Nonlinear Optics (WiSe 2017/18)

Lecture 4: October 26, 2017

4 Frequency doubling

4.1 Without depletion of fundamental wave

4.2 With depletion of fundamental wave

4.3 Wave propagation in linear non-isotropic media

4.3.1 Ordinary wave

4.3.2 Extraordinary wave

4.4 Phase matching

4.4.1 Birefringent phase matching

4.4.2 Frequency doubling of Gaussian beams

4.4.3 Frequency doubling of pulses

4.4.4 Effective nonlinear coefficient d_{eff}

4.4.5 Quasi-phase matching (QPM)

Repetition: Nonlinear Wave Equation

Employing the dispersion relation $k^2 = \mu_0 \varepsilon_0 \varepsilon_r \omega^2$ the two leading terms cancel, and within the SVEA (3.6),(3.7) it follows

$$2jk\frac{\partial}{\partial z}E + 2j\frac{\omega}{c^2}\frac{\partial}{\partial t}E = -j\omega\mu_0\sigma E + \mu_0\omega^2 P_{NL}\left(\hat{\mathbf{e}}\cdot\hat{\mathbf{p}}\right)e^{j(k-k_p)z},$$

where we introduced the velocity of light in the linear medium as $c = \sqrt{\mu_0\varepsilon_0\varepsilon_r}^{-1}$. We divide this equation by $2jk$ and transform it into a comoving time frame using $t' = t - z/c$, $(z = z')$, and obtain

$$\frac{\partial}{\partial z}E(z,t') = -\alpha E(z,t') - \frac{1}{2}j\omega Z_\omega P_{NL}(z,t')\left(\hat{\mathbf{e}}\cdot\hat{\mathbf{p}}\right)e^{j(k-k_p)z}, \qquad (3.8)$$

with the damping constant $\alpha = \sigma Z_\omega/2$ and the impedance of the medium $Z_\omega = \frac{1}{\varepsilon_0\sqrt{\varepsilon_{r,\omega}}c_0} = \sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_{r,\omega}}}$.

Some remarks on

$$\frac{\partial}{\partial z}E(z,t') = -\alpha E(z,t') - \frac{1}{2}j\omega Z_\omega P_{NL}(z,t')\,(\hat{\mathbf{e}}\cdot\hat{\mathbf{p}})\,e^{j(k-k_p)z}, \qquad (3.8)$$

•The medium **conductivity σ** leads to **losses** and therefore **damping** of the propagating wave.

•The medium's nonlinear polarization can lead to both **gain or damping**, depending on the **relative phase** between the electric field and the polarization (parametric amplification, frequency conversion, stimulated scattering processes as Raman and Brillouin scattering, multi-photon absorption).

•If the nonlinear polarization is **in phase or in opposite phase** of the electric field, it corresponds to a a nonlinear change of the refractive index, leading to a **phase shift of the electric field** (Pockels effect, Kerr effect).

•If the polarization is advancing the field by 90^o, the polarization is supplying energy to the field. In the opposite case, the polarization is extracting energy from the field.

•**phase relation is changing during propagation**, if no phase matching of the process, i.e., $k = k_p$, is achieved.

4. Frequency doubling

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GENERATION OF OPTICAL HARMONICS*

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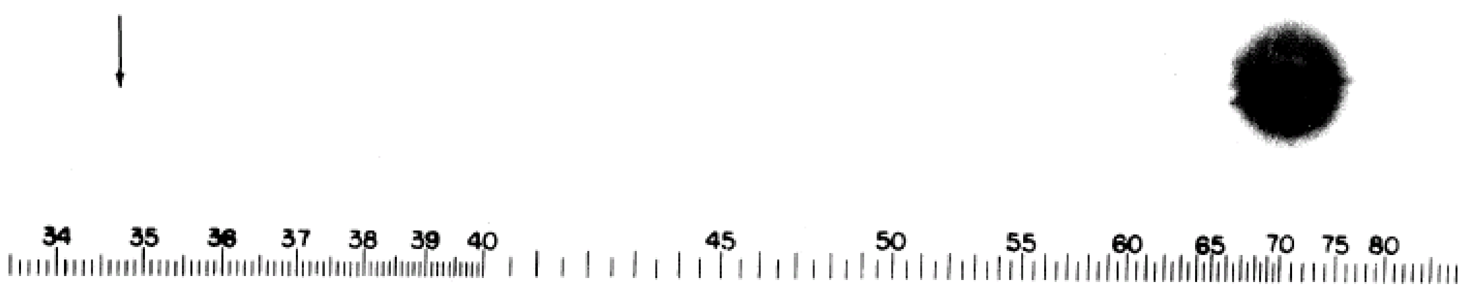
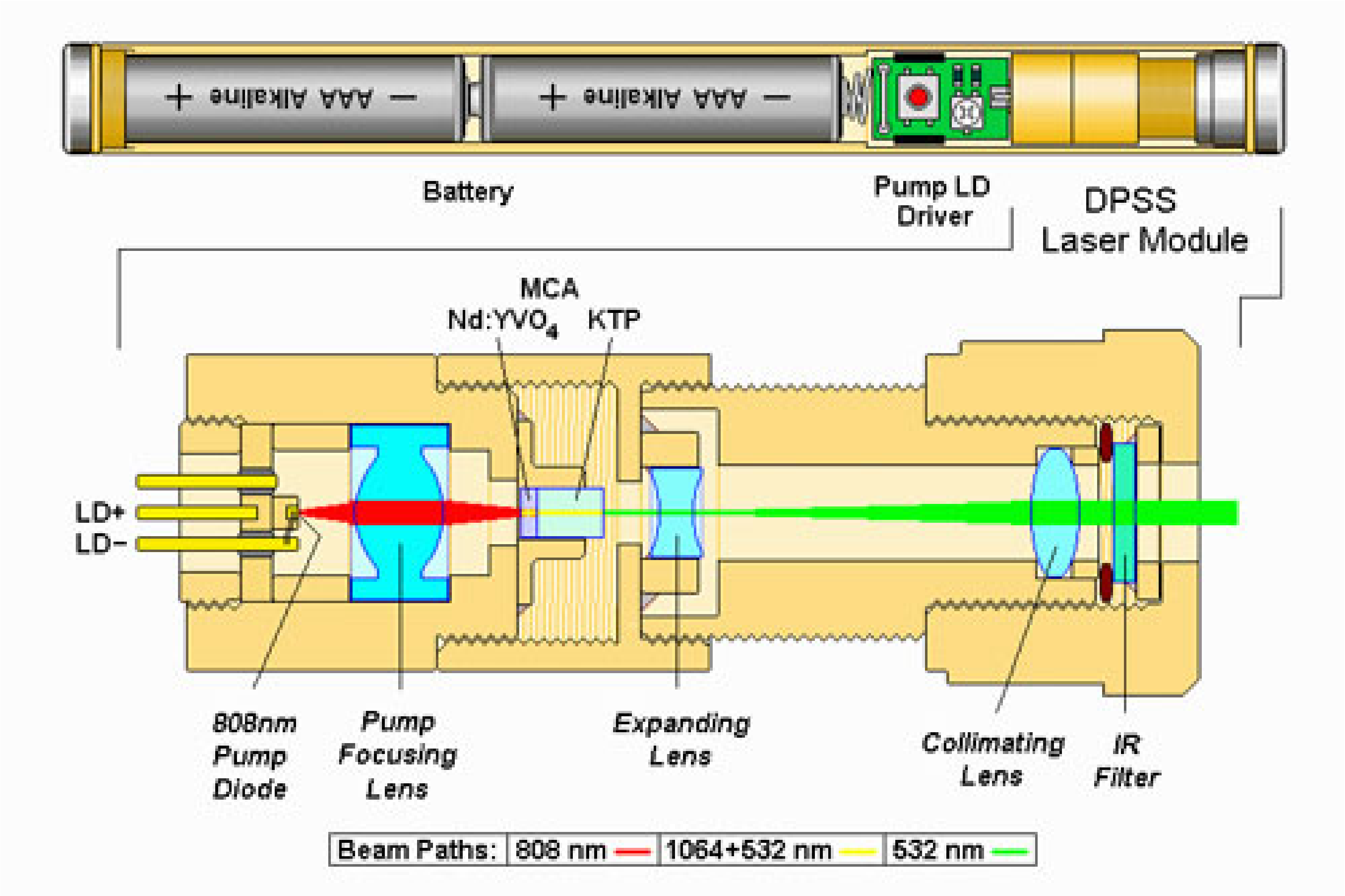


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt and didn’t ask the authors anymore.

SHG in daily life: green laser pointer



Second harmonic generation (SHG)

$$\hat{P}(2\omega) = \varepsilon_0 d_{eff}(2\omega;\omega,\omega)\hat{E}(\omega,z)\hat{E}(\omega,z). \qquad (4.1)$$

We neglect any losses for the moment ($\alpha = 0$), and $Z_\omega = \frac{1}{n_\omega}\sqrt{\frac{\mu_0}{\varepsilon_0}} = \frac{1}{n_\omega}\frac{1}{\varepsilon_0 c_0}$ from Eq..(3.8)

$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c_0} d_{eff}(2\omega;\omega,\omega)\hat{E}(\omega,z)\hat{E}(\omega,z)\mathrm{e}^{j(k(2\omega)-2k(\omega))z} \qquad (4.2)$$

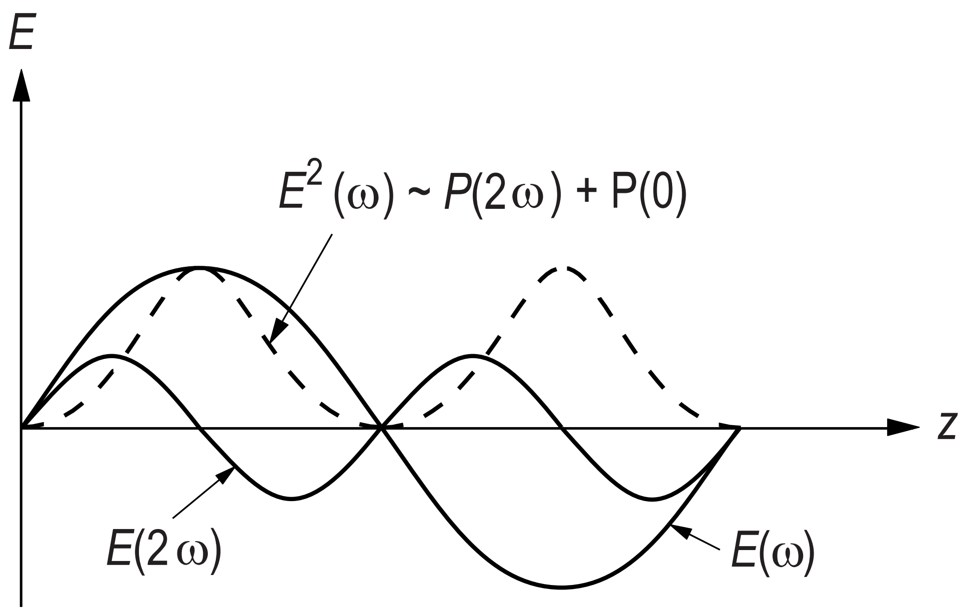


Fig. 1: Phase relationships between fundamental, second harmonic and nonlinear polarization.

4.1 Without depletion of fundamental wave

$$\hat{E}(2\omega, z = \ell) = -\frac{j\omega d_{eff}}{n_{2\omega} c_0} \hat{E}^2(\omega) \int_0^\ell \mathrm{e}^{j\Delta k z} dz$$

where $\Delta k = k(2\omega) - 2k(\omega)$ is the difference in wave number between the second harmonic light and twice the wavenumber of the fundamental light or the driving second order nonlinear Polarization.

Second-harmonic generation (SHG)

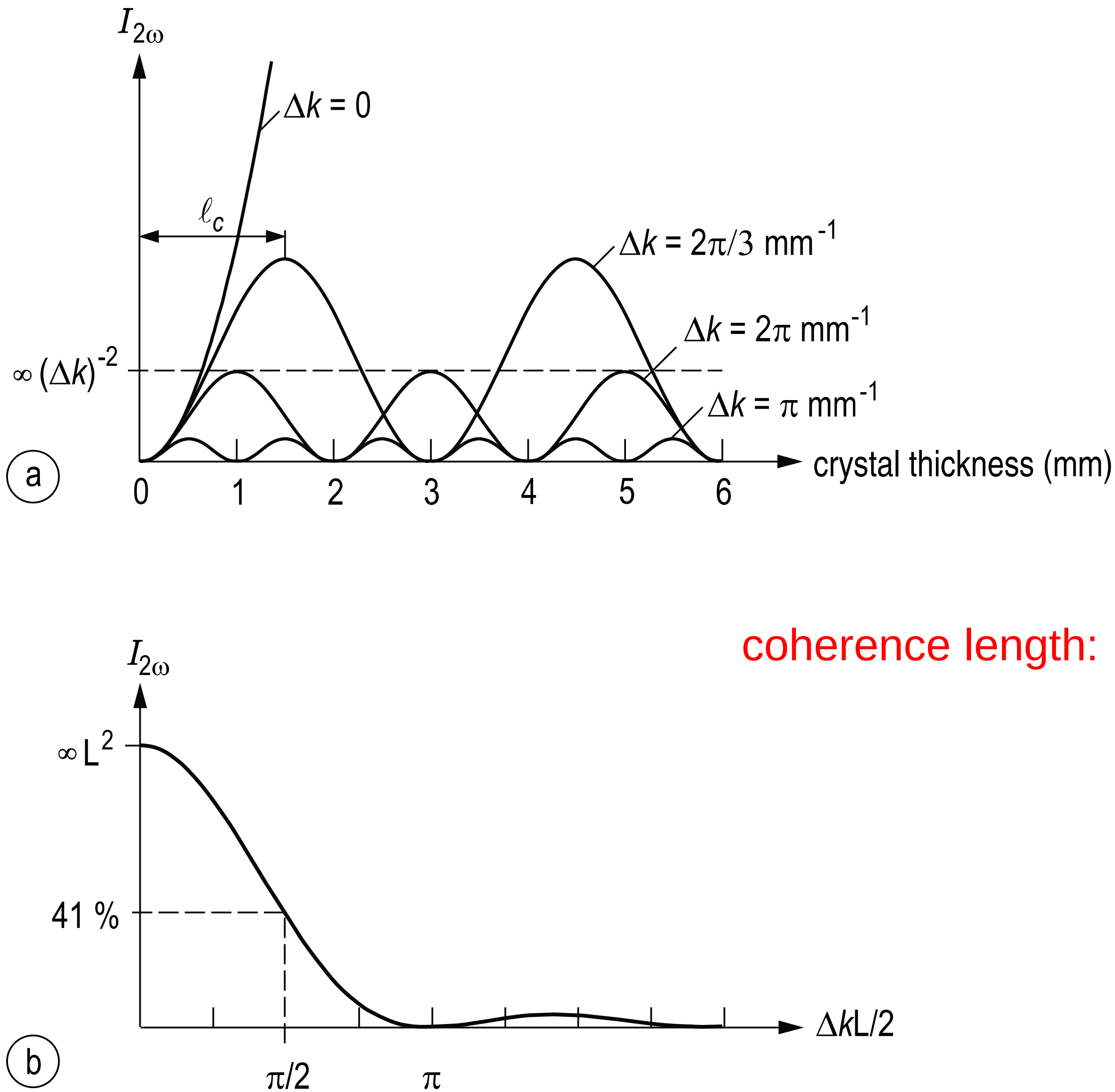
$$\hat{E}(2\omega, \ell) = -\frac{j\omega d_{eff}}{n_{2\omega} c_0} \hat{E}^2(\omega)\ell \cdot \left[\frac{\sin \Delta k\ell/2}{\Delta k\ell/2}\right] \mathrm{e}^{j\Delta k\ell/2}. \qquad (4.3)$$

Introducing the intensities of the fundamental and second harmonic waves

$$I_{\omega,2\omega} = \frac{n_{\omega,2\omega}}{2} \sqrt{\varepsilon_0/\mu_0} |\hat{E}_{\omega,2\omega}|^2$$

wie obtain

$$I(2\omega, \ell) = \frac{2\omega^2 d_{eff}^2}{n_{2\omega} n_\omega^2 c_0^3 \varepsilon_0} \ell^2 I^2(\omega) \left[\frac{\sin \Delta k\ell/2}{\Delta k\ell/2}\right]^2. \qquad (4.4)$$



coherence length: $\ell_c = \dfrac{\pi}{\Delta k}$

Figure 4.2: Second-harmonic generation as function of phase mismatch.

If phase matching can be achieved, one can use Eq. (4.4) to define an inverse conversion length Γ as

$$\Gamma = \frac{\omega d_{eff}}{nc} |\hat{E}(\omega)|, \text{ with } n = \sqrt{n_\omega n_{2\omega}}, \tag{4.6}$$

and

$$I(2\omega, \ell) = \Gamma^2 \ell^2 I(\omega). \tag{4.7}$$

If the medium length reaches the conversion length, i.e., $\Gamma\ell = 1$, then Eq. (4.7) would indicate, that all fundamental light is converted to the second harmonic, which contradicts the assumption of small conversion, and therefore we have to work a little more to correct for it.

4.2 With depletion of the fundamental wave

$$\hat{P}(\omega) = \varepsilon_0 d'_{eff}(\omega; 2\omega, -\omega)\hat{E}(2\omega)\hat{E}^*(\omega).$$

The coupled equations are

$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c_0} d_{eff}\hat{E}(\omega)\hat{E}(\omega)\mathrm{e}^{j\Delta kz} \tag{4.8}$$

and

$$\frac{\partial \hat{E}(\omega)}{\partial z} = -\frac{j\omega}{n_\omega c_0} d'_{eff}\hat{E}(2\omega)\hat{E}^*(\omega)\mathrm{e}^{-j\Delta kz}. \tag{4.9}$$

Both equations describe the energy exchange between fundamental and second-harmonic wave. The intensities are

$$I_\omega = \frac{n_\omega}{2Z_0}\left|\hat{E}(\omega)\right|^2 \text{ and } I_{2\omega} = \frac{n_{2\omega}}{2Z_0}\left|\hat{E}(2\omega)\right|^2 \tag{4.10}$$

lossless media, i.e., d'_{eff} and d_{eff} are real

$$
\begin{aligned}
2Z_0 \frac{dI_{2\omega}}{dz} &= n_{2\omega} \left[\hat{E}^*(2\omega) \frac{\partial \hat{E}(2\omega)}{\partial z} + c.c. \right] = \\
&= -\frac{j\omega}{c_0} d_{eff} \hat{E}^*(2\omega) \hat{E}(\omega) \hat{E}(\omega) \mathrm{e}^{j\Delta k z} + c.c. \\
2Z_0 \frac{dI_{\omega}}{dz} &= n_{\omega} \left[\hat{E}(\omega) \frac{\partial \hat{E}^*(\omega)}{\partial z} + c.c. \right] = -2Z_0 \frac{dI_{2\omega}}{dz}, \text{ if } d'_{eff} = d^*_{eff}.
\end{aligned}
$$

Energy conservation demands permutation symmetry of the conversion coefficients

$$
n_{2\omega} |\hat{E}(2\omega)|^2 + n_{\omega} |\hat{E}(\omega)|^2 = \text{ const.} \equiv n_{\omega} \hat{E}_0^2 = \text{const.} \tag{4.11}
$$

Separating the wave amplitudes with respect to amplitude and phase

$$\hat{E}(\omega) = |\hat{E}(\omega)|\mathrm{e}^{j\Phi(\omega)}$$

$$\hat{E}(2\omega) = |\hat{E}(2\omega)|\mathrm{e}^{j\Phi(2\omega)}$$

$$\frac{d\hat{E}(2\omega)}{dz} = \frac{d|\hat{E}(2\omega)|}{dz}\mathrm{e}^{j\Phi(2\omega)} + j\frac{d\Phi(2\omega)}{dz}|\hat{E}(2\omega)|\mathrm{e}^{j\Phi(2\omega)} \tag{4.12}$$

$$\frac{d|\hat{E}(2\omega)|}{dz} = \mathrm{Re}\left\{-\frac{j\omega d_{eff}}{n_{2\omega}c_0}|\hat{E}(\omega)|^2\mathrm{e}^{2j\Phi(\omega)-j\Phi(2\omega)}\mathrm{e}^{j\Delta kz}\right\} \tag{4.13}$$

$$= \mathrm{Re}\left\{-\frac{j\omega d_{eff}}{n_{\omega}c_0}\{\hat{E}_0^2 - |\hat{E}(2\omega)|^2\}\mathrm{e}^{2j\Phi(\omega)-j\Phi(2\omega)}\mathrm{e}^{j\Delta kz}\right\}. \tag{4.14}$$

General solution: Jacobi elliptic function!

For Δk=0, second harmonic builds up such that

$$-j\mathrm{e}^{2j\Phi(\omega)-j\Phi(2\omega)} = 1$$

Solution for Δk=0

$$\int_0^{|\hat{E}(2\omega)|_\ell} \frac{d|\hat{E}(2\omega)|}{\hat{E}_0^2 - |\hat{E}(2\omega)|^2} = -\int_0^\ell \frac{\omega d_{eff}}{n_\omega c_0} dz. \tag{4.15}$$

Using the integral

$$\int \frac{dx}{a^2 - x^2} = \frac{1}{a} \tanh^{-1}[x/a] \tag{4.16}$$

we obtain

$$|\hat{E}(2\omega)|_{z=\ell} = \hat{E}_0 \tanh\left\{\hat{E}_0 \left(\frac{\omega d_{eff}}{n_\omega c_0}\right) \ell\right\} \tag{4.17}$$

or for the intensity

$$I(2\omega, \ell) = I(\omega, 0) \tanh^2 \left\{\frac{\hat{E}_0 \omega d_{eff}}{n_\omega c_0} \cdot \ell\right\} \tag{4.18}$$

With the conversion rate $\Gamma = \frac{\omega d_{eff}}{n_\omega c_0} \hat{E}_0$ introduced above, we obtain

$$I(2\omega, \ell) = I(\omega, 0) \tanh^2\{\Gamma\ell\} \tag{4.19}$$

With $1-\tanh^2 = \cosh^{-2} = \mathrm{sech}^2$

$$I(\omega,\ell) = I(\omega,0)\mathrm{sech}^2\{\Gamma\ell\}. \qquad (4.20)$$

For perfect phase matching, 100% conversion possible for $\Gamma\ell$ **>> 1**

What to do if there is phase mismatch?

4.3 Wave propagation in linear non-isotropic media

$$\ddot{\mathbf{D}} = \varepsilon\ddot{\mathbf{E}}$$

$$\varepsilon = \varepsilon_0 \begin{bmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{bmatrix}$$

$$\nabla\times\nabla\times\hat{\mathbf{E}} = -\omega^2\mu_0\varepsilon\hat{\mathbf{E}} \qquad (4.21)$$

Wave propagation in linear non-isotropic media

As in isotropic media, there are plane-wave solutions with

$$\hat{\mathbf{E}} = \hat{\mathbf{E}}_0 e^{-j\mathbf{k}\cdot\mathbf{r}} \tag{4.22}$$

that obey

$$\mathbf{k}\times\mathbf{k}\times\hat{\mathbf{E}} = -\omega^2\mu_0\varepsilon\hat{\mathbf{E}} \tag{4.23}$$

The wave vector is orthogonal to the displacement vector but in general not anymore to the electric field

$$\mathbf{k} \perp (\varepsilon\hat{\mathbf{E}} = \hat{\mathbf{D}}).$$

From Faraday's law we have

$$j\mathbf{k}\times\hat{\mathbf{E}} = -\omega\hat{\mathbf{B}} \tag{4.24}$$

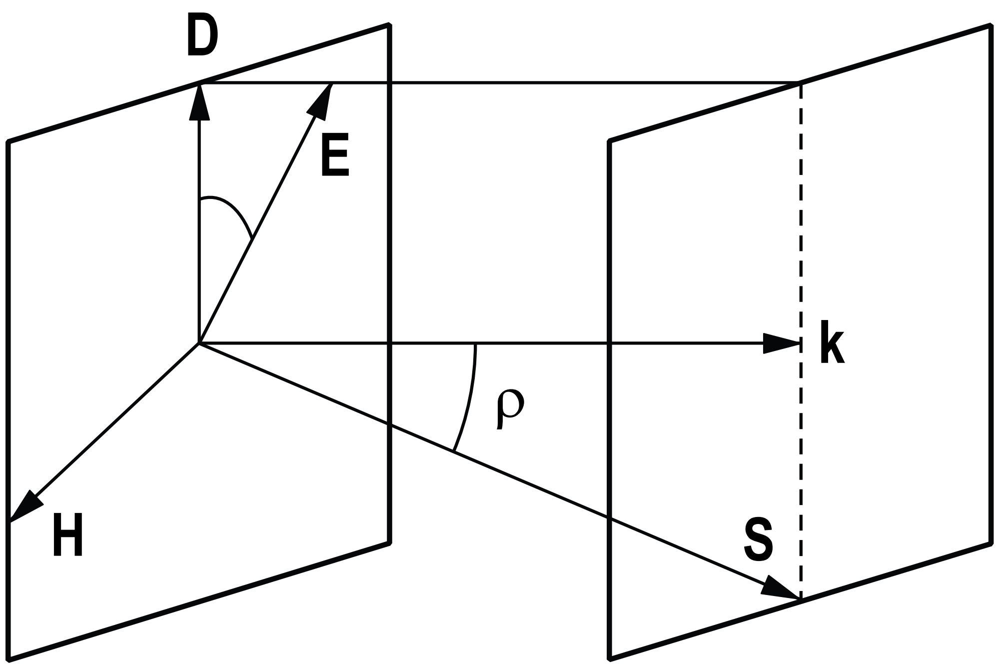
and therefore, as in the isotropic case, we have

$$\mathbf{k} \perp \hat{\mathbf{B}} \parallel \hat{\mathbf{H}}.$$

$\hat{\mathrm{E}} \parallel \hat{\mathrm{D}}$ **: only when parallel to a main axis**

Poynting vector $\mathbf{S} = \mathbf{E} \times \mathbf{H}$, is always normal to $\mathbf{E}$ and $\mathbf{H}$

not necessarily parallel to the wave vector



D parallel to phase fronts

E in general not parallel to phase fronts

S not necessarily parallel to **k**

Figure 4.3: Relationship between field vectors, wave vector and Poynting vector of a plane wave in birefringent media.

Form of dielectric susceptibility tensor

isotropic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & xx \end{bmatrix}$	cubic
uniaxial	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$	tetragonal trigonal hexagonal
biaxial	$\begin{bmatrix} xx & 0 & 0 \\ 0 & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$	orthorhombic
	$\begin{bmatrix} xx & 0 & xz \\ 0 & yy & 0 \\ xz & 0 & zz \end{bmatrix}$	monoclinic
	$\begin{bmatrix} xx & xy & xz \\ xy & yy & yz \\ xz & yz & zz \end{bmatrix}$	triclinic

Table 4.1: Form of the dielectric susceptibility tensor for the different crystal systems.

In the following, we consider the uniaxial case

$$\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_1 \neq \varepsilon_{zz} = \varepsilon_3$$

The corresponding refractive indices are called ordinary and extraordinary indices.

$$n_1 = n_o \neq n_3 = n_e.$$

Further one distinguishes between positive uniaxial, $n_e > n_o$, and negativ

Propagation different from main axes

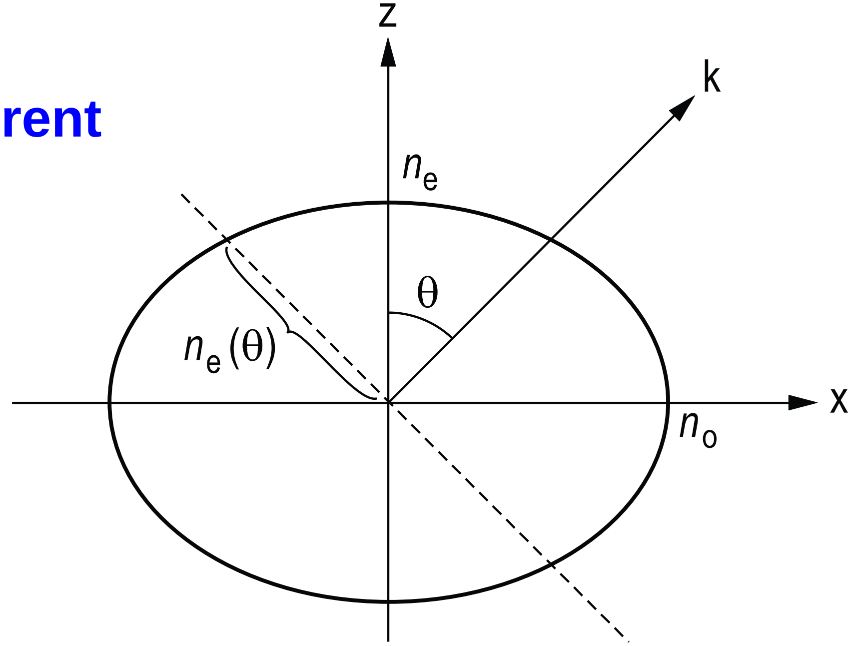


Figure 4.4: Index ellipsoid

Nonlinear optical susceptibilities

$$k^2\hat{\mathbf{E}} + \omega^2\mu_0\varepsilon\hat{\mathbf{E}} = \mathbf{0}. \tag{4.25}$$

$$\begin{pmatrix} k_0^2 n_o^2 + k_x^2 - k^2 & & k_x k_z \\ & k_0^2 n_o^2 - k^2 & \\ k_z k_x & & k_0^2 n_e^2 + k_z^2 - k^2 \end{pmatrix} \hat{\mathbf{E}} = \mathbf{0} \tag{4.26}$$

y-polarized wave decouples → ordinary wave $k^2 = k_0^2 n_o^2.$

As the wave in an isotropic medium, it is purely transversal, $\mathbf{k} \perp \hat{\mathbf{E}} \perp \hat{\mathbf{H}}$.

Wave in the x-z plane with polarization in x-z plane: extraordinary wave

$$\det \begin{vmatrix} k_0^2 n_o^2 + k_x^2 - k^2 & k_x k_z \\ k_z k_x & k_0^2 n_e^2 + k_z^2 - k^2 \end{vmatrix} = \mathbf{0}$$

or after some brief transformations

$$\frac{k_z^2}{n_o^2} + \frac{k_x^2}{n_e^2} = k_0^2. \tag{4.27}$$

With $k_x = k \sin(\theta)$, $k_z = k \cos(\theta)$ and $k = n(\theta) k_0$ we obtain for the refractive index of the extraordinary wave

$$\frac{1}{n(\theta)^2} = \frac{\cos^2(\theta)}{n_o^2} + \frac{\sin^2(\theta)}{n_e^2}. \tag{4.28}$$

$$v_g = \nabla_k \omega(\mathbf{k}) \parallel \mathbf{S},$$

normal to index ellipsoid and parallel to Poynting vector

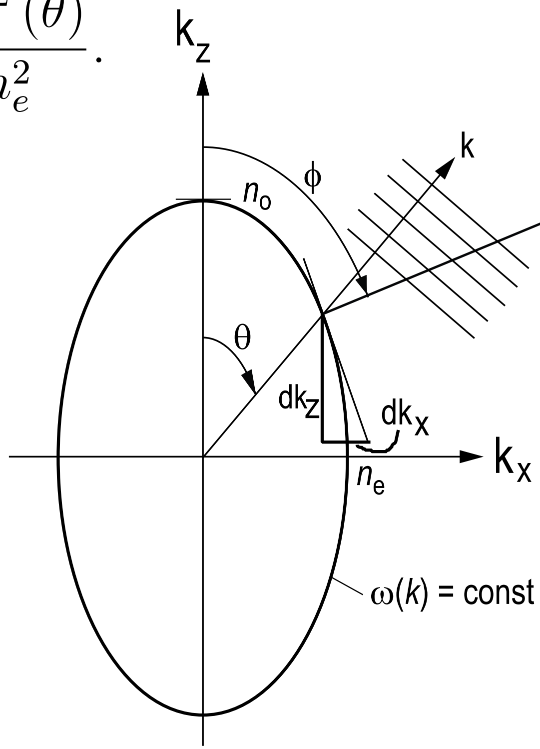


Figure 4.5: Cut through the surface of the index ellipsoid with constant free-space value $k_o(k_x, k_y, k_z)$ or frequencies.

and is normal to the index ellipsoid. To determine the “walk-off” angle between the Poynting vector and the wave vector, we consider

$$\tan\theta = \frac{k_x}{k_z}$$

$$\tan\phi = -\frac{dk_z}{dk_x}\,.$$

From Eq. (4.27) we find

$$\frac{2k_z dk_z}{n_o^2} + \frac{2k_x dk_x}{n_e^2} = 0, \tag{4.29}$$

and

$$\tan\phi = \frac{n_o^2 k_x}{n_e^2 k_z} = \frac{n_o^2}{n_e^2}\tan\theta\,.$$

, we obtain for the walk-off angle between Poynting vector and wave number vector

$$\tan\varrho = \tan\left(\theta - \phi\right) = \frac{\tan\theta - \tan\phi}{1 + \tan\theta\tan\phi} \tag{4.30}$$

$$\tan\varrho = \frac{\left(1 - \frac{n_o^2}{n_e^2}\right)\tan\theta}{1 + \frac{n_0^2}{n_e^2}\tan^2\theta}.$$