) = \frac{\omega_0 d_{eff}}{4nc_0} \cdot \ell \cdot A_1^2(t). \tag{4.78}$$

In the case of $\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell \gg t_p =$ pulse length, we obtain from Eq. (4.77) a rectangularly shaped pulse with duration

very small doubling bandwidth

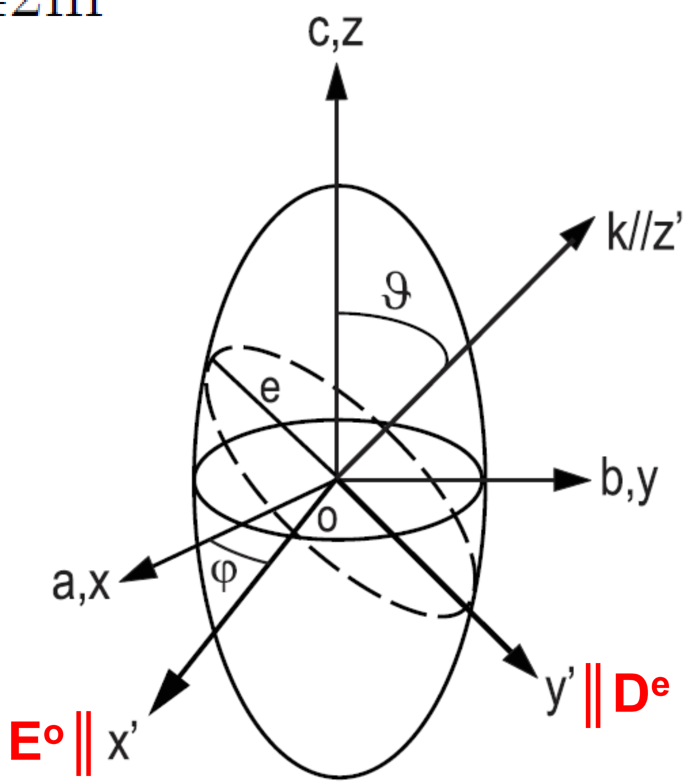
$$\ell \left\{ \left.\frac{1}{v_g}\right|_{2\omega} - \left.\frac{1}{v_g}\right|_{\omega} \right\}.$$

4.4.3 Effective nonlinear coefficients

Fig. 4.12. The d tensor of the crystal in a coordinate system $(\mathbf{x}, \mathbf{y}, \mathbf{z})$ aligned with the main axis $(\mathbf{a}, \mathbf{b}, \mathbf{c})$ of the index ellipsoid is in diagonal form. For the purpose of phase matching the crystal is rotated such that the beams propagate in direction $\mathbf{z}'$ of a new coordinate system $(\mathbf{x}', \mathbf{y}', \mathbf{z}')$. The new coordinate system follows from the old one by two transformations, a rotation around the $\mathbf{z}$-axis by an angle φ and another rotation around the $\mathbf{x}'$-axis by an angle $-\vartheta$. The transformation of a vector $\mathbf{u}$ from the old to the new coordinate system

point group $\bar{4}2m$

KDP

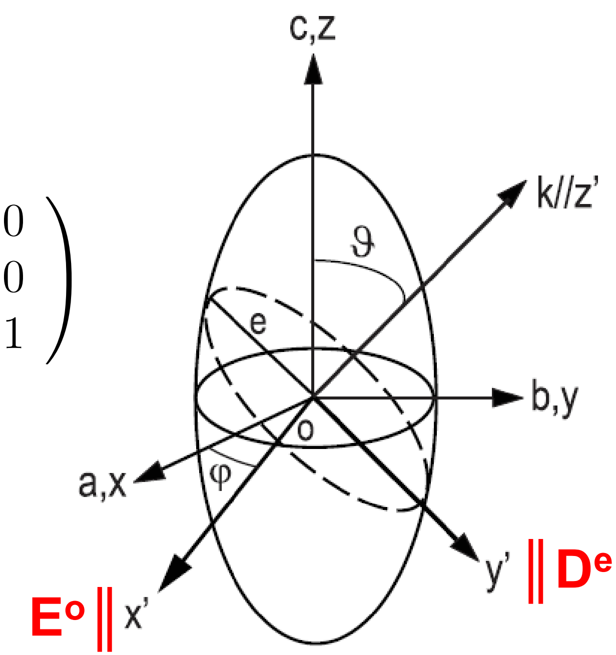


4.4.3 Effective nonlinear coefficients

$$\begin{pmatrix} u_{x'} \\ u_{y'} \\ u_{z'} \end{pmatrix} = \mathbf{T} \cdot \begin{pmatrix} u_x \\ u_y \\ u_z \end{pmatrix} \tag{4.79}$$

with the transformation matrix **T**

$$\begin{aligned} \mathbf{T} &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\vartheta & -\sin\vartheta \\ 0 & \sin\vartheta & \cos\vartheta \end{pmatrix} \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix} \\ &= \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi\cos\vartheta & \cos\varphi\cos\vartheta & -\sin\vartheta \\ -\sin\varphi\sin\vartheta & \cos\varphi\sin\vartheta & \cos\vartheta \end{pmatrix}. \end{aligned}$$



The inverse is

$$\mathbf{T}^{-1} = \mathbf{T}^T = \begin{pmatrix} \cos\varphi & -\sin\varphi\cos\vartheta & -\sin\varphi\sin\vartheta \\ \sin\varphi & \cos\varphi\cos\vartheta & \cos\varphi\sin\vartheta \\ 0 & -\sin\vartheta & \cos\vartheta \end{pmatrix}. \tag{4.81}$$

The fundamental and second-harmonic waves are ordinary or extraordinary waves. The ordinary wave, $(\mathbf{E}\|\mathbf{D})$, is polarized along the x'-axis

$$\mathbf{E}^o = \hat{E}^o \cdot \mathbf{x}' = \hat{E}^o \left(\cos\varphi \cdot \mathbf{x} + \sin\varphi \cdot \mathbf{y}\right) \tag{4.82}$$

The dielectric displacement of the extraordinary beam ($\mathbf{E} \nparallel \mathbf{D}$), is polarized along the y'-axis

$$\mathbf{D}^e = D^e \cdot \mathbf{y}' = D^e \left(-\sin\varphi\cos\vartheta \cdot \mathbf{x} + \cos\varphi\cos\vartheta \cdot \mathbf{y} - \sin\vartheta \cdot \mathbf{z}\right). \tag{4.83}$$

There are two possible ways to determine the effective nonlinear coefficient. One way is by transforming the d tensor to a new coordinate system or by substitution of the fundamental and second-harmonic waves in the old coordinate system and decomposing the second-harmonic fields. For example, for frequency doubling with KDP, which is a negative uniaxial crystal belonging to the point group $\bar{4}2$m, with type-I phase matching:

$$\begin{aligned} \text{fundamental} &: \quad \mathbf{E}(\omega) = \mathbf{E}^o \| \mathbf{D}^o \\ \text{second harmonic} &: \quad \mathbf{D}(2\omega) = \mathbf{D}^e \end{aligned}$$

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \varepsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \cdot \begin{bmatrix} E_x(\omega)^2 \\ E_y(\omega)^2 \\ E_z(\omega)^2 \\ 2E_y(\omega)E_z(\omega) \\ 2E_x(\omega)E_z(\omega) \\ 2E_x(\omega)E_y(\omega) \end{bmatrix}$$

type-I PM

type-II PM

$$= \varepsilon_0 \begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix} \cdot \begin{bmatrix} \cos^2\varphi \\ \sin^2\varphi \\ 0 \\ 0 \\ 0 \\ \sin(2\varphi) \end{bmatrix} \hat{E}^{o2}$$

$2\cos\varphi\sin\varphi$

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \varepsilon_0 \begin{bmatrix} 0 \\ 0 \\ d_{36}\sin(2\varphi) \end{bmatrix} \hat{E}^{o2}$$

In the new system this corresponds to the polarization multiplication with **T**

$$\begin{bmatrix} P_{x'}^{(2)}(2\omega) \\ P_{y'}^{(2)}(2\omega) \\ P_{z'}^{(2)}(2\omega) \end{bmatrix} = \varepsilon_0 d_{36}\sin(2\varphi) \begin{bmatrix} 0 \\ -\sin\vartheta \\ \cos\vartheta \end{bmatrix} \hat{E}^{o2} \qquad (4.84)$$

since the polarization $P_{y'}^{(2)}(2\omega)$ is related to the dielectric displacement of the extraordinary beam. To see that, we would need to rederive Eq. (3.8) in non-isotropic media for the dielectric displacement, instead of the electric fields

$$d_{eff} = -d_{36} \sin(2\varphi) \sin\vartheta. \tag{4.85}$$

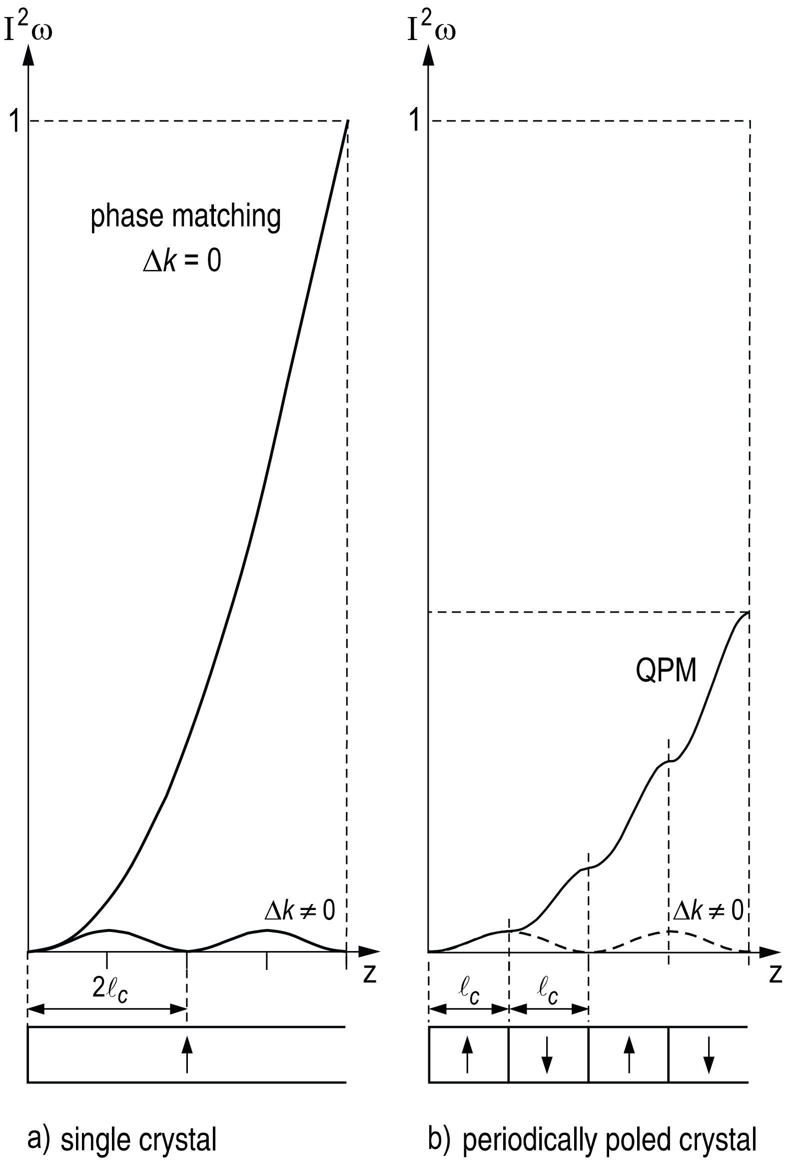
Because of Kleinman symmetry $d_{36} = d_{14}$. The effective nonlinear coefficients for type-I phase matching for the different point groups are given in Table 4.3.

crystal class	$2e \to o$	$2o \to e$
6,4	0	$d_{15} \sin\vartheta$
622,422	0	0
6mm,4mm	0	$d_{15} \sin\vartheta$
$\bar{6}$m2	$d_{22} \cos^2\vartheta \cos 3\varphi$	-$d_{22} \cos\vartheta \sin 3\varphi$
3m	$d_{22} \cos^2\vartheta \cos 3\varphi$	$d_{15} \sin\vartheta - d_{22} \cos\vartheta \sin 3\varphi$
$\bar{6}$	$(d_{11} \sin 3\varphi + d_{22} \cos 3\varphi) \cos^2\vartheta$	$(d_{11} \cos 3\varphi - d_{22} \sin 3\varphi) \cos\vartheta$
3	$(d_{11} \sin 3\varphi + d_{22} \cos 3\varphi) \cos^2\vartheta$	$d_{15} \sin\vartheta + (d_{11} \cos 3\varphi - d_{22} \sin 3\varphi) \cos\vartheta$
32	$d_{11} \sin 3\varphi \cos^2\vartheta$	$d_{11} \cos 3\varphi \cos\vartheta$
$\bar{4}$	$(d_{14} \cos 2\varphi - d_{15} \sin 2\varphi) \sin 2\vartheta$	$-(d_{14} \cos 2\varphi + d_{15} \cos 2\varphi) \sin\vartheta$
$\bar{4}$2m	$d_{14} \cos 2\varphi \sin 2\vartheta$	$-d_{14} \sin 2\varphi \sin\vartheta$

Table 4.3: Effective conversion coefficient d_{eff}, if Kleinman symmetry is valid.

4.4.5 Quasi-phase matching (QPM)

Sometimes to achieve phase matching of a nonlinear process in the desired wavelength range is not possible by birefringence only. In that case, or for achieving a collinear interaction of waves, one can use quasi-phase matching (QPM), a technique introduced by N. Bloembergen, Nobel Prize in Physics 1981 (J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interactions between Light Waves in a Nonlinear Dielectric," Phys. Rev. **127**, 6



high technological relevance!

custom-engineer phase matching
e.g., mid-IR, THz generation

fan-out QPM gratings

chirped QPM gratings

waveguide QPM devices etc.

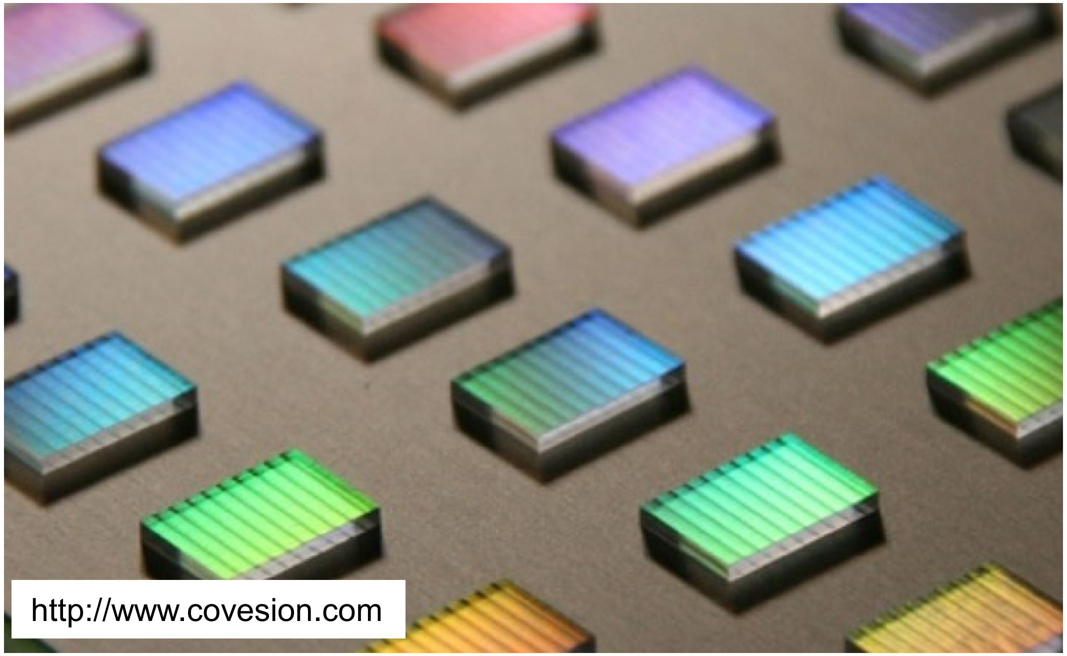


Figure 4.13: Growth of second harmonic as a function of distance z in a crystal for different cases: a) homogeneous crystal and b) periodically poled crystal.

occurs. Due to phase mismatch the second harmonic runs out of phase with the driving wave and therefore the generating polarization. If the sign of the nonlinearity is switched in the second layer, a phase advance by π is introduced in the driving polarization, which rephases it with the already present second harmonic and the process continues with maximum efficiency, see Fig. 4.13.

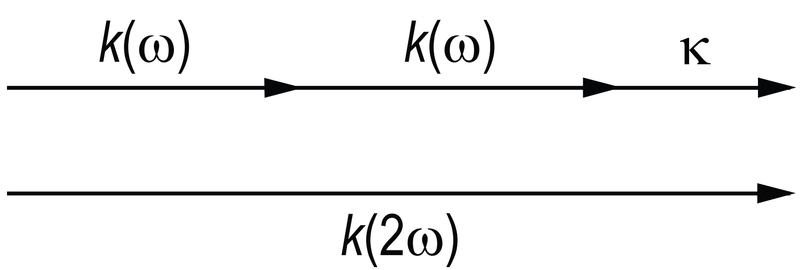
$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c} d_{eff}(z)\hat{E}(\omega)\hat{E}(\omega)\mathrm{e}^{j(k(2\omega)-2k(\omega))z}. \tag{4.86}$$

Since the spatial modulation is periodic, we can represent it as a Fourier series

$$d_{eff}(z) = \sum_{m=-\infty}^{+\infty} d_m e^{jm\kappa z}. \tag{4.87}$$

If the period of the nonlinear coefficient corresponds to twice the coherence length at a given frequency, i.e., $\kappa = k(2\omega) - 2k(\omega)$, then SHG is rephased and grows over multiple periods on average like

$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c} d_{-1}\hat{E}(\omega)\hat{E}(\omega) \tag{4.88}$$



$\Delta k_{total} = \Delta k_{process} + 2\pi/\Lambda(z)$

$\Lambda(z)$ grating period

no walk-off

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$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c} d_{eff}(z)\hat{E}(\omega)\hat{E}(\omega)\mathrm{e}^{j(k(2\omega)-2k(\omega))z}. \tag{4.86}$$

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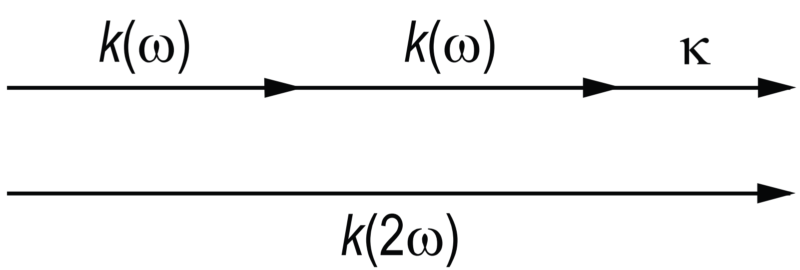

$\Delta k_{total} = \Delta k_{process} + 2\pi/\Lambda(z)$

$\Lambda(z)$ grating period

no walk-off

4.5 Optical rectification

Beside frequency doubling, the $\chi^{(2)}$ nonlinearity also gives rise to optical rectification, that results in a DC voltage in the nonlinear optical medium

$$P_i(0) = \varepsilon_0 \chi_{ijk}(0; \omega_1, -\omega_1) \hat{E}_j(\omega_1) \hat{E}_k^*(\omega_1). \tag{4.89}$$

Due to dispersion, in general

$$\chi_{ijk}(0 : \omega_1, -\omega_1) \neq \chi_{ijk}(2\omega : \omega_1, \omega_1), \tag{4.90}$$

but due to the symmetry relations for χ in lossless media, it holds

$$\chi_{ijk}(0 : \omega_1, -\omega_1) = \chi_{kji}(\omega_1 : \omega_1, 0). \tag{4.91}$$

This ensures that the coefficients for optical rectification are the same as for the Pockels effect. Optical rectification can be used to generate short THz pulses via rectification of femtosecond laser pulses.

4.6 Manley-Rowe relations

Three plane waves propagating in z-direction with frequencies ω_1, ω_2, and ω_3, and interacting via a $\chi^{(2)}$ nonlinearity, can be described by the coupled equations

$$\frac{d\hat{E}(\omega_1)}{dz} = -j\kappa_1 \hat{E}(\omega_3)\hat{E}^*(\omega_2)e^{-j\Delta kz}, \tag{4.92}$$

$$\frac{d\hat{E}(\omega_2)}{dz} = -j\kappa_2 \hat{E}(\omega_3)\hat{E}^*(\omega_1)e^{-j\Delta kz}, \tag{4.93}$$

$$\frac{d\hat{E}(\omega_3)}{dz} = -j\kappa_3 \hat{E}(\omega_1)\hat{E}(\omega_2)e^{+j\Delta kz}, \tag{4.94}$$

with coupling coefficients and difference wave number

$$\kappa_i = \omega_i d_{eff}/n_i c_0, \qquad \text{and} \qquad \Delta k = k_3 - k_1 - k_2. \tag{4.95}$$

We multiply Eq. (4.92) by $n_i c_0 \varepsilon_0 \hat{E}^*(\omega_1)/2$, and add the complex conjugated part, thus obtaining

$$\left(\frac{1}{\omega_1}\right)\frac{dI(\omega_1)}{dz} = \frac{j\varepsilon_0 d_{eff}}{2}\hat{E}(\omega_3)\hat{E}^*(\omega_2)\hat{E}^*(\omega_1)e^{-j\Delta kz} + c.c.$$

We again assume a lossless medium, i.e., $d_{eff} = d^*_{eff}$, and treat Eqs. (4.93), (4.94) similar to Eq. (4.92), and obtain

$$\frac{1}{\omega_1}\frac{dI(\omega_1)}{dz} = \frac{1}{\omega_2}\frac{dI(\omega_2)}{dz} = -\frac{1}{\omega_3}\frac{dI(\omega_3)}{dz}. \tag{4.96}$$

I.e., for each photon, that is created (annihilated) at frequency ω_3, one photon at frequency ω_1 and one photon at frequency ω_2 must be annihilated (created). The corresponding spatial variations of the intensities $\frac{dI(\omega_i)}{dz}$ scale with the frequencies ω_i. This is an interesting result, because no quantum-mechanical treatment has been used to obtain it. Nevertheless, this classical nonlinear electrodynamical treatment already strongly suggests a photon hypothesis $E = n \cdot h\nu$.

4.7 Sum-frequency generation (SFG)

If the nonlinear medium is irradiated by two input fields with frequencies ω_1 and ω_2, it is possible to to generate the sum frequency $\omega_3 = \omega_1 + \omega_2$ via a $\chi^{(2)}$-process. The corresponding coupled-wave equations describing the amplitudes at the three frequencies are

$$\frac{\partial \hat{E}(\omega_3)}{\partial z} = -j\kappa_3 \hat{E}(\omega_1)\, \hat{E}(\omega_2)\, e^{j\Delta kz} \tag{4.97}$$

$$\frac{\partial \hat{E}(\omega_2)}{\partial z} = -j\kappa_2 \hat{E}(\omega_3)\, \hat{E}^*(\omega_1)\, e^{-j\Delta kz} \tag{4.98}$$

$$\frac{\partial \hat{E}(\omega_1)}{\partial z} = -j\kappa_1 \hat{E}(\omega_3)\, \hat{E}^*(\omega_2)\, e^{-j\Delta kz}, \tag{4.99}$$

with $\Delta k = k(\omega_3) - k(\omega_1) - k(\omega_2)$ and $\kappa_i = \omega_i d_{eff}/n_i c_0$. In the special case $\omega_1 = \omega_2$, we again obtain frequency doubling. In the low-conversion case, we can solve Eqs. (4.97)-(4.99) in an analogous manner to the case of frequency doubling. Assuming $\hat{E}(\omega_2)$ and $\hat{E}(\omega_1)$ to be constant, we obtain

$$I(\omega_3, \ell) = \frac{2\kappa_3^2 n_3}{n_1 n_2 c_0 \varepsilon_0} \cdot \ell^2 I(\omega_1)\, I(\omega_2) \left\{ \frac{\sin \Delta k\ell/2}{\Delta k\ell/2} \right\}^2 . \tag{4.100}$$

One important spectroscopic application of sum-frequency generation is the up-conversion of a weak signal at frequency ω_1 with a strong signal at frequency ω_2. This can be used to time-resolve weak light-emission dynamics or to convert a signal in the far infrared into the visible spectral range, where much better photodetectors are available. By using a strong signal at ω_2, the weak signal can even significantly be enhanced. If the pump signal at frequency ω_2 is a short pulse, a short slice can temporally be "gated" out of the input signal. Under the assumption of a strong pump signal at ω_2 and phase matching, we obtain

$$\frac{\partial \hat{E}(\omega_3)}{\partial z} = -\left\{ j\kappa_3 \hat{E}(\omega_2) \right\} \hat{E}(\omega_1), \tag{4.101}$$

$$\frac{\partial \hat{E}(\omega_1)}{\partial z} = -\left\{ j\kappa_1 \hat{E}^*(\omega_2) \right\} \hat{E}(\omega_3). \tag{4.102}$$

In addition, the boundary conditions $E(\omega_3, z = 0) = 0$ and $E(\omega_1, z = 0) = E_0(\omega_1)$ apply. Since the system (4.101)-(4.102) is linear, we try an exponential ansatz of the form $\hat{E}(\omega_{1,3}, z) = \hat{E}_0(\omega_{1,3})\, \mathrm{e}^{\pm j\gamma z}$. With this ansatz we obtain from (4.101)-(4.102)

$$\pm j\gamma \hat{E}_0(\omega_3) + \left\{ j\kappa_3 \hat{E}_0(\omega_2) \right\} \hat{E}_0(\omega_1) = 0$$

$$\left\{ j\kappa_1 \hat{E}^*(\omega_2) \right\} \hat{E}_0(\omega_3) \pm j\gamma \hat{E}_0(\omega_1) = 0.$$

For solutions to exist (the determinant of the coefficient matrix must vanish), it must hold

$$\gamma^2 = \kappa_1\kappa_3|\hat{E}_0(\omega_2)|^2 \tag{4.103}$$

or

$$\gamma = \left\{\frac{2\kappa_1\kappa_3 I(\omega_2)}{n_2 c_0 \varepsilon_0}\right\}^{1/2}. \tag{4.104}$$

The fundamental solutions are cosine and sine functions, and together with the boundary conditions it follows

$$\hat{E}(\omega_3) = \hat{A}\sin\gamma z \tag{4.105}$$

and

$$\hat{E}(\omega_1) = \hat{E}_0(\omega_1)\cos\gamma z. \tag{4.106}$$

Substitution into Eq.(4.101) yields

$$\hat{A} = -j\sqrt{\frac{\omega_3 n_1}{\omega_1 n_3}}\hat{E}_0(\omega_1)\, e^{j\phi(\omega_2)}, \tag{4.107}$$

where

$$e^{j\phi(\omega_2)} = \frac{\hat{E}_0(\omega_2)}{|\hat{E}_0(\omega_2)|}$$

contains the phase of the pump wave. The factor $-j$ in Eq. (4.107) again implies, that the driving polarization advances the generated electric field by $\pi/2$. For the intensities we then obtain

$$I(\omega_3) = \frac{\omega_3}{\omega_1} I_0(\omega_1) \sin^2 \gamma z \tag{4.108}$$

$$I(\omega_1) = I_0(\omega_1) \cos^2 \gamma z \tag{4.109}$$

We realize that for $\gamma z > \pi/2$ again backconversion into ω_1 occurs. As in the best case each photon at frequency ω_1 is converted into a photon at frequency ω_3, it follows for the maximum enhancement in intensity

$$\frac{I_{\max}(\omega_3)}{I_0(\omega_1)} = \frac{\omega_3}{\omega_1}, \tag{4.110}$$

that again corresponds to the Manley-Rowe relation.

4.8 Difference-frequency generation (DFG)

Next we look at the generation of the difference frequency $\omega_1 = \omega_3 - \omega_2$, $\Delta k = k(\omega_1) + k(\omega_2) - k(\omega_3)$. Again in the low-conversion limit with $\hat{E}(\omega_3)$ and $\hat{E}(\omega_2)$ assumed to be constant, we obtain

$$I(\omega_1, \ell) = \frac{2\kappa_1^2 n_1}{n_2 n_3 c_0 \epsilon_0} \cdot \ell^2 I(\omega_3) I(\omega_2) \left\{ \frac{\sin \Delta k \ell/2}{\Delta k \ell/2} \right\}^2 . \tag{4.111}$$

This can, e.g., be used to generate infrared light from two visible light fields for spectroscopic applications. Another case is, if a strong pump wave at ω_3 converts light from ω_2 to ω_1. For a strong pump wave, $\hat{E}(\omega_3)$ is constant,

*E** in contrast to SFG

$$\frac{\partial \hat{E}(\omega_1)}{\partial z} = -j\kappa_1 \hat{E}(\omega_3) \hat{E}^*(\omega_2) e^{j\Delta k z} \tag{4.112}$$

$$\frac{\partial \hat{E}(\omega_2)}{\partial z} = -j\kappa_2 \hat{E}(\omega_3) \hat{E}^*(\omega_1) e^{j\Delta k z}. \tag{4.113}$$

The boundary conditions are now $E(\omega_2, z = 0) = E_0(\omega_2)$ and $E(\omega_1, z = 0) = 0$. With the ansatz $\hat{E}(\omega_{1,2}, z) = \hat{E}_0(\omega_{1,2})\, \mathrm{e}^{\pm\gamma z}$, we obtain when phase-matched

$$\pm\gamma \hat{E}_0(\omega_1) + j\left\{\kappa_1 \hat{E}(\omega_3)\right\} \hat{E}_0^*(\omega_2) = 0$$

$$\left\{-j\kappa_2 \hat{E}^*(\omega_3)\right\} \hat{E}_0(\omega_1) \pm \gamma \hat{E}_0^*(\omega_2) = 0,$$

which enforces

$$\gamma^2 = \kappa_1 \kappa_2 |\hat{E}(\omega_3)|^2. \tag{4.114}$$

In comparison to sum-frequency generation, the fundamental solutions are now hyperbolic functions

$$\hat{E}(\omega_1) = \hat{A} \sinh \gamma z$$

and

$$\hat{E}(\omega_2) = \hat{E}_0(\omega_2) \cosh \gamma z + \hat{B} \sinh \gamma z$$

After back substitution, it again follows $\hat{B} = 0$ and

$$\hat{A} = -j\sqrt{\frac{\omega_1 n_2}{\omega_2 n_1}} e^{j\phi(\omega_3)} \hat{E}_0^*(\omega_2).$$

Thus it follows

$$I(\omega_1) = (\omega_1/\omega_2) I_0(\omega_2) \sinh^2 \gamma z,$$

$$I(\omega_2) = I_0(\omega_2) \cosh^2 \gamma z.$$

This is indeed a different behavior compared to sum-frequency generation. Both waves grow! At first glimpse, it seems that energy is not conserved. Of course, this can not be the case. The required energy actually comes from the pump wave, however, this can not be observed because of the assumption $\hat{E}(\omega_3) = \text{const}$. For high conversion, we obtain for the intensity ratio of the low-frequency waves

$$\frac{I(\omega_1)}{I(\omega_2)} = \frac{\omega_1}{\omega_2}.$$

4.9 Optical parametric amplification (OPA)

Already for difference-frequency generation it became obvious, that in this frequency mixing process, gain can be achieved. We inspect this process now more generally taking into account losses and without the assumption of perfect phase matching. This will allow us to derive the underlying equations governing the amplification process, its gain bandwidth and associated amplifier noise. We keep the assumption of a constant pump wave at ω_3. Then it holds

symmetric in 1 and 2

$$\frac{\partial \hat{E}(\omega_1)}{\partial z} + \alpha_1 \hat{E}(\omega_1) = -j\kappa_1 \hat{E}\left(\omega_3\right) \hat{E}^*\left(\omega_2\right) e^{j\Delta k z} \tag{4.115}$$

$$\frac{\partial \hat{E}(\omega_2)}{\partial z} + \alpha_2 \hat{E}(\omega_2) = -j\kappa_2 \hat{E}\left(\omega_3\right) \hat{E}^*\left(\omega_1\right) e^{j\Delta k z}. \tag{4.116}$$

We again look for an exponential solution $\hat{E}\left(\omega_1\right) \sim \hat{E}_0\left(\omega_1\right) e^{\gamma' z + j\Delta k z/2}$, and $\hat{E}\left(\omega_2\right) \sim \hat{E}_0\left(\omega_2\right) e^{\gamma' z + j\Delta k z/2}$, with suitable initial values denoted by the index 0. It follows

$$\left\{\gamma' + \alpha_1 + \frac{j\Delta k}{2}\right\} \hat{E}_0\left(\omega_1\right) + \left\{j\kappa_1 \hat{E}\left(\omega_3\right)\right\} \hat{E}_0^*\left(\omega_2\right) = 0 \tag{4.117}$$

$$-\left\{j\kappa_2 \hat{E}^*\left(\omega_3\right)\right\} \hat{E}_0\left(\omega_1\right) + \left\{\gamma' + \alpha_2 - \frac{j\Delta k}{2}\right\} \hat{E}_0^*\left(\omega_2\right) = 0. \tag{4.118}$$

From the determinant condition, it follows

$$\gamma'^2 + \gamma'\{\alpha_1 + \alpha_2\} + \alpha_1\alpha_2 + (\Delta k/2)^2 + (\alpha_2 - \alpha_1)\frac{j\Delta k}{2} - \kappa_1\kappa_2\left|\hat{E}(\omega_3)\right|^2 = 0$$

$$\Rightarrow \gamma' = -\frac{(\alpha_1 + \alpha_2)}{2} \pm \left\{\left[\frac{\alpha_1 - \alpha_2}{2} + \frac{j\Delta k}{2}\right]^2 + \kappa_1\kappa_2\left|\hat{E}(\omega_3)\right|^2\right\}^{1/2} \quad (4.119)$$

For the case of phase matching and no losses, we find

$$\gamma^2 = \kappa_1\kappa_2\left|\hat{E}(\omega_3)\right|^2 \quad (4.120)$$

For the case of equal losses $\alpha_1 = \alpha_2 = \alpha$, we obtain

$$\gamma' = -\alpha \pm \left\{\gamma^2 - (\Delta k/2)^2\right\}^{1/2} = -\alpha \pm g, \quad (4.121)$$

where

$$g = \left\{\gamma^2 - (\Delta k/2)^2\right\}^{1/2}. \quad (4.122)$$

The solutions of the coupled equations (4.115)-(4.116) are of the form

$$\hat{E}(\omega_1, \ell) = e^{-\alpha\ell + j(\Delta k/2)\ell}\left\{\hat{E}_0(\omega_1)\cosh g\ell + B\sinh g\ell\right\} \quad (4.123)$$

$$\hat{E}(\omega_2, \ell) = e^{-\alpha\ell - j(\Delta k/2)\ell}\left\{\hat{E}_0(\omega_2)\cosh g\ell + D\sinh g\ell\right\} \quad (4.124)$$

Insertion into the coupled equations (4.115)-(4.116) yields

$$B = -j\frac{\Delta k}{2g}\hat{E}_0(\omega_1) - j\frac{\kappa_1}{g}\hat{E}(\omega_3)\hat{E}_0^*(\omega_2) \tag{4.125}$$

$$D = -j\frac{\Delta k}{2g}\hat{E}_0(\omega_2) - j\frac{\kappa_2}{g}\hat{E}(\omega_3)\hat{E}_0^*(\omega_1). \tag{4.126}$$

Note the symmetry between both solutions. Anyway, the two waves are generally called signal and idler wave. If the optical parametric amplifier (OPA) is seeded by one wave only and if $\omega_1 \neq \omega_2$, then this wave is amplified independent of the phase

$\sinh^2 = \cosh^2 - 1$

$$\left|\frac{\hat{E}(\omega_1,\ell)e^{\alpha\ell}}{\hat{E}_0(\omega_1)}\right|^2 = \left\{\cosh^2 g\ell + (\Delta k/2g)^2 \sinh^2 g\ell\right\} = \frac{1}{g^2}\left\{\gamma^2\cosh^2 g\ell - (\Delta k/2)^2\right\} \tag{4.127}$$

We define the parametric gain as

$$G_1(\ell) = \left|\frac{\hat{E}(\omega_1,\ell)e^{\alpha\ell}}{E_0(\omega_1)}\right|^2 - 1 \Rightarrow G_1(\ell) = (\gamma\ell)^2\frac{\sinh^2 gl}{(g\ell)^2} \tag{4.128}$$

Note that for small gain, i.e., $\gamma < \Delta k$, the gain has the form

g becomes imaginary: $\sinh \to \sin$

$$G_1(\ell) = (\gamma\ell)^2\frac{\sin^2\left\{\left[(\Delta k/2)^2 - \gamma^2\right]^{1/2}\ell\right\}}{\ell^2\left\{(\Delta k/2)^2 - \gamma^2\right\}}, \tag{4.129}$$

that can me further simplified to

$$G_1\ (\ell) = (\gamma\ell)^2 \frac{\sin^2(\Delta k\ell/2)}{(\Delta k\ell/2)^2}$$

for $\gamma \ll \Delta k/2$. In the opposite limit for large gain $\gamma \gg \Delta k/2$

$$G_1\ (\ell) = (\gamma\ell)^2 \frac{\sinh^2 g\ell}{(g\ell)^2} \Rightarrow \frac{1}{4}e^{2g\ell}. \tag{4.130}$$

We define the bandwidth of the OPA via

$$\left\{(\Delta k/2)^2 - \gamma^2\right\}^{1/2} \ell = \pi, \tag{4.131}$$

that for small gain implies $\Delta k = 2\pi/\ell$, as we already found for frequency doubling. In general, we obtain from Eq. (4.131)

$$\frac{2\pi\Delta f}{c} = \Delta k = \left\{1 + \left(\frac{\gamma\ell}{\pi}\right)^2\right\}^{1/2} (2\pi/\ell)\,. \tag{4.132}$$

This means that the bandwidth increases with the gain. For the ratio between the bandwidths at high and low gain, we obtain

$$\frac{\Delta f_{\text{ High Gain}}}{\Delta f_{\text{ Low Gain}}} = \left\{1 + (\gamma\ell/\pi)^2\right\}^{1/2}. \tag{4.133}$$

Fig. 4.15 shows the gain as function of $\Delta k\ell$ for various values of $\gamma\ell$.

gain bandwidth increases with stronger pumping

noncollinear type-I OPAs feature ultrabroad bandwidth for few-optical-cycle pulse generation (→ later lecture)

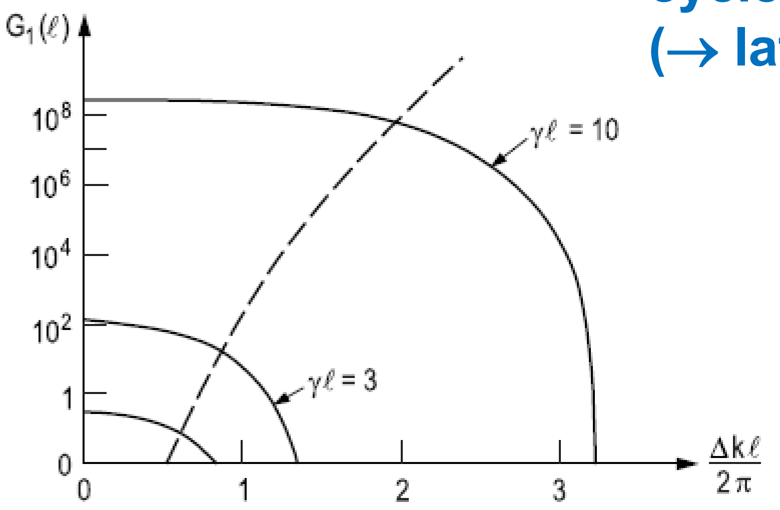

Figure 4.15: Gain of an optical parametric amplifier (OPA) as function of the wave number difference and gain.

4.10 Optical parametric oscillation (OPO)

In a single pass through a parametric amplifier medium, which is described by Eqs. (4.123)-(4.126), the signal and idler waves grow when phase-matched ($\Delta k = 0$) according to

$$\hat{E}(\omega_1, \ell)\, e^{\alpha_1 \ell} = \hat{E}_0(\omega_1) \cosh \gamma\ell - j\frac{\kappa_1}{\gamma} \hat{E}(\omega_3)\, \hat{E}_0^*(\omega_2) \sinh \gamma\ell \qquad (4.134)$$

$$\hat{E}(\omega_2, \ell)\, e^{\alpha_2 \ell} = \hat{E}_0(\omega_2) \cosh \gamma\ell - j\frac{\kappa_2}{\gamma} \hat{E}(\omega_3)\, \hat{E}_0^*(\omega_1) \sinh \gamma\ell. \qquad (4.135)$$

Most often parametric amplifiers only permit gain for passage in a single direction. In the other direction, only damping occurs

$$\hat{E}'(\omega_1, \ell) = \hat{E}_0(\omega_1)\, e^{-\alpha_1 \ell}$$

$$\hat{E}'(\omega_2, \ell) = \hat{E}_0(\omega_2)\, e^{-\alpha_2 \ell}$$

If feedback of the parametric amplifier is realized by means of a Fabry-Pérot resonator and if the field is larger after a round trip than at the beginning, so the amplifier is turned into a self-starting oscillator. The threshold condition is that the losses must equal the gain

$$\hat{E}_0'(\omega_1) = \hat{E}_0(\omega_1)$$

$$\hat{E}_0'(\omega_2) = \hat{E}_0(\omega_2)$$

or inserted into Eqs. (4.134)-(4.135) it follows with $e^{-2\alpha\ell} \sim 1 - 2\alpha\ell$

$$\frac{\hat{E}_0(\omega_1)}{1-2\alpha_1\ell} = \hat{E}_0(\omega_1)\cosh\gamma\ell - j\frac{\kappa_1}{\gamma}\hat{E}(\omega_3)\,\hat{E}_0^*(\omega_2)\sinh\gamma\ell$$

$$\frac{\hat{E}_0^*(\omega_2)}{1-2\alpha_2\ell} = \hat{E}_0^*(\omega_2)\cosh\gamma\ell + j\frac{\kappa_2}{\gamma}\hat{E}^*(\omega_3)\,\hat{E}_0(\omega_1)\sinh\gamma\ell.$$

Again the solution of this equation system is only non-zero, if the determinant of the coefficient matrix vanishes, i.e.,

$$\left[\cosh\gamma\ell - \frac{1}{1-2\alpha_1\ell}\right]\left[\cosh\gamma\ell - \frac{1}{1-2\alpha_2\ell}\right] = \frac{\kappa_1\kappa_2}{\gamma^2}\left|\hat{E}(\omega_3)\right|^2\sinh^2\gamma\ell = \sinh^2\gamma\ell$$

so that

$$1-\cosh\gamma\ell\left(\frac{1}{1-2\alpha_1\ell}+\frac{1}{1-2\alpha_2\ell}\right)+\left(\frac{1}{1-2\alpha_2\ell}\right)\left(\frac{1}{1-2\alpha_1\ell}\right)=0 \quad (4.136)$$

or

$$\cosh\gamma\ell = 1+\frac{2\alpha_1\alpha_2\ell^2}{1-\alpha_1\ell-\alpha_2\ell}. \quad (4.137)$$

For $\alpha_1 \approx \alpha_2 \approx \alpha$ and the case of small losses or small gain $\alpha\ell$, $\gamma\ell \ll 1$, it follows

$$(\gamma\ell)^2 \approx 4\alpha\ell. \qquad \mathbf{\cosh x = 1 + \frac{x^2}{2!}} \quad (4.138)$$

One distinguishes between doubly resonant parametric oscillators (DROs) and singly resonant ones (SRO). In the first case, both signal and idler waves are resonant, in the second case only the signal wave. The threshold for SROs is many times higher than for DRO. Nevertheless, most OPOs are singly resonant, because it is much more difficult to operate a DRO.