NLO Lecture 7: Phase Matching

4 Frequency doubling

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4.4.3 Frequency doubling of pulses

4.4.4 Effective nonlinear coefficient d_{eff}

4.4.5 Quasi-phase matching (QPM)

4.5 Optical rectification

4.6 Manley-Rowe relations

4.4.3 Frequency doubling of pulses

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

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

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

With Eqs. (4.63) and (4.64)

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

where

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

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

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

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

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

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

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

with

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

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

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

where

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

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

and

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

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

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

where

$$\begin{aligned} g(t) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} G\left(\omega\right) e^{j\omega t} d\omega \qquad\qquad f(t)=E_1(t)^2 \\ &= \left\{ \begin{array}{ll} \underbrace{e^{j2\omega_0 t}}_{\text{phase}} \frac{\omega_0 d_{eff}}{4nc_0} \frac{1}{\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)}, & 0 < t < \left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell \\ 0, & \text{elsewhere} \end{array} \right\} \end{aligned} \tag{4.76}$$

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

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

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

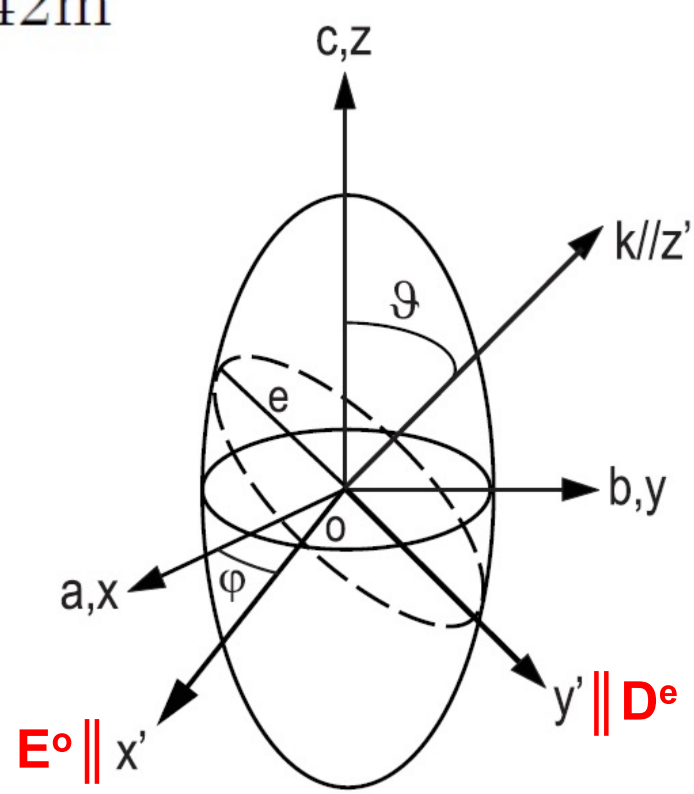
$$\ell \left\{ \left.\frac{1}{v_g}\right|_{2\omega} - \left.\frac{1}{v_g}\right|_{\omega} \right\}.$$

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 $\bar{4}2m$

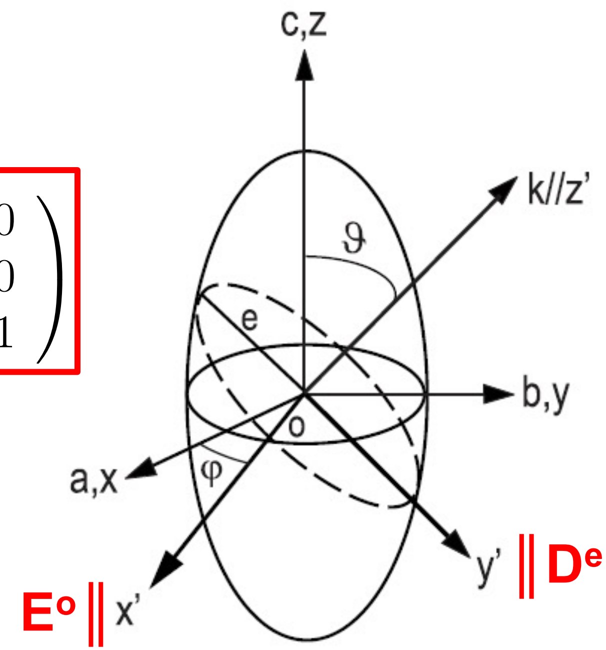
KDP



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 $\mathbf{T}$



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

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

The inverse is

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

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:

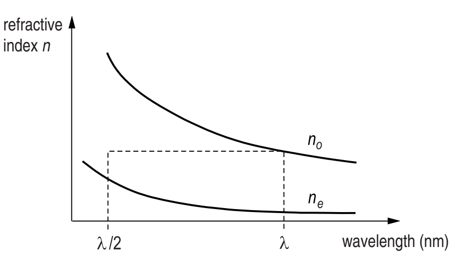


Figure 4.7: Type-I critical phase matching.

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

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

type-I PM

type-II PM

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

2cosφsinφ

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

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 $P^{(2)}_{y'}(2\omega)$ 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

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

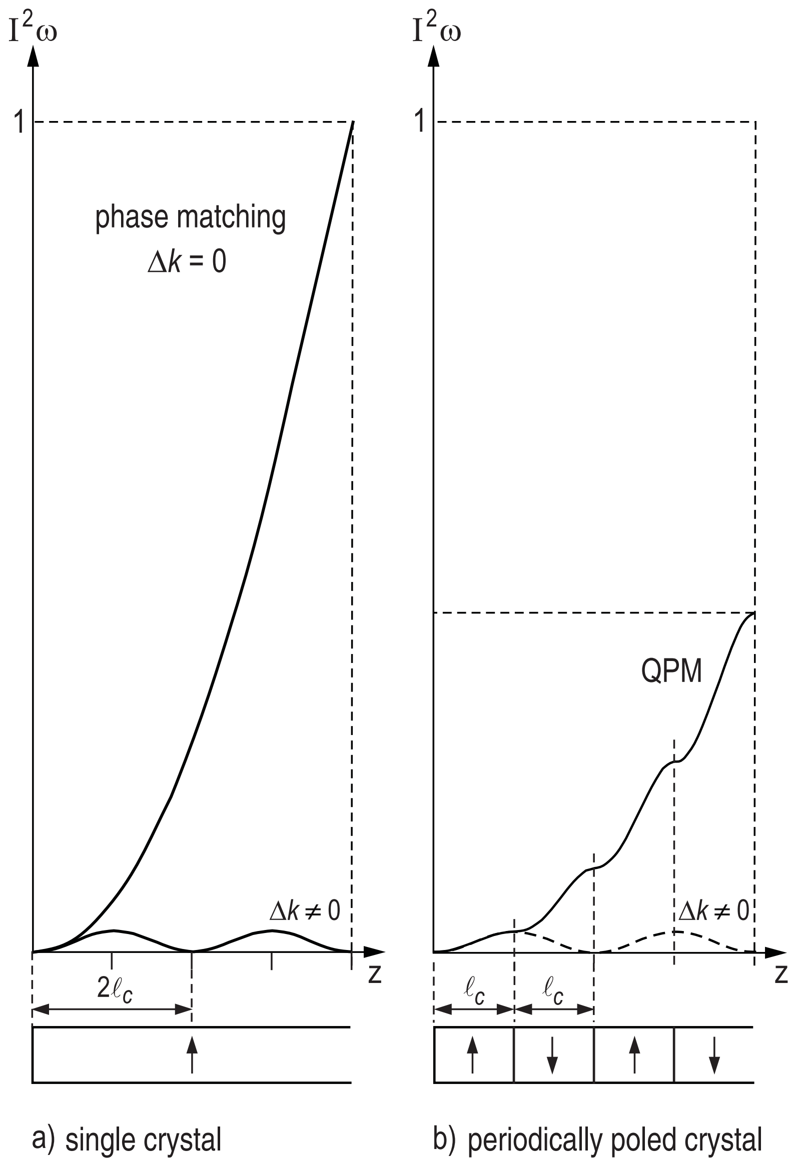
Because of Kleinman symmetry $d_{36} = d_{14}$. The effective nonlinear coefficients for type-I phase matching for the different point groups are given in Table 4.3.

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.

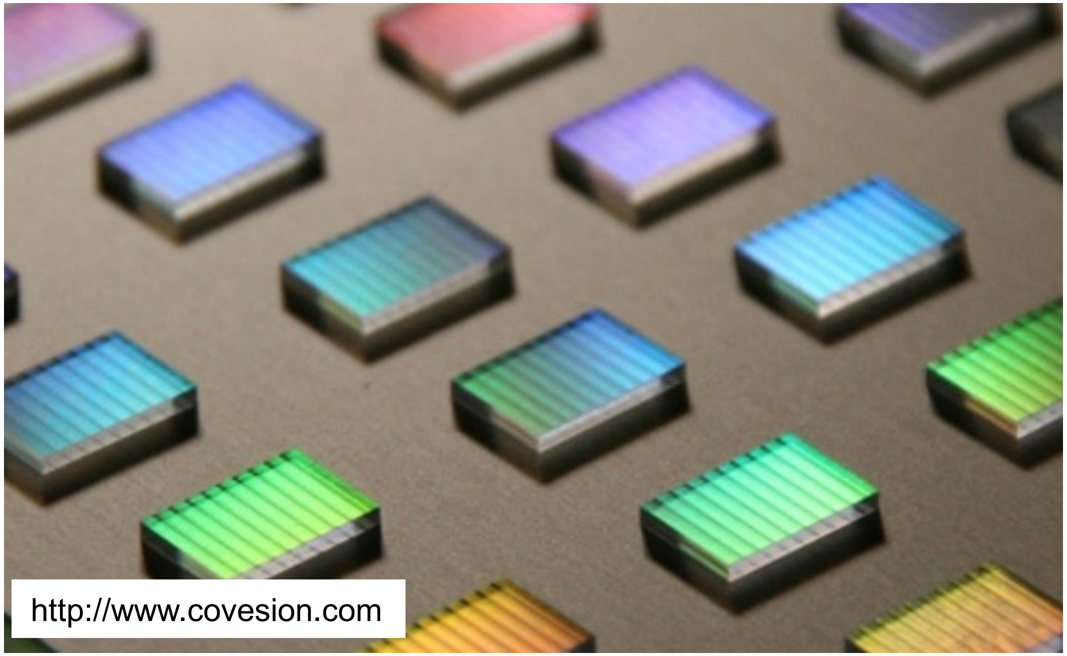


Figure 4.13: Growth of second harmonic as a function of distance z in a crystal for different cases: a) homogeneous crystal and b) periodically poled crystal.

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.

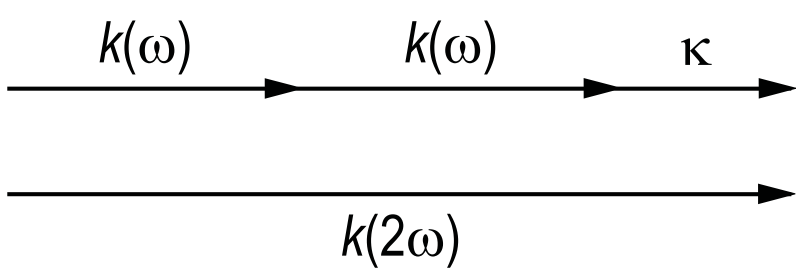
$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c} d_{eff}(z) \hat{E}(\omega) \hat{E}(\omega) e^{j(k(2\omega)-2k(\omega))z}. \tag{4.86}$$

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

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

$$
\boxed{
\begin{aligned}
\frac{d\hat{E}(\omega_1)}{dz} &= -j\kappa_1 \hat{E}(\omega_3)\,\hat{E}^*(\omega_2)\,e^{-j\Delta kz}, && (4.92)\\
\frac{d\hat{E}(\omega_2)}{dz} &= -j\kappa_2 \hat{E}(\omega_3)\,\hat{E}^*(\omega_1)\,e^{-j\Delta kz}, && (4.93)\\
\frac{d\hat{E}(\omega_3)}{dz} &= -j\kappa_3 \hat{E}(\omega_1)\,\hat{E}(\omega_2)\,e^{+j\Delta kz}, && (4.94)
\end{aligned}
}
$$

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 $E = n \cdot h\nu$.