3 Wave propagation in nonlinear media

Starting point for the nonlinear wave equation are Maxwell's equations and the nonlinear polarisation $P = P_{lin} + P_{NL}$. To derive the nonlinear wave equation we investigate the simultaneous propagation of three electric fields E_1, E_2, E_3 which finally results in a system of coupled differential equations describing effects like second harmonic generation and parametric processes.

The Maxwell equations for non-magnetic media are

$$\vec{\nabla}\times\vec{H} = \vec{j} + \frac{\partial}{\partial t}\vec{D}\,, \qquad \vec{\nabla}\times\vec{E} = -\mu_0\frac{\partial}{\partial t}\vec{H}$$

with current density $\vec{j} = \sigma\vec{E}$ and conductivity σ. For the dielectric displacement we have

$$\vec{D} = \varepsilon_0\,\vec{E} + \vec{P} = \varepsilon_0\,\vec{E} + \varepsilon_0\,\chi\,\vec{E} + \vec{P}_{NL}$$

with $\varepsilon = 1+\chi$ and component $P_{NL,\,i}$ of the nonlinear polarisation

$$P_{NL,\,i} = \sum_{j,k} d_{ijk}\;E_j\,E_k\;\;.$$

Insertion into Maxwell's equation results in

$$\vec{\nabla}\times\vec{H} = \sigma\,\vec{E} + \varepsilon\,\varepsilon_0\,\frac{\partial}{\partial t}\,\vec{E} + \frac{\partial}{\partial t}P_{NL}$$

Taking the curl operator *rot* on both sides of the second Maxwell equation leads to the nonlinear wave equation

$$\nabla^2\,\vec{E} = \mu_0\,\sigma\,\frac{\partial}{\partial t}\,\vec{E} + \mu_0\,\varepsilon\,\varepsilon_0\,\frac{\partial^2}{\partial t^2}\,\vec{E} + \mu_0\,\frac{\partial^2}{\partial t^2}\,\vec{P}_{NL}$$

We assume an isotropic medium with $\vec{E}\,||\,\vec{P}_{NL}$ and go over to a scalar notation.

Let us consider the propagation of three plane waves E_1, E_2 and E_3 with frequencies ω_1, ω_2 and ω_3 along the z-direction of the medium:

$$E_1^{(\omega_1)}(z,t) = \frac{1}{2}\left(E_{01}^{(\omega_1)}(z)\,e^{i\,(\omega_1 t - k_1 z)} + c.\,c.\right)$$

$$E_2^{(\omega_2)}(z,t) = \frac{1}{2}\left(E_{02}^{(\omega_2)}(z)\,e^{i(\omega_2 t - k_2 z)} + c.\,c.\right)$$

$$E_3^{(\omega_3)}(z,t) = \frac{1}{2}\left(E_{03}^{(\omega_3)}(z)\,e^{i\,(\omega_3 t - k_3 z)} + c.\,c.\right)$$

The total instantaneous light field is

$$E = E_1^{(\omega_1)} + E_2^{(\omega_2)} + E_3^{(\omega_3)}$$

and has to fulfil the nonlinear wave equation. Now we have a look at the different frequencies that may contribute to the nonlinear polarization P_{NL} and which are of the form $d_{ijk}\, E_j\, E_k$, for example

$$P_{NL}^{(\omega_1+\omega_2)} = \mathrm{Re}\left(d^{(\omega_1+\omega_2)}\, E_{01}\, E_{02}\, e^{i\left[(\omega_1+\omega_2)t-(k_1+k_2)z\right]} \right)$$

or

$$P_{NL}^{(\omega_3-\omega_2)} = \mathrm{Re}\left(d^{(\omega_3-\omega_2)}\, E_{03}\, E_{02}^{*}\, e^{i\left[(\omega_3-\omega_2)t-(k_3-k_2)z\right]} \right)$$

In general all different sums and differences of the frequencies $\omega_{1,\,2,\,3}$ may contribute to P_{NL}. However, only those terms that are synchronous with either ω_1, ω_2 or ω_3 can drive an oscillation at these frequencies, while all others being nonsynchronous.

An example of the last statement is the case

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

where the term $P_{NL}^{(\omega_1+\omega_2)}$ oscillates at frequency ω_3 and thus forms a source of an oscillating wave with frequency ω_3. In physical terms, we have a power flow from the fields at ω_1 and ω_2 into that of ω_3, or vice versa.

In what follows we assume the case $\omega_3 = \omega_1 + \omega_2$ to be fulfilled. The nonlinear wave equation has to be valid for all times t, i.e. we can treat the terms oscillating at different frequencies independently. For this term oscillating at frequency ω_1 we get:

$$\nabla^2 E_1^{(\omega_1)} = \mu_0\,\sigma_1\,\frac{\partial}{\partial t}E_1^{(\omega_1)} + \mu_0\,\varepsilon_1\,\varepsilon_0\,\frac{\partial}{\partial t^2}E_1^{(\omega_1)}$$

$$+ \mu_0\, d\,\frac{\partial^2}{\partial t^2}\left[\frac{1}{2}E_{03}^{(\omega_3)}\,E_{02}^{(\omega_2)^*}\,e^{i\left[(\omega_3-\omega_2)t-(k_3-k_2)z\right]} + c.\,c.\right]$$

Because we have assumed a propagation along the z-direction only, the Laplace operator ∇^2 reduces to the second derivative along z

$$\nabla^2 E_1^{(\omega_1)} = \frac{1}{2}\frac{\partial^2}{\partial z^2}\left[E_{o1}^{(\omega_1)}\,e^{i(\omega_1 t-k_1 z)} + c.\,c.\right]$$

$$= \frac{1}{2}\left[\frac{\partial^2}{\partial z^2}E_{01}^{(\omega_1)} - 2ik_1\frac{\partial}{\partial z}E_{01}^{(\omega_1)} - k_1^2\,E_{01}^{(\omega_1)}\right]e^{i(\omega_1 t-k_1 z)} + c.\,c.$$

Next we assume slowly varying amplitudes of the propagating waves $E_{0i}^{(\omega_i)}$, i.e. we use the approximation

$$\left| \frac{\partial^2}{\partial z^2} E_{01}^{(\omega_1)} \right| << \left| k_1 \frac{\partial}{\partial z} E_{01}^{(\omega_1)} \right|$$

This leads to

$$\nabla^2 E_1^{(\omega_1)} = -\frac{1}{2}\left[k_1^{\,2} E_{01}^{(\omega_1)} + 2ik_1 \frac{\partial}{\partial z} E_{01}^{(\omega_1)} \right] e^{i(\omega_1 t - k_1 z)} + c.c.$$

All components oscillate with $e^{i\omega_1 t}$, and with $\partial/\partial t \to i\omega_1$ it follows

$$-\frac{1}{2}\left[k_1^{\,2} E_{01}^{(\omega_1)} + 2ik_1 \frac{\partial}{\partial z} E_{01}^{(\omega_1)} \right] e^{-ik_1 z} + c.c.$$

$$= (i\omega_1 \mu_0 \sigma_1 - \omega_1^2 \mu_0 \varepsilon_1 \varepsilon_0) \left[\frac{1}{2} E_{01}^{(\omega_1)} e^{-ik_1 z} + c.c. \right]$$

$$-\omega_1^2 \mu_0 d \left[\frac{1}{2} E_{03}^{(\omega_3)} E_{02}^{(\omega_2)^*} e^{-i(k_3 - k_2)z} + c.c. \right]$$

For the wave number we find

$$k_1 = \frac{2\pi}{\lambda} n = \omega_1 \sqrt{\mu_0 \varepsilon_1 \varepsilon_0}$$

or $k_1^2 = \omega_1^2 \mu_0 \varepsilon_1 \varepsilon_0$. Multiplication of the not complex conjugated part of the equation before last with $i/k_1\, e^{ik_1 z}$ results in

$$-\frac{1}{2}\left[\omega_1^2 \mu_0 \varepsilon_1 \varepsilon_0 \frac{i}{k_1} E_{01}^{(\omega_1)} + 2ik_1 \frac{i}{k_1} \frac{\partial E_{01}^{(\omega_1)}}{\partial z} \right]$$

$$= \left(i\omega_1 \mu_0 \sigma_1 \frac{i}{k_1} - \omega_1^2 \mu_0 \varepsilon_1 \varepsilon_0 \frac{i}{k_1} \right) \frac{1}{2} E_{01}^{(\omega_1)}$$

$$-\omega_1^2 \mu_0 \frac{i}{k_1} d \; \frac{1}{2} E_{03}^{(\omega_3)} E_{02}^{(\omega_2)^*} e^{-i(-k_1 + k_3 - k_2)z}$$

As a result we now have a system of three coupled differential equations that describe the evolution of the complex field amplitudes $E_{0\,i}$ along the propagation direction z:

$$\frac{\partial E_{01}^{(\omega_2)}}{\partial z} = -\frac{\sigma_1}{2}\sqrt{\frac{\mu_0}{\varepsilon_1\varepsilon_0}}\,E_{01}^{(\omega_1)} - \frac{i\omega_1}{2}\sqrt{\frac{\mu_0}{\varepsilon_1\varepsilon_0}}\,d\,E_{03}^{(\omega_3)}\,E_{02}^{(\omega_2)^*}\,e^{-i(k_3-k_2-k_1)z}$$

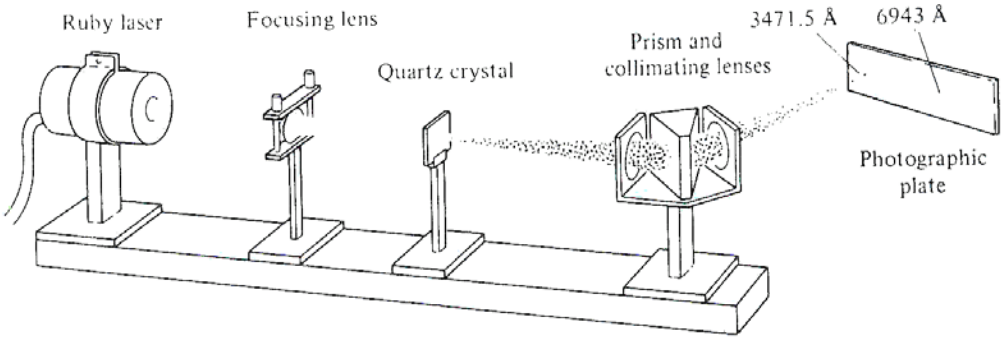
$$\frac{\partial E_{02}^{(\omega_2)}}{\partial z} = -\frac{\sigma_2}{2}\sqrt{\frac{\mu_0}{\varepsilon_2\varepsilon_0}}\,E_{02}^{(\omega_2)} - \frac{i\omega_2}{2}\sqrt{\frac{\mu_0}{\varepsilon_2\varepsilon_0}}\,d\,E_{01}^{(\omega_1)^*}\,E_{03}^{(\omega_3)}\,e^{-i(k_3-k_1-k_2)z}$$

$$\frac{\partial E_{03}^{(\omega_3)}}{\partial z} = -\frac{\sigma_3}{2}\sqrt{\frac{\mu_0}{\varepsilon_3\varepsilon_0}}\,E_{03}^{(\omega_3)} - \frac{i\omega_3}{2}\sqrt{\frac{\mu_0}{\varepsilon_3\varepsilon_0}}\,d\,E_{01}^{(\omega_1)}\,E_{02}^{(\omega_2)}\,e^{-i(k_1+k_2-k_3)z}$$

The first term on the right hand side of each equation describes damping of the wave, while the second one accounts for the nonlinear coupling of the interacting waves („wave mixing“). This term also includes a phase term, which leads to an oscillation of the coupling strength along the propagation direction.

3.1 Frequency doubling

The first experiment in nonlinear optics in 1961 by Franken et al. (Phys. Rev. Lett. 7, 118 (1961)) consisted of generating the second of a Ruby laser 694 nm (red) → 347 nm (UV) into the ultra violet spectral region.



The conversion efficiency achieved in this first experiment, i.e. the ratio of power of UV light compared to the red pump light power, was about $P_{UV}/P_{rot} \approx 10^{-8}$. Today conversion efficiencies up to 30 % in single-pass operation and up to 80 % inside an optical resonators can be obtained.

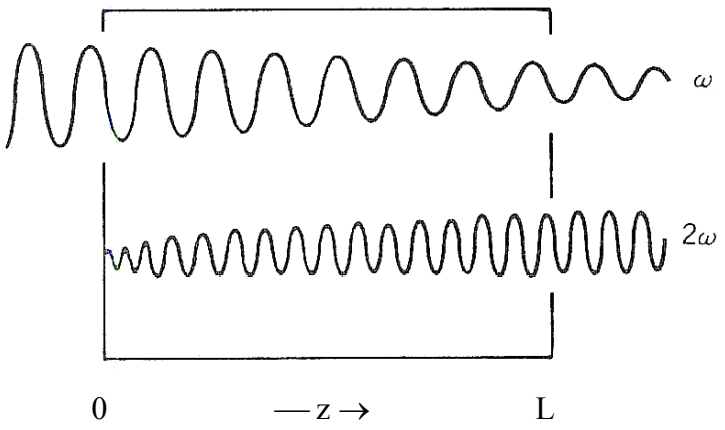
Second harmonic generation is a special case of the differential equation system derived before for $\omega_1 = \omega_2 = \omega$, $\omega_3 = 2\omega$ and $E_{01} = E_{02} = E^{(\omega)}$, $E_{03} = E^{(2\omega)}$. Furthermore we will neglect absorption in the following discussion, i.e. we assume $\sigma_1 = \sigma_2 = \sigma_3 = 0$.

This results in

$$\frac{d\,E^{(2\omega)}}{d\,z} = -i\,\omega\,\sqrt{\frac{\mu_0}{\varepsilon\,\varepsilon_0}}\,d\,\left[E^{(\omega)}\right]^2\,e^{i\,\Delta k\,z}$$

with $\Delta k \equiv k_3 - 2\,k_1 = k^{(2\omega)} - 2\,k^{(\omega)}$

We now focus on frequency doubling in a crystal sample of length L:



Let us first assume that conversion efficiency is low, so the relation $E^{(\omega)}(z) \approx E^{(\omega)}(0) = const.$ is fulfilled, which is the case of non-depleted pump approximation. Only wave $E^{(\omega)}$ is used as input, so the boundary condition is $E^{(2\omega)}(0) = 0$.

Integration of the two above differential equations along the propagation direction z from 0 to L gives us the solution for the amplitude $E^{(2\omega)}(L)$ at the rear face of the crystal

$$E^{(2\omega)}(L) = -\,i\,\omega\,\sqrt{\frac{\mu_0}{\varepsilon_{2\omega}\,\varepsilon_0}}\,d\,\left[E^{(\omega)}\right]^2\,\int_0^L e^{i\,\Delta k\,z}\,dz$$

$$= -\,i\,\omega\,\sqrt{\frac{\mu_0}{\varepsilon_{2\omega}\,\varepsilon_0}}\,d\,\left[E^{(\omega)}\right]^2\,\frac{e^{i\,\Delta k\,L} - 1}{i\,\Delta k}$$

The intensity of the frequency doubled wave is proportional to

$$I^{(2\omega)} \sim E^{(2\omega)}(L)\,E^{(2\omega)^*}(L) = -\omega^2\,\frac{\mu_0}{\varepsilon_0\,\varepsilon_{2\omega}}\,d^2\,|\,E^{(\omega)}\,|^4\,\frac{\left(e^{i\,\Delta k\,L} - 1\right)\left(e^{-i\,\Delta k\,L} - 1\right)}{(\Delta k)^2}$$

$$= \omega^2\,\frac{\mu_0}{\varepsilon_0}\,\frac{1}{n_{2\omega}^2}\,d^2\,|\,E^{(\omega)}\,|^4\,L^2\,\frac{\left(e^{i\,\Delta k\,L} - 1\right)\left(1 - e^{-i\,\Delta k\,L}\right)}{4\,(\Delta k\,L/2)^2}$$

$$= \frac{\mu_0}{\varepsilon_0}\,\frac{\omega^2\,d^2}{n_{2\omega}^2}\,\left|E^{(\omega)}\right|^4\,L^2\,\frac{\sin^2\left(\Delta k\,L/2\right)}{(\Delta k\,L/2)}$$

Intensity is defined as power per area

$$I_{2\omega} \equiv \frac{P_{2\omega}}{F} \quad \text{and} \quad I_{\omega} = \frac{P_{\omega}}{F}$$

with the effective beam area F. This results in

$$P_{\omega} = F\, I_{\omega} = F\, \frac{c\, n_{\omega}\, \varepsilon_0}{2}\, \left| E^{(\omega)} \right|^2$$

$$\left| E^{(\omega)} \right|^2 = \frac{2}{c\, n_{\omega}\, \varepsilon_0}\, \frac{P_{\omega}}{F}$$

The efficiency of second harmonic generation is given by the ratio

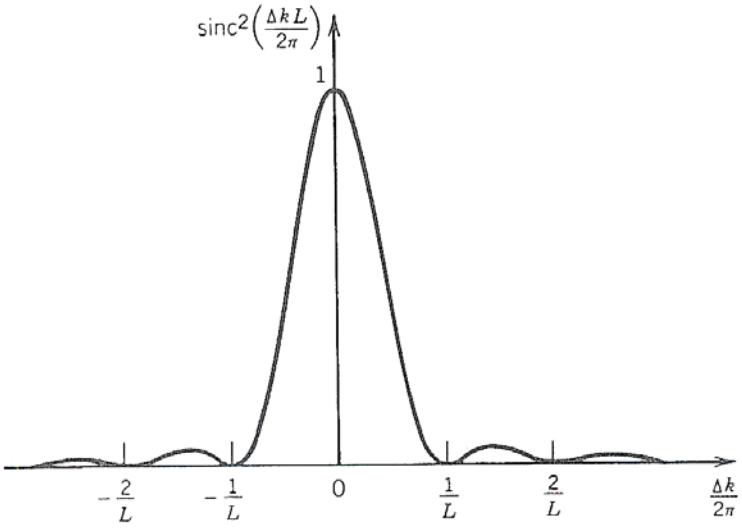
$$\eta \equiv \frac{P_{2\omega}}{P_{\omega}} = \frac{\left| E^{(2\omega)} \right|^2}{\left| E^{(\omega)} \right|^2} \ ,$$

$$\eta = \frac{\left| E^{(2\omega)} \right|^2}{\left| E^{(\omega)} \right|^2} = \frac{\mu_0}{\varepsilon_0}\, \frac{\omega^2\, d^2\, L^2}{n_{2\omega}^2}\, \left| E^{(\omega)} \right|^2\, \frac{\sin^2(\Delta k\, L/2)}{(\Delta k\, L/2)^2}$$

$$= \frac{2}{c\, n_{\omega}\, \varepsilon_0}\, \frac{\mu_0}{\varepsilon_0}\, \frac{\omega^2\, d^2\, L^2}{n_{2\omega}^2}\, \frac{\sin^2\left(\Delta k\, L/2\right)}{\left(\Delta k\, L/2\right)^2}\, \frac{P_{\omega}}{F}$$

$$= 2 \left(\frac{\mu_0}{\varepsilon_0} \right)^{3/2}\, \frac{\omega^2\, d^2\, L^2}{n_{2\omega}^2\, n_{\omega}}\, \frac{\sin^2\left(\Delta k\, L/2\right)}{\left(\Delta k\, L/2\right)^2}\, \frac{P_{\omega}}{F} \ .$$

An important result is the proportionality of η to the squares of both sample length L^2 (i.e. large interaction lengths) and d^2 (i.e. large nonlinear coefficients) and the intensity I_{ω} of the pump wave.



Now we want to discuss second harmonic generation for the case of pump wave depletion, i.e. we now have $E^{(\omega)}(L) < E^{(\omega)}(0)$. For simplification we use the notation

$$A_l = \sqrt{\frac{n_l}{\omega_l}}\, E_l \quad \text{with } l = 1,2,3$$

For the intensity of each wave $l = 1, 2, 3$ we get

$$I_l = \frac{P_l}{F} = \frac{c\, n_l\, \varepsilon_0}{2} \left|E_l\right|^2$$

$$= \frac{c\, \omega_l\, \varepsilon_0}{2} \left|A_l\right|^2$$

The three coupled differential equations now read

$$\frac{d\,A_1}{d\,z} = -\frac{\alpha_1}{2}\, A_1 - i\,\kappa\, A_2^*\, A_3\, e^{-i\,\Delta k\, z}$$

$$\frac{d\,A_2}{d\,z} = -\frac{\alpha_2}{2}\, A_2 - i\,\kappa\, A_1^*\, A_3\, e^{-i\,\Delta k\, z}$$

$$\frac{d\,A_3}{d\,z} = -\frac{\alpha_3}{2}\, A_3 - i\,\kappa\, A_1\, A_2\, e^{i\,\Delta k\, z}$$

with

$$\kappa = \frac{d}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}\,\frac{\omega_1\,\omega_2\,\omega_3}{n_1\,n_2\,n_3}}$$

$$\alpha_l = \sigma_l\,\sqrt{\mu_0 / \varepsilon_0\,\varepsilon_l} \quad , \quad l = 1, 2, 3$$

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

For the case of second harmonic generation with $A_1 = A_2$ for the two pump wave amplitudes and with $\omega_1 = \omega_2 = \omega$, we neglect again the influence of damping, $\alpha_l = 0$. As a result we find for the amplitude A_3 of the frequency-doubled wave at frequency $\omega_3 = 2\omega$

$$\frac{d\,A_1}{d\,z} = -\,i\,\kappa\, A_3\, A_1^*\, e^{-i\,\Delta k\, z}$$

$$\frac{d\,A_3}{d\,z} = -\,i\,\kappa\, A_1^2\, e^{i\,\Delta k\, z}$$

We further assume phase matching to be fulfilled, i.e. we take $\Delta k = 0$.

Without loss of generality we chose a phase reference of the differential equation system by assuming $A_1(0)$ to be a real quantity. As a direct consequence now also $A_1(z)$ is a real number and we get the following system

$$\frac{d A_1}{d z} = -\kappa \hat{A}_3 A_1 \ ,$$

$$\frac{d \hat{A}_3}{d z} = \kappa A_1^2 \ ,$$

with $\hat{A}_3 = +i A_3$ or $A_3 = -i \hat{A}_3$.

This can be interpreted in a way that the wave with amplitude A_3 is shifted by $\pi/2$ relative to that with amplitude A_1, i.e. the amplitudes A_1 and $\hat{A}_3$ are in phase.

For the nonlinear interaction with boundary condition $A_3(0) = 0$ we have conservation of energy in the form

$$A_1^2(z) + \hat{A}_3^2(z) = const.$$

$$= A_1^2(0)$$

Thus we may write

$$\frac{d \hat{A}_3}{d z} = \kappa A_1^2 = \kappa \left[A_1^2(0) - \hat{A}_3^2(z) \right]$$

or

$$\frac{d \hat{A}_3}{A_1^2(0) - \hat{A}_3^2(z)} = \kappa \, dz$$

Integration and using the relation

$$\int \frac{dx}{a^2 - x^2} = \frac{1}{a} \operatorname{atanh}\left(\frac{x}{a}\right)$$

gives the amplitude of the frequency-doubled wave

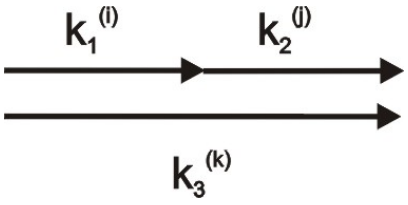
$$\hat{A}_3(z) = A_1(0) \tanh\left(\kappa A_1(0) z\right) .$$

The efficiency of second harmonic generation is now

$$\eta = \frac{P^{(2\omega)}}{P^{(\omega)}} = \frac{\left|A_3(z)\right|^2}{\left|A_1(0)\right|^2} = \tanh^2\left(\kappa A_1(0) z\right) \ .$$

3.2 Phase matching

Prerequisite for efficient second harmonic generation is phase matching of the interacting waves $\Delta k = 0$, or in other words equal phase velocity of pump and frequency-doubled beam. Using the deBroglie relation $p = \hbar\, k$ this is identical with conservation of momentum.



i, j, k : polarization direction

with $\Delta k = k_3^{(k)} - k_1^{(i)} - k_1^{(j)}$, $\omega_3 = 2\omega$, $\omega_1 = \omega_2 = \omega$ and

$$k_l = \frac{2\pi n_l}{\lambda_l} = \frac{\omega_l}{c}\, n_l \quad , \qquad l = 1, 2, 3$$

Phase matching $\Delta k = 0$ is fulfilled if

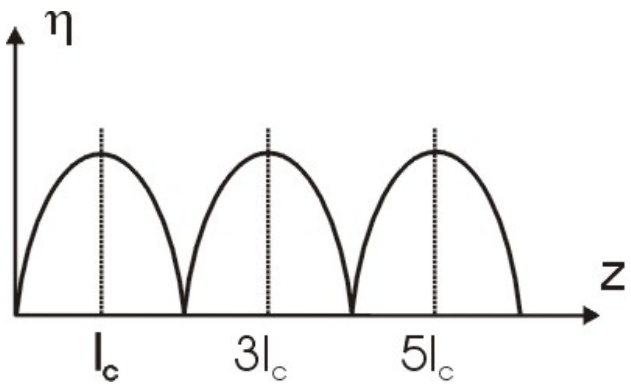
$$\frac{1}{c}\left(2\omega n_k^{(2\omega)} - \omega n_i^{(\omega)} - \omega n_j^{(\omega)}\right) = 0$$

or $$2n_k^{(2\omega)} = n_i^{(\omega)} + n_j^{(\omega)}$$

In the most simple case $n_i^{(\omega)} = n_j^{(\omega)}$ of a linearly polarized pump wave with $i = j$, this requires

$$n^{(2\omega)} = n^{(\omega)} \; .$$

For the case that phase matching is not exactly fulfilled, $\Delta k \neq 0$, the frequency-doubled wave propagates either faster or slower then the pump wave. This leads to a phase mismatch between the two waves that grows during propagation along z. As a result the power conversion oscillates as well among pump wave and second harmonic and follows a sinc function $\sin^2(\Delta k\, x/2)/(\Delta k\, x/2)^2$.



The spatial coherence length l_c is given by

$$l_c = \frac{\pi}{\Delta k} = \frac{\pi}{k^{(2\omega)} - 2k^{(\omega)}} = \frac{\lambda}{4(n^{(2\omega)} - n^{(\omega)})}$$

At $z = l_c$ both waves are π out of phase, i.e. power starts to be transferred back to the pump wave. For high conversion efficiency exact phase matching is therefore inevitable.

Numerical example: wavelength $\lambda = 1\ \mu\text{m}$, dispersion $n^{(2\omega)} - n^{(\omega)} \simeq 10^{-2}$:

$$l_c = \frac{10^{-6}\,\text{m}}{4\times 10^{-2}} = 25\ \mu\text{m}$$

The crystal length is assumed to be $L = 1\ \text{cm}$. This results in an increase of efficiency as a result of exact phase matching of $n^{(2\omega)} = n^{(\omega)}$ (compared to the case without phase matching) of more then five orders of magnitude

$$\frac{\eta_{\text{opt}}}{\eta} \sim \left(\frac{L}{l_c}\right)^2 = 1.6\times 10^5\ .$$

To allow for exact phase matching the birefringence of anisotropic crystals is of advantage. Nonlinear crystals that are used for frequency doubling lack a center of inversion symmetry, and therefore always have at least two different refractive indices. For the case of an optically uniaxial sample these are ordinary n_o and extraordinary refractive index n_e, respectively. In this case and with a purely ordinary or extraordinarily polarized pump wave this process is called type I phase matching; if the pump wave has a mixed polarization the phase matching is of type II.

Because with the usual situation of normal dispersion the refractive index increases with light frequency ω, i.e. for identical polarisation we have $n^{(2\omega)} > n^{(\omega)}$. Using birefringence the two possible solutions are

$$n_e^{(2\omega)} = n_o^{(\omega)} \qquad \text{or} \qquad n_o^{(2\omega)} = n_e^{(\omega)}\ .$$

In the following table the refractive indices of the nonlinear crystal KDP (KH_2PO_4) are given. Exact phase matching is thus possible for a pump wavelength of about 2 μm. However, in a real system the exact wavelength of pump light is usually fixed, e.g. by typical pump lasers like Nd:YAG or Nd:YVO_4 at 1.064 μm. Therefore an additional degree of freedom to allow for phase matching is necessary. This becomes possible for example by changing the propagation direction of wave inside the crystal.

Table 8-2 Index of Refraction Dispersion Data of KH_2PO_4 (After Reference [8].

		Index		
Wavelength, μm		n_o (ordinary ray)		n_e (extraordinary ray)
0.2000		1.622630		1.563913
0.3000		1.545570		1.498153
0.4000		1.524481		1.480244
0.5000		1.514928		1.472486
0.6000		1.509274		1.468267
0.7000		1.505235		1.465601
0.8000		1.501924		1.463708
–> 0.9000		1.498930	ω –>	1.462234
1.0000		1.496044		1.460993
1.1000		1.493147		1.459884
1.2000		1.490169		1.458845
1.3000		1.487064		1.457838
1.4000		1.483803		1.456838
1.5000		1.480363		1.455829
1.6000		1.476729		1454797
1.7000		1.472890		1.453735
1.8000		1.468834		1.452636
–> 1.9000	2ω –>	1.464555		1.451495
2.0000		1.460044		1.450308

As an example we assume an unaxial crystal with negative birefringence $(n_e < n_o)$ and take into account that the extraordinary refractive index depends on the propagation direction of the wave, i.e. it is a function of the angle θ between Poynting vector and the optical axis of the crystal:

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

with $$n_e^{(2\omega)} < n_e^{(2\omega)}(\theta) < n_o^{(2\omega)} \quad .$$

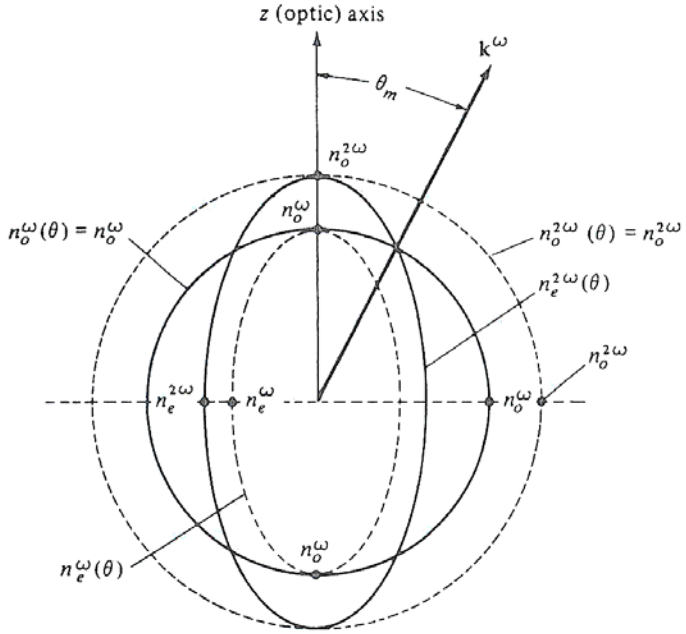
As a result there may exist an angle (this depends actually on the magnitude of birefringence) θ_m, so that the following relation is fulfilled:

$$n_e^{(2\omega)}(\theta_m) = n_o^{(\omega)} \quad .$$

In this case we have an ordinarily polarized pump wave $(i = j)$ and an extraordinarily polarized second harmonic $(k \neq i)$ propagating in the same direction. The phase matching angle θ_m can be obtained by equating the above equation for the case of negative birefringence $(n_e < n_o)$ and with the relation $n_o^{(\omega)} = n_e^{(2\omega)}(\theta_m)$:

$$\frac{1}{(n_o^{(\omega)})^2} = \frac{\cos^2(\theta_m)}{(n_o^{(2\omega)})^2} + \frac{\sin^2(\theta_m)}{(n_e^{(2\omega)})^2} \quad ,$$

$$\sin^2(\theta_m) = \frac{(n_o^{(\omega)})^{-2} - (n_o^{(2\omega)})^{-2}}{(n_e^{(2\omega)})^{-2} - (n_o^{(2\omega)})^{-2}} \quad .$$



As an example we take frequency doubling of a Ruby laser in a KDP crystal with pump wavelength $\lambda^{(\omega)} = 693\ \mathrm{nm}$. With the numbers

$$n_e^{(\omega)} = 1.466 \qquad n_e^{(2\omega)} = 1.487$$
$$n_o^{(\omega)} = 1.506 \qquad n_o^{(2\omega)} = 1.534$$

we find the phase matching angle to be $\theta_m = 50.4\,°$.

an alternative way to obtain exact phase matching is to use a mixed polarization of the pump wave (type II phase matching). Here the pump wave is polarized at an angle of 45° to the optical axis $(i \neq j)$, i.e. we now have

$$n_e^{(2\omega)}(\theta_m) = \frac{1}{2}\left[n_o^{(\omega)} + n_e^{(\omega)}(\theta_m)\right].$$

Again we make use of the dependence of refractive index on propagation direction

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

and

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

From the phase matching condition it follows that

$$\left(\frac{\cos^2(\theta_m)}{(n_o^{(2\omega)})^2} + \frac{\sin^2(\theta_m)}{(n_e^{(2\omega)})^2}\right)^{-\frac{1}{2}} = \frac{1}{2}\left(n_o^{(\omega)} + \left[\frac{\cos^2(\theta_m)}{(n_o^{(\omega)})^2} + \frac{\sin^2(\theta_m)}{(n_e^{(\omega)})^2}\right]^{-\frac{1}{2}}\right).$$

Summarizing the conditions for phase matching

Type I: both pump waves $E_i^{(\omega)}$, $E_j^{(\omega)}$ have the same polarization, $i = j$

Type II: pump waves $E_i^{(\omega)}$, $E_j^{(\omega)}$ have different polarization, $i \neq j$

	positive birefringent $n_e > n_o$	negative birefringent $n_e < n_o$
type I	$n_o^{(2\omega)} = n_e^{(\omega)}(\theta_m)$	$n_e^{(2\omega)}(\theta_m) = n_o^{(\omega)}$
type II	$n_o^{(2\omega)}(\theta_m) = \frac{1}{2}\left(n_o^{(\omega)} + n_e^{(\omega)}(\theta_m)\right)$	$n_e^{(2\omega)}(\theta_m) = \frac{1}{2}\left(n_e^{(\omega)}(\theta_m) + n_o^{(\omega)}\right)$

Here it is important to mention that not all crystals can be phase-matched by changing the propagation direction for a given pump wavelength. For example, the mixed crystal SBN75 ($Sr_{0.75}Ba_{0.25}Nb_2O_6$, strontium-barium niobate), which has rather large SHG coefficients d, but at the same time has a small birefringence $n_o - n_e \leq 0.012$ but a larger dispersion $n_e^{(2\omega)} - n_e^{(\omega)} \approx 0.2$, is not phase-matchable in the wavelength range $0.5\ \mu\text{m} < \lambda < 3\ \mu\text{m}$.

Already for smaller deviations of the exact phase matching condition the efficiency of second harmonic generation is considerable reduced:

$$\eta = \frac{P^{(2\omega)}}{P^{(\omega)}} \sim \frac{\sin^2(\Delta k\, L/2)}{(\Delta k\, L/2)^2} \ .$$

Let δ be the deviation of the exact phase matching angle

$$\delta = \theta - \theta_m \ .$$

For small values of δ we have the approximation

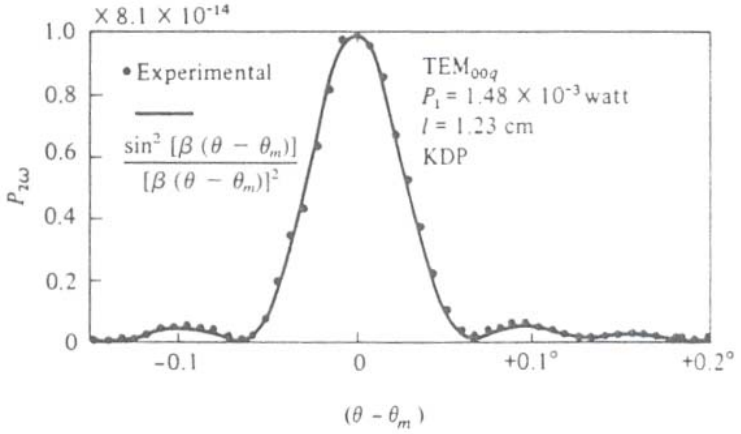
$$\Delta k(\theta) = k_e^{(2\omega)}(\theta) - 2k_o^{(\omega)} \approx 2\beta\delta \ ,$$

with $$\beta = \beta(n_o^{(\omega)}, n_o^{(2\omega)}, n_e^{(2\omega)}) = const. \ .$$

With the definition $\psi := \beta\delta L$ we can rewrite the efficiency in the form

$$\eta = \frac{P^{(2\omega)}}{P^{(\omega)}} \sim \frac{\sin^2\psi}{\psi^2} \ .$$

An experimental proof of this relation is given in the following diagram for the case of a KDP crystal.



Another option to achieve phase matching is to use the temperature dependence of refractive indices, where one can take advantage of the fact that in most case the refractive indices n_o and n_e have different thermooptic coefficients. For optical uniaxial samples one finds

$$\frac{\partial n_e}{\partial T} > \frac{\partial n_o}{\partial T} \quad .$$

As an example we take the uniaxial crystal $LiNbO_3$ with $n_e < n_o$. Lithium niobate is a ferroelectric material with a Curie temperature $T_c \approx 1210°\ \mathrm{C}$. At temperatures above the Curie temperature, $T > T_c$, the crystal is in the paraelectric phase and shows inversion symmetry. On the other hand, at temperatures below T_c the crystal is ferroelectric and thus possesses a spontaneous polarization P_s (where the direction of polarization is related to the sign of the tensor elements d_{ijk}) as well as different refractive indices $n_e \neq n_o$. Starting from room temperature an increase in T usually results in a decrease of birefringence and thus leads to $n_e \to n_o$ for $T \to T_c$. Here one has to take into account that the thermooptic coefficient dn/dT is a function of both temperature and wavelength and may show a more complicated behaviour. Typical numbers for $LiNbO_3$ are

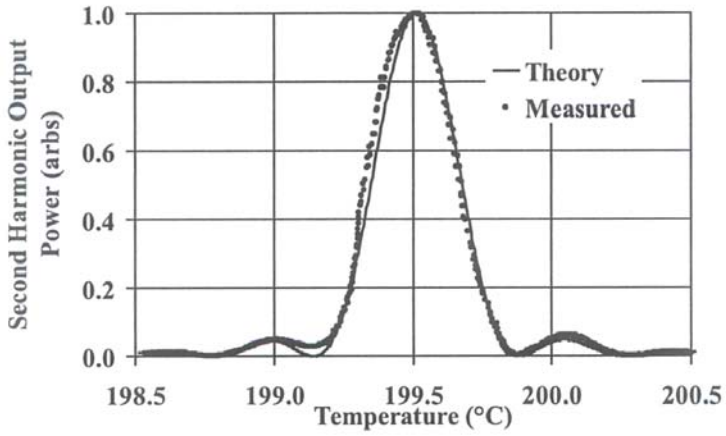
$$\lambda = 0.5\,\mu\mathrm{m} \quad : \quad \frac{d n_e}{d T} \approx 10^{-4} \quad , \quad \frac{d n_o}{d T} \approx 3\times10^{-5}$$

$$\lambda = 1\,\mu\mathrm{m}: \quad \frac{d n_e}{d T} \approx 4\times10^{-5} \quad , \quad \frac{d n_o}{d T} \approx 0$$

As an approximation we may use

$$\frac{d (n_e - n_o)}{d T} \approx \frac{d n_e}{d T} \quad .$$

For the case of type I phase matching for the material KDP (see table in this section) frequency doubling of a Nd:YAG laser at a wavelength λ = 1.064 μm is possible by tuning the crystal temperature to the phase matching temperature T_{pm}. An example is given in the next diagram.



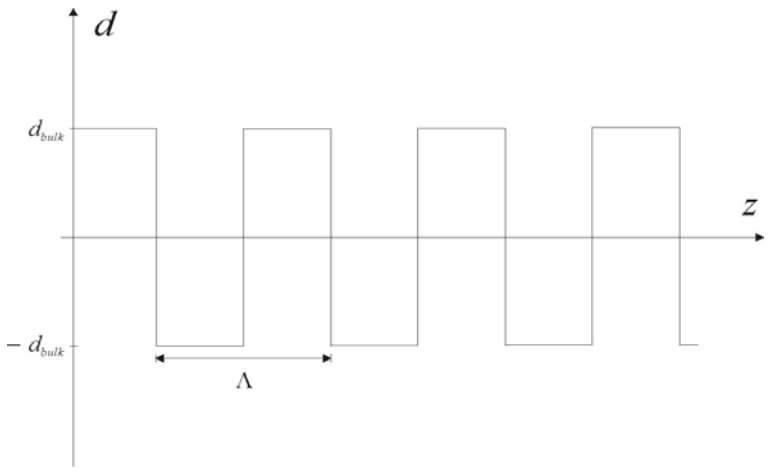
3.3 Quasi phase matching

In many cases materials that may be used for frequency doubling, for example the ferroelectric crystals $LiNbO_3$ and $LiTaO_3$, shown rather large diagonal elements of the d Tensors (e.g. d_{333}), which cannot be used by type I or type II phase matching. Therefore, as has been shown in the proceeding section, the pump and frequency-doubled wave have orthogonal polarizations, thus using for example the coefficient $d_{311} \hat{=} d_{31}$ (or $d_{312} = d_{36}$). In the case of $LiNbO_3$ or $LiTaO_3$ the ratio of the involved coefficients is $d_{333}/d_{311} \approx 3$. Consequently, using the larger coefficient d_{333} would result in an increase of SHG efficiency of about one order of magnitude.

An alternative technique for achieving phase matching is referred to as quasi phase-matching (QPM), where the nonlinear coefficient is periodically modulated along the propagation direction $d = d(z)$. The periodic modulation of the nonlinear coefficient d can be expanded in a Fourier series

$$d(z) = d_{bulk} \sum_m -i\, a_m \exp\left(i\, m \frac{2\pi}{\Lambda} z \right)$$

with d_{bulk} being the nonlinearity of the homogeneously polarized crystal, the period length Λ and the Fourier coefficients a_m of order m. An example that is close to application is a purely binary modulation $d = d(z)$ with amplitudes $\pm d_{bulk}$.



Insertion of the Fourier expansion of $d = d(z)$ into the differential equation system (for the case $E_{01}^{(\omega_1)} = E_{02}^{(\omega_2)}$) results in

$$\frac{d\,E_{01}^{(\omega_1)}}{d\,z} = -\frac{\sigma_1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0\,\varepsilon_1}}\,E_{01}^{(\omega_1)} - \frac{i\,\omega_1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0\,\varepsilon_1}}\,d_{bulk}\,E_{03}^{(\omega_3)}\,E_{01}^{*(\omega_1)}\sum_m -i\,a_m \exp\left[i\left(m\frac{2\pi}{\Lambda} - k_3 + k_1 + k_1\right)z\right]$$

For QPM the phase matching condition is

$$m\,\frac{2\pi}{\Lambda} = \Delta k = k_3 - 2\,k_1 \qquad m = 1, 2, 3, ...$$

This condition is assumed to be fulfilled for a certain $m = n$; for all other $m \neq n$ their contributions averages out to zero over distances that are large compared to the coherence length, i.e. for $L >> \Lambda$. The magnitude of the coefficients a_m can be obtained by applying the orthogonality of the different terms in the sum over m:

$$\frac{d(z)}{d_{bulk}} = \sum_m -i\,a_m \exp\left(i\,m\,\frac{2\pi}{\Lambda}\,z\right)$$

$$\frac{1}{\Lambda}\int_0^{\Lambda}\frac{d(z)}{d_{bulk}}\,i\,\exp\left(-i\,m\,\frac{2\pi}{\Lambda}\,z\right)$$

$$= \frac{1}{\Lambda}\int_0^{\Lambda}\sum_m a_m \exp\left(i\,m\,\frac{2\pi}{\Lambda}\,z\right)\exp\left(-i\,n\,\frac{2\pi}{\Lambda}\,z\right)dz \;= a_m\,\delta_{mn}$$

$$a_m = \frac{1}{\Lambda}\int_0^{\Lambda} i\,\frac{d(z)}{d_{bulk}}\exp\left(-i\,m\,\frac{2\pi}{\Lambda}\,z\right)dz$$

As an example we again treat the case of $d = d(z)$ where the sign changes every halve grating period:

$$a_m = \frac{i}{\Lambda}\int_0^{\Lambda}\frac{d(z)}{d_{bulk}}\exp\left(-i\,m\,\frac{2\pi}{\Lambda}\,z\right)dz \qquad \text{(with } m \neq 0\text{)}$$

$$= \frac{i}{\Lambda}\left[\int_0^{\Lambda/2}\exp\left(-i\,m\,\frac{2\pi}{\Lambda}\,z\right)dz + \int_{\Lambda/2}^{\Lambda} -\exp\left(-i\,m\,\frac{2\pi}{\Lambda}\,z\right)dz\right]$$

$$= \frac{i}{\Lambda}\,\frac{-\Lambda}{i\,m\,2\pi}\left[\exp(-i\,m\,\pi) - 1 - \exp(-i\,m\,2\pi) + \exp(-i\,m\,\pi)\right]$$

$$= \frac{1}{m\,\pi}\,\frac{1}{2}\left[1 - 2\exp(-i\,m\,\pi) + \exp(-i\,m\,2\pi)\right]$$

$$= \frac{1}{2\,m\,\pi}\left[1 - 2\cos(-m\,\pi) - \underbrace{2\,i\sin(-m\,\pi)}_{=0} + \underbrace{\cos(-m\,2\pi)}_{=1} + \underbrace{\sin(-m\,2\pi)}_{=0}\right]$$

$$= \frac{1}{m\,\pi}\,\frac{1}{2}\left[2 - 2\cos(m\,\pi)\right] = \frac{1 - \cos(m\,\pi)}{m\,\pi}\;.$$

For the case $m = 1$ we obtain

$$d_{eff} = a_m \, d_{bulk} = \frac{2}{\pi} \, d_{bulk} \; .$$

Compared to the situation of true phase matching the efficiency of quasi phase-matching is reduced by a factor $\eta_{QPM} / \eta = (2/\pi)^2 \approx 0.4$.

A numerical example of quasi phase-matching for SHG using $LiNbO_3$ at a wavelength of 1 μm is given below:

$$\Delta k = k_3 - 2k_1 = \frac{4\pi (n_e^{(2\omega)} - n_e^{(\omega)})}{\lambda} = \frac{4\pi}{1\mu\text{m}} (2.25 - 2.16) \approx 1.2 \times 10^6 \, \text{m}^{-1}$$

$$\Lambda = \frac{2\pi}{\Delta k} = \frac{\lambda}{2} \, (n_e^{(2\omega)} - n_e^{(\omega)})^{-1} \approx 5 \, \mu\text{m}$$

The case Fall $m = 1$ requires a period length of only 5 μm or a coherence length of $l_c = 2.5$ μm, respectively. Such small period lengths (the structure size is only half the period length) may be not easy to achieve in real applications. Therefore also quasi phase matching with $m > 1$ may be used. For the case $m = 3$ one requires less-demanding periods of 5 μm, however, at the same time the relative efficiency is further reduced to $\eta_{QPM,\,m=3} / \eta = (2/3\pi)^2 \approx 0.05$.

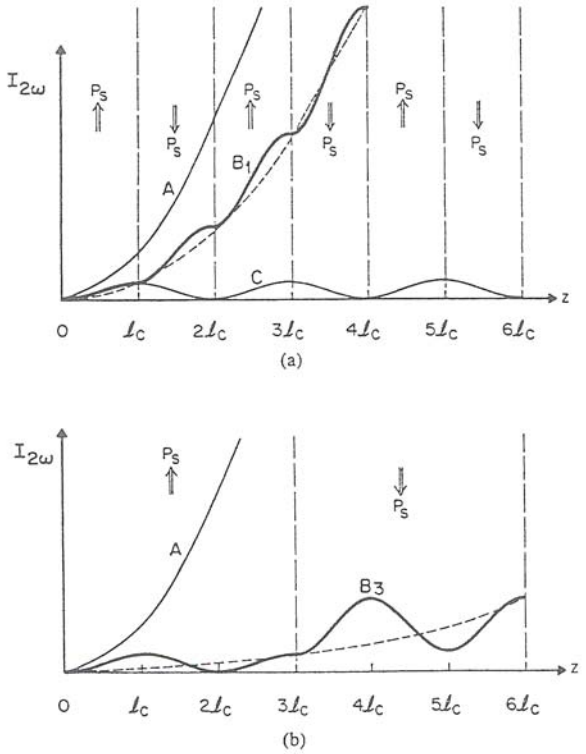


Fig. 1. Effect of phase matching on the growth of second harmonic intensity with distance in a nonlinear crystal. (a) A: perfect phase matching in a uniformly poled crystal; *C*: nonphase-matched interaction; B_1: first-order QPM by flipping the sign of the spontaneous polarization every coherence length of the interaction of curve C. (b) A: perfect phase matching; B_3: third-order QPM by flipping P_S every three coherence lengths.

For the fabrication of periodic structures for quasi phase matching the spontaneous polarization P_s of a ferroelectric crystal has to be reversed periodically. This can be achieved by applying an external voltage to grating-like ("finger") electrodes, which are formed by lithographic techniques on the crystal's surface. The dependence $P_s(E)$ shows the typical hystersis behaviour of ferroelectric media.

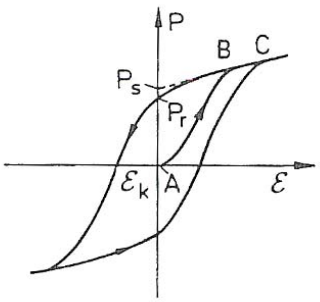
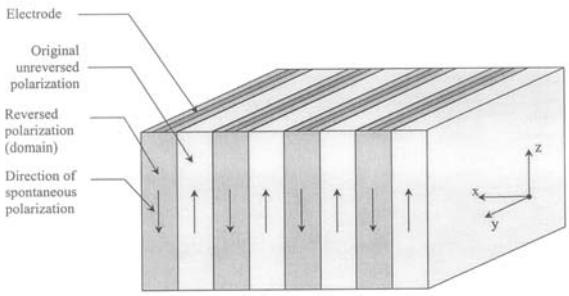


Fig. 4.14
Hysteresekurve eines ferroelektrischen Kristalls, siehe Text.

The necessary coercive field E_c, at which the spontaneous polarization is reversed, is usually higher then the electrical break-down threshold of air (i.e., for $LiNbO_3$ $E_c = 22\ \mathrm{kV/mm}$), thus a treatment in isolating atmosphere (e.g., silicone oil) has to be used.



The poling procedure cannot be understood as a simple switching of domains. Instead, this is a more complicated, time-dependent process, with the growing speed of the domains (which by itself strongly depend on the external poling field) as the most important parameter. A scheme of the poling process can be seen in the following figure. In a first step, needle-like start growing at the edges of the finger electrodes, because there the local electric field has a maximum value (a-c). Then the domains start to grow in lateral direction (d,e), which accidentally may result also in fusion of neighboured domains. Finally, if the poling procedure is stopped, a periodic modulation of the spontaneous polarization is achieved (f). The Figure on the right shows a photograph of the domain structure which has been made visible by etching of the crystal's surface.