Chapter 2

Basics of nonlinear optics

2.1 Nonlinear susceptibility

Linear dependence (1.13) is just an approximation. Generally, the polarization of a medium is an arbitrary function of the past history of the applied electric field. This function can be decomposed into a Taylor series

$$\begin{aligned}\vec{P}(t) \;=\;& \epsilon_0 \frac{1}{2\pi} \int\limits_{-\infty}^{+\infty} \tilde{\vec{\vec{\chi}}}^{(1)}(\tau)\vec{E}(t-\tau)\mathrm{d}\tau \\ &+\epsilon_0 \frac{1}{(2\pi)^2} \iint\limits_{-\infty}^{+\infty} \tilde{\vec{\vec{\chi}}}^{(2)}(\tau_1,\tau_2)\vec{E}(t-\tau_1)\vec{E}(t-\tau_2)\mathrm{d}\tau_1\mathrm{d}\tau_2 \\ &+\epsilon_0 \frac{1}{(2\pi)^3} \iiint\limits_{-\infty}^{+\infty} \tilde{\vec{\vec{\chi}}}^{(3)}(\tau_1,\tau_2,\tau_3)\vec{E}(t-\tau_1)\vec{E}(t-\tau_2)\vec{E}(t-\tau_3)\mathrm{d}\tau_1\mathrm{d}\tau_2\mathrm{d}\tau_3 + \ldots,\end{aligned} \tag{2.1}$$

where $\tilde{\vec{\vec{\chi}}}^{(i)}(\tau_1,\ldots,\tau_i)$ is the *nonlinear susceptibility tensor of order i*. This tensor has rank $i+1$. From now on, we will omit the tensor sign.

Problem 2.1 Show[1] that in the Fourier domain Eq. (2.1) takes the form

$$\begin{aligned}\vec{P}_F(\omega) \;=\;& \epsilon_0\chi^{(1)}(\omega)E_F(\omega) \\ &+\epsilon_0 \iint\limits_{-\infty}^{+\infty} \chi^{(2)}(\omega_1,\omega_2)\delta(\omega-\omega_1-\omega_2)\vec{E}_F(\omega_1)\vec{E}_F(\omega_2)\mathrm{d}\omega_1\mathrm{d}\omega_2 \\ &+\epsilon_0 \iiint\limits_{-\infty}^{+\infty} \chi^{(3)}(\omega_1,\omega_2,\omega_3)\delta(\omega-\omega_1-\omega_2-\omega_3)\vec{E}_F(\omega_1)\vec{E}_F(\omega_2)\vec{E}_F(\omega_3)\mathrm{d}\omega_1\mathrm{d}\omega_2\mathrm{d}\omega_3 \\ &+\ldots,\end{aligned} \tag{2.2}$$

where subscript F indicates the Fourier transform defined similarly to Eq. (1.14).

Note 2.1 Because $E_F(-\omega) = E_F^*(\omega)$, the frequency-to-time Fourier transform

$$\vec{E}(t) = \int_{-\infty}^{+\infty} E_F(\omega)e^{-i\omega t}\mathrm{d}\omega \tag{2.3}$$

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[1] For full credit, it is enough to perform the calculation only for the second term.

can be written as

$$\vec{E}(t) = \int\limits_0^{+\infty} \left[E_F(\omega)e^{-i\omega t} + E_F^*(\omega)e^{i\omega t}\right] \mathrm{d}\omega. \tag{2.4}$$

This notation is commonly used, along with the convention that $E_F(\omega)$ has only positive frequency components. The same is true for the polarization $\vec{P}(t)$. The nonlinear susceptibility, on the other hand, is a function of many variables and cannot be written in the form (2.4). It may contain negative frequency components.

Note 2.2 Because of the presence of delta functions in Eq. (2.2), the frequency-domain susceptibilities are sometimes written as, for example,

$$\chi(\omega_1, \omega_2) \equiv \chi(\omega, \omega_1, \omega_2) \equiv \chi(\omega = \omega_1 + \omega_2). \tag{2.5}$$

Problem 2.2 Verify that the dimension of $\chi^{(1)}(\omega)$ is unity; the dimension of $\chi^{(2)}(\omega)$ is m/V; the dimension of $\chi^{(2)}(\omega)$ is m^2/V^2.

Problem 2.3 Suppose the field has only two components:

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c. \tag{2.6}$$

Calculate $P(t)$ due to the second-order nonlinearity. For simplicity, work in one dimension.
Answer:

$$\begin{aligned} P^{(2)}(t) \quad = \quad & \epsilon_0 \Big[\underbrace{\chi^{(2)}(\omega_1, \omega_1)E_1^2 e^{-2i\omega_1 t} + \chi^{(2)}(\omega_2, \omega_2)E_2^2 e^{-2i\omega_2 t}}_{\text{second-harmonic generation (SHG)}} \\ & + \underbrace{2\chi^{(2)}(\omega_1, \omega_2)E_1 E_2 e^{-i(\omega_1+\omega_2)t}}_{\text{sum-frequency generation (SFG)}} + \underbrace{2\chi^{(2)}(\omega_1, -\omega_2)E_1 E_2^* e^{-i(\omega_1-\omega_2)t}}_{\text{difference-frequency generation (DFG)}} \\ & + \underbrace{\chi^{(2)}(\omega_1, -\omega_1)|E_1|^2 + \chi^{(2)}(\omega_2, -\omega_2)|E_2|^2}_{\text{optical rectification (OR)}} + c.c. \Big] \end{aligned} \tag{2.7}$$

Note 2.3 Notice the factor of 2 in the SFG and DFG terms that is absent in SHG. It appears that the nonlinear effect of two slightly non-degenerate fields is different, by a factor of two, from the nonlinear effect of two identical fields. Is this correct?

Problem 2.4 Repeat Problem 2.3 for the third-order nonlinearity with three fields.

2.2 Symmetries

Problem 2.5 Consider the framework of the classical theory of dispersion (Sec. 1.4) with unharmonic spring, whose potential energy is given by

$$U(x) = \frac{1}{2}\kappa x^2 + \frac{1}{3}max^3 + \frac{1}{4}mbx^4. \tag{2.8}$$

Calculate the (a) second- and (b) third- order nonlinear susceptibility due to the unharmonicity. Assume that the nonlinearities are small.
Hint [part (a)]: Assume that two fields are applied akin to Problem 2.3. Write the equation of motion:

$$\ddot{x} = -\omega_0^2 x - \gamma\dot{x} - ax^2 + Ee/m \tag{2.9}$$

(where we neglect the b-term responsible for the third-order nonlinearity). First calculate $x(t)$ neglecting the nonlinear term. Then substitute this solution into (2.9) and solve this equation again,

for example, for the sum frequency component of the oscillation spectrum.
Answer:

$$\chi^{(2)}(\omega_1,\omega_2) = \frac{Ne^3a}{\epsilon_0 m^2}\frac{1}{D(\omega_1+\omega_2)D(\omega_1)D(\omega_2)}; \tag{2.10}$$

$$\chi^{(3)}(\omega_1,\omega_2,\omega_3) = \frac{Ne^4b}{\epsilon_0 m^3}\frac{1}{D(\omega_1+\omega_2)D(\omega_1)D(\omega_2)D(\omega_3)}, \tag{2.11}$$

where

$$D(\omega) = \omega_0^2 - \omega^2 - i\omega\gamma. \tag{2.12}$$

Note 2.4 Based on the previous calculation, we obtain *Miller's rule*: the quantity

$$\frac{\chi^{(2)}(\omega_1,\omega_2)}{\chi^{(1)}(\omega_1+\omega_2)\chi^{(1)}(\omega_1)\chi^{(1)}(\omega_2)}$$

is frequency-independent.

Problem 2.6 Estimate the order of magnitude of the first-, second- and third-order nonlinear susceptibilities in a crystal assuming that the anharmonicity of the potential becomes significant when the position x of the electron is on a scale of the lattice constant d: $kd \sim mad^2 \sim mbd^3$. Assume that all frequencies are in the optical range, but the optical fields are far away from the resonance at ω_0.
Answer:

$$\begin{aligned}\chi^{(1)} &\sim \frac{e^2}{\epsilon_0 d^3 m\omega^2} \sim 1 \\ \chi^{(2)} &\sim \frac{e^3}{\epsilon_0 d^4 m^2\omega^4} \sim 10^{-12}\mathrm{m/V} \\ \chi^{(3)} &\sim \frac{e^4}{\epsilon_0 d^5 m^3\omega^6} \sim 10^{-24}\mathrm{m^2/V^2}\end{aligned} \tag{2.13}$$

Problem 2.7 Show that, far away from the resonance (lossless media, see Note 1.8), using the model of Problem 2.5,

$$\chi^{(2)}(\omega_3=\omega_1+\omega_2) = \chi^{(2)}(\omega_1=\omega_3-\omega_2) = \chi^{(2)}(\omega_2=\omega_3-\omega_1). \tag{2.14}$$

Note 2.5 A more careful calculation taking into account tensor nature of the susceptibilities shows that

$$\chi^{(2)}_{kij}(\omega_3=\omega_1+\omega_2) = \chi^{(2)}_{ikj}(\omega_1=\omega_3-\omega_2) = \chi^{(2)}_{jki}(\omega_2=\omega_3-\omega_1) \tag{2.15}$$

(where i,j,k are spatial coordinates), i.e. the tensor indices must be permuted together with the frequencies. A similar result is valid for the third-order nonlinearity.

An even more powerful symmetry is obtained under the assumption that the susceptibilities are frequency independent. In this case, any permutation of indices is allowed, for example:

$$\chi^{(2)}_{xyz} = \chi^{(2)}_{xzy} = \chi^{(2)}_{yxz} = \chi^{(2)}_{yzx} = \chi^{(2)}_{zxy} = \chi^{(2)}_{zyx},$$

or

$$\chi^{(2)}_{xxz} = \chi^{(2)}_{xzx} = \chi^{(2)}_{zxx}.$$

This result is called *Kleinman's symmetry*; it is also valid for the third-order nonlinearity. Note that Kleinman's symmetry does not mean that, for example, $\chi^{(2)}_{xxz} = \chi^{(2)}_{xxy}$.

Problem 2.8 Show that there are only 10 independent tensor elements of $\chi^{(2)}$ under Kleinman's symmetry.

Problem 2.9 Show that the second-order nonlinear susceptibility in any medium with mirror symmetry completely vanishes.

Note 2.6 This result demonstrates that second-order nonlinear effects are impossible in liquids or gases unless the symmetry is broken, for example, by an external field.

Problem 2.10 Materials without mirror symmetry are called *chiral.*

a) Can an isotropic medium (e.g. liquid or a gas) be chiral?

b) What tensor elements of the second-order susceptibility may not vanish in an isotropic chiral medium?

Problem 2.11 Show that in an isotropic non-chiral medium, the only nonvanishing elements of the third order nonlinear susceptibility are χ_{iijj}, $\chi^{(3)}_{ijij}$, and $\chi^{(3)}_{ijji}$. Show that each of these elements is the same for all i and j as long as $i \neq j$ (i.e., for example, $\chi^{(3)}_{xxyy} = \chi^{(3)}_{yyzz}$). Show that

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyxy} + \chi^{(3)}_{xyyx}.$$

Note 2.7 An alternative notation[2] for the second-order susceptibility with Kleinman's symmetry is tensor d_{il} with

$$d_{il} = \frac{1}{2}\chi^{(2)}_{ijk}$$

and the pair of indices jk map onto a single index l as follows:

xx	yy	zz	$yz = zy$	$xz = zx$	$xy = yx$
1	2	3	4	5	6

For example, $d_{12} = \frac{1}{2}\chi^{(2)}_{xyy}$, $d_{36} = \frac{1}{2}\chi^{(2)}_{zyx}$, etc.

Problem 2.12 Which of the d's are equal to each other under Kleinman's symmetry?

Note 2.8 The values of d's for most important nonlinear crystals, defined with respect to crystallographic axes, are tabulated and can be found, for example, in the *Handbook of nonlinear optical crystals* together with the recipe to determine the effective nonlinearity (see below). Due to external symmetries, many crystals have fewer independent d's than 10.

2.3 Frequency conversion

Note 2.9 Consider the sum frequency term in Eq. (2.7):

$$\vec{P}^{(2)}(t) = [2\vec{\chi}^{(2)}(\omega_1,\omega_2)\vec{E}_1\vec{E}_2 e^{-i(\omega_1+\omega_2)t} + c.c] \tag{2.16}$$

If we are interested only in the amplitude of the polarization along a particular direction, we can rewrite the above as

$$P^{(2)}(t) = \epsilon_0[4d_{\text{eff}}(\omega_1,\omega_2)E_1E_2e^{-i(\omega_1+\omega_2)t} + c.c] \tag{2.17}$$

where the information about directions is accumulated in

$$d_{\text{eff}} = \frac{1}{2}\sum_{ijk}\chi^{(2)}_{ijk}\frac{P^{(2)}_i(t)}{|\vec{P}^{(2)}(t)|}\frac{(E_1)_j}{|\vec{E}_1|}\frac{(E_2)_k}{|\vec{E}_2|}, \tag{2.18}$$

the *effective nonlinearity* of the material associated with the given set of directions for the input fields and nonlinear polarization. The definition of effective nonlinearity can be straightforwardly extended to other nonlinear effects.

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[2] This convention does not appear very logical. First, the factor of 1/2 does not reflect any physics. Second, there are 3 × 6 = 18 d's while the number of independent χ's is only 10.

Problem 2.13 Derive *the coupled-wave equations for sum-frequency generation* $(1+2 \to 3)$ in the continuous-wave regime:

$$\partial_z \mathcal{E}_1 = \frac{2id_{\text{eff}}\omega_1}{n_1 c}\mathcal{E}_3\mathcal{E}_2^* e^{-i\Delta kz}, \tag{2.19}$$

$$\partial_z \mathcal{E}_2 = \frac{2id_{\text{eff}}\omega_2}{n_2 c}\mathcal{E}_3\mathcal{E}_1^* e^{-i\Delta kz}, \tag{2.20}$$

$$\partial_z \mathcal{E}_3 = \frac{2id_{\text{eff}}\omega_3}{n_3 c}\mathcal{E}_1\mathcal{E}_2 e^{i\Delta kz}, \tag{2.21}$$

where $\delta_k = k_1 + k_2 - k_3$ is the *phase mismatch*, $\mathcal{E}$'s are the time-independent slowly-varying envelope amplitudes (see Sec. 1.2).

Problem 2.14 The process of sum-frequency generation can be visualized at the quantum level as conversion of two photons from waves 1 and 2 into a single photon of wave 3. From this visualisation, we obtain the *Manley-Rowe relations*

$$\frac{1}{\omega_1}\partial_z I_1 = \frac{1}{\omega_2}\partial_z I_2 = -\frac{1}{\omega_3}\partial_z I_3. \tag{2.22}$$

Derive these relations from the classical coupled-wave equations for sum-frequency generation (2.19)–(2.21). **Hint:** $I_i = 2n_i\epsilon_0 c|\mathcal{E}_i|^2$.

Problem 2.15 Waves 1 and 2 with intensities I_1 and I_2, respectively, are incident on the front face $(z = 0)$ of a nonlinear crystal with effective nonlinearity d_{eff} and generate a sum-frequency wave. Solve the coupled-wave equations to find the dependencies of I_1 and the sum-frequency intensity I_3 on the propagation distance z. Assume that $I_2 \gg I_1$ so wave 2 does not get substantially depleted in the process.

a) Assume that the propagation distance is sufficiently short so wave 1 does not get depleted, either.

b) Assume perfect phase matching $(\Delta k = 0)$ instead.

c) Do not make any of the above assumptions.
Answer:

$$I_1(z) = I_1(0)\left[\frac{\Delta k^2}{4g^2} + \frac{1}{L_{\text{NL}}^2}\frac{\cos^2 gz}{g^2}\right]; \tag{2.23}$$

$$I_3(z) = I_1(0)\frac{\omega_3}{\omega_1}\frac{1}{L_{\text{NL}}^2}\frac{\sin^2 gz}{g^2}, \tag{2.24}$$

$$\tag{2.25}$$

where

$$L_{\text{NL}} = \sqrt{\frac{\epsilon_0 c^3 n_1 n_2 n_3}{2\omega_1\omega_3 I_2 d_{\text{eff}}^2}} \tag{2.26}$$

is the characteristic *nonlinear length* over which the energy is converted from wave 1 to wave 3 (Fig. 2.1) and

$$g = \sqrt{\frac{1}{L_{\text{NL}}^2} + \frac{\Delta k^2}{4}}. \tag{2.27}$$

Problem 2.16 A wave with intensity I_1 and frequency ω is incident on the front face $(z = 0)$ of a nonlinear crystal with effective nonlinearity d_{eff} and generates a second-harmonic wave. Write the coupled-wave equations and find the behavior of I_1 and the second-harmonic intensity I_2 as functions of z.

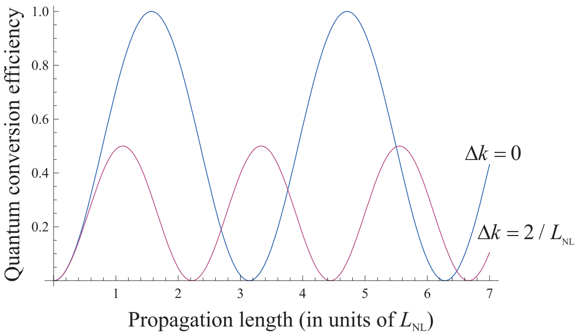

Figure 2.1: SFG conversion efficiency as a function of the propagation distance (Problem 2.15).

a) Assume that the propagation distance is sufficiently short so wave 1 does not get depleted.
Answer [Fig. 2.2(a)]:

$$I_2(z) = I_1 \frac{z^2}{L_{\text{NL}}^2} \operatorname{sinc}^2 \frac{\Delta k z}{2} \tag{2.28}$$

where

$$L_{\text{NL}} = \sqrt{\frac{\epsilon_0 c^3 n_1^2 n_2}{2\omega^2 I_1 d_{\text{eff}}^2}}. \tag{2.29}$$

b)* Assume perfect phase matching ($\Delta k = 0$) instead.
Answer [Fig. 2.2(b)]:

$$I_2(z) = I_1 \tanh^2 \frac{z^2}{L_{\text{NL}}^2} \tag{2.30}$$

c)* Do not make any of the above assumptions.
Answer [Fig. 2.2(b)]:

$$I_2(z) = I_1 \kappa \operatorname{sn}^2(\kappa^{-1/2} z / L_{\text{NL}}, \kappa) \tag{2.31}$$

where $\operatorname{sn}(\cdot,\cdot)$ is Jacobi's elliptic sine and

$$\kappa = \left[\sqrt{1 + \frac{\Delta k^2}{16 L_{\text{NL}}^2}} - \frac{\Delta k}{4 L_{\text{NL}}} \right]^2 . \tag{2.32}$$

Note 2.10 As evident from Eqs. (2.28) and (2.29), the second harmonic intensity is proportional to the square of the fundamental intensity. For this reason, a pulsed laser will produce more second harmonic than a continuous-wave laser of a similar power. SHG power can also be enhanced by focusing a laser into the crystal.

Problem 2.17 Focussing the laser beam into the crystal too tightly will result in strong angular divergence due to diffraction, so high intensity is present only over a short length (Fig. 2.3). Estimate the beam waist radius w_0 inside the crystal which will optimize the SHG. Estimate the generated second harmonic power. Use the parameters from Problem 2.16; the crystal length is $L \ll L_{NL}$. Neglect any phase mismatch.

Note 2.11 The exact solution to this problem has been found in 1968 by G. D. Boyd and D. A. Kleinman. The focusing is optimized when L/kw_0^2=2.84 (where w_0 is the $1/e^2$ radius of the waist).

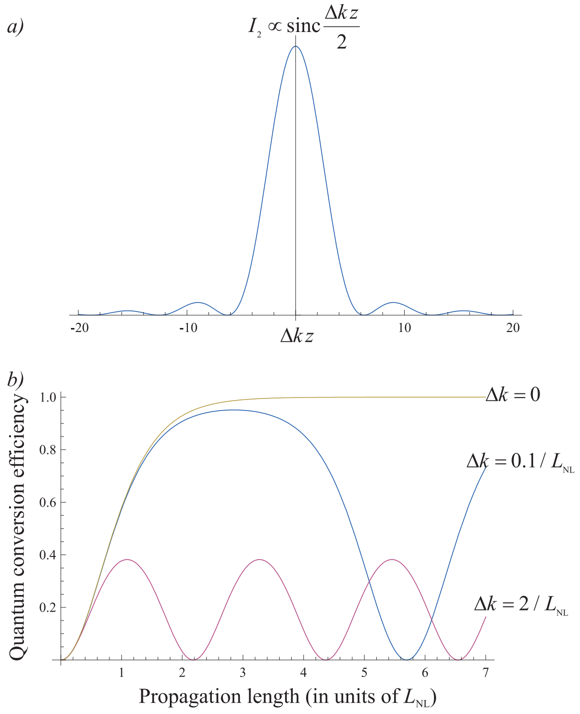

Figure 2.2: SHG conversion efficiency: (a) as a function of the phase mismatch for small propagation distances [Problem 2.16(a)]; (b) as a function of the propagation distance for various values of the phase mismatch [Problem 2.16(b,c)].

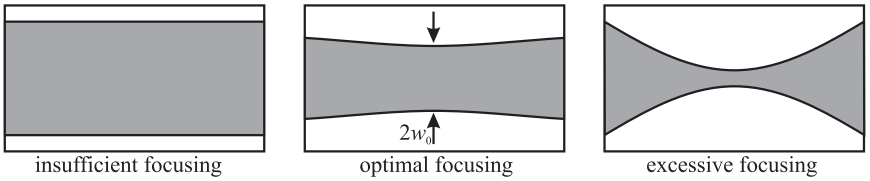

Figure 2.3: Focusing a laser beam into the crystal (Problem 2.17).

2.4 Linear optics in crystals

It follows from linear algebra that the first-order susceptibility tensor can be diagonalized by reorientation of the reference frame[3].

$$\begin{pmatrix} \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{zx} & \chi_{zy} & \chi_{zz} \end{pmatrix} \rightarrow \begin{pmatrix} \chi_{xx} & 0 & 0 \\ 0 & \chi_{yy} & 0 \\ 0 & 0 & \chi_{zz} \end{pmatrix} \tag{2.33}$$

We will be working in the reference frame in which χ is diagonalized. The coordinate axes are then called *dielectric* or axes of the crystal[4]. The planes xy, xz and yz are called *principal planes.* With each χ_{ii}, we associate a *principal value of the refractive index* $n_i = \sqrt{1 + \chi_{ii}}$.

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[3] In this section, we talk only about $\chi^{(1)}$. The supersctipt (1) will be omitted. The crystal is assumed non-magnetic.

[4] The dielectric (crystallophysical) axes coincide with the crystallographic axes, but are denoted differently. Crystallographic axes are denoted a, b, c, and dielectric x, y, z. The conventional assignment of axes may not be straightforward, e. g. for LBO it is $abc \rightarrow xzy$. In the literature, the table in Note 2.7 may refer to either crystallographic or dielectric axes. In this course, however, we will always define objects with respect to the dielectric axes, unless otherwise indicated.

Problem 2.18 Consider a wave of frequency ω propagating in the principal plane yz with the k vector oriented at angle θ with respect to the z axis. The wave is polarized so that its electric field vector is also in the yz plane.

a) Show that the index of refraction for this wave is given by

$$\frac{1}{n(\theta)^2} = \frac{\sin^2\theta}{n_z^2} + \frac{\cos^2\theta}{n_y^2} \tag{2.34}$$

b) Show that the Poynting vector $\vec{S} = \frac{1}{\mu_0}\vec{E}\times\vec{B}$ is not parallel to $\vec{k}$, but oriented at angle

$$\tan\theta' = \frac{n_y^2}{n_z^2}\tan\theta \tag{2.35}$$

with respect to the z axis.

Hint: Because the susceptibility is non-scalar, we have $\vec{P} \nparallel \vec{E}$ and thus $\vec{D} \nparallel \vec{E}$. Therefore you have to start from Maxwell's equations and determine the magnitude of k and the direction of $\vec{E}_0$ under which the wave $\vec{E}_0 e^{i\vec{k}\vec{r}-i\omega t}$ satisfies these equations. It is convenient to solve the problem in the vector form. Notice that because of Eq. (1.1) we must have $\vec{D} \perp \vec{k}$. Therefore, if $\vec{k} = \begin{pmatrix} 0 \\ k\sin\theta \\ k\cos\theta \end{pmatrix}$, then $\vec{D}_0 = \begin{pmatrix} 0 \\ D_0\cos\theta \\ -D_0\sin\theta \end{pmatrix}$ and thus $\vec{E}_0 = \frac{1}{\epsilon_0}\begin{pmatrix} 0 \\ D_0\cos\theta/n_y^2 \\ -D_0\sin\theta/n_z^2 \end{pmatrix}$.

Note 2.12 The angle between $\vec{k}$ and $\vec{S}$ is called the *walk-off angle*. The walk-off effect is the principle of operation of many optical devices, such as calcite beam displacers (`http://www.thorlabs.com/newgrouppage9.cfm?objectgroup_id=745`).

Problem 2.19 Show that, if the wave in Problem 2.18 is polarized orthogonally to the yz plane, it behaves as a regular electromagnetic wave in an isotropic medium, i.e. $\vec{E} \perp \vec{k}$ and there is no walk-off. Find the index of refraction for this wave.

Note 2.13 If the wave enters the crystal with its polarization outside one of the principal planes, it will experience birefringence, i.e. its polarization vector will rotate as the wave propagates. In other words, no expression of the form $\vec{E}_0 e^{i\vec{k}\vec{r}-i\omega t}$ will satisfy Maxwell's equations.

Note 2.14 Dependent on the lattice symmetry, there can be three possibilities for n_x, n_y, and n_z.

- All three refractive indices are equal (e.g. in a cubic lattice), leading to *isotropic* optical properties.
- Only two refractive indices are equal. Crystals with such structure are called *uniaxial*. Examples: trigonal, tetragonal, hexagonal lattices.
- All three are unequal — *biaxial* crystals. Examples: triclinic, monoclinic, orthorhombic.

Note 2.15 If there exists a direction such that a wave of arbitrary polarization, propagating in the crystal along this direction, does not experience birefringence, this direction is called the *optical axis* of the crystal. The optical axis may not coincide with any of the crystallographic axes.

Note 2.16 In a uniaxial crystal, the dielectric axes are typically defined so that $n_x = n_y$. The associated refractive index is called *ordinary* (n_O) while the refractive index n_z is called *extraordinary* (n_E). In $n_E > n_O$, the crystal is called *positive*, otherwise *negative*. The refractive index behaves as shown in Fig. 2.4; there is only one optical axis, which is the z axis.

Any wave propagating through the crystal can be decomposed into two polarization components: *ordinary* (electric field perpendicular to the optical axis) and *extraordinary* (orthogonal to ordinary).

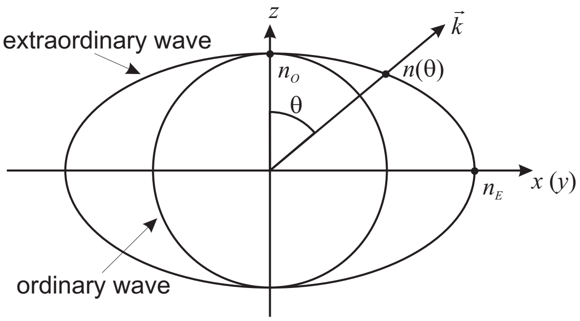

Figure 2.4: Dependence of the refractive index on the propagation direction in a positive uniaxial crystal.

Problem 2.20 Show that the index of refraction for the ordinary wave is n_O whereas for the extraordinary wave it is given by Eq. (2.34).

Note 2.17 We see that the index of refraction for the extraordinary wave varies with θ and does not always equal n_E. Thus the terms "extraordinary index of refraction" and "index of refraction for the extraordinary wave" mean different things. However, in some literature they are used interchangeably, which may create confusion.

Problem 2.21 Consider a biaxial crystal with $n_x < n_y < n_z$. For each principal plane, make a plot similar to Fig. 2.4. Verify that there are two optical axes. Show that the angle between each optical axis and the z axis is given by

$$\sin\theta = \frac{1/n_x^2 - 1/n_y^2}{1/n_x^2 - 1/n_z^2}. \tag{2.36}$$

Note 2.18 The dielectric axes of biaxial crystals are conventionally defined in such way that either $n_x < n_y < n_z$ or $n_z < n_y < n_x$ so the optical axes are always in the xz plane (their azimuthal angle $\varphi = 0$).

Note 2.19 In a biaxial crystal, the terms "ordinary" and "extraordinary" apply to waves propagating in one of the principal planes. The electric field vector of the extraordinary wave is in the same principal plane as $\vec{k}$, and it behaves as discussed in Problem 2.18. The ordinary wave is polarized orthogonally to this plane and behaves in accordance with Problem 2.19.

2.5 Phase matching

As we saw in Sec. 2.3, the frequency conversion is maximized when the phase mismatch vanishes. In application, for example, to SHG, this means $k_2 = 2k_1$. Given that $k_i = n_i\omega_i/c$ and $\omega_2 = 2\omega_1$, we see that phase matching condition for SHG implies that $n_2 = n_1$. This condition is not satisfied automatically, because the refraction index depends on the wavelength. However, it may be possible that for a certain propagation direction the refractive indices match for one of the waves being ordinary, and the other extraordinary. This technique is called *critical phase matching.* The crystal must be manufactured so that its optical surfaces are roughly perpendicular to the intended propagation direction consistent with the chosen phase matching configuration.

Problem 2.22 You need to generate the second harmonic of laser light at wavelength $\lambda = 1064$ nm in a beta barium borate (BBO) crystal. This is a uniaxial crystal with $n_O = 1.6551$, $n_E = 1.5425$ at 1064 nm and $n'_O = 1.6749$, $n'_E = 1.5555$ at 532 nm.

a) At which angle to the optical axis should the beam propagate to achieve phase matching?

b) Which wave (fundamental or second harmonic) is ordinary, which one is extraordinary?

c) Find the walk-off angle for both waves.

Note 2.20 For given a crystal cut for optimal phase matching, the following parameters can be defined.

- *Group velocity mismatch* GVM $= 1/v_{gr,1} - 1/v_{gr,2}$. This parameter is important when the laser is pulsed. For nonzero GVM, the generated second harmonic pulse will overtake (or fall behind) the fundamental, potentially resulting in a longer second harmonic pulse width.
- *Phase matching bandwidth*, i.e. the range of wavelengths over which the phase-matching condition holds for a constant propagation angle. It is normally defined in terms of the FWHM[5] intensity range, within which
$$\operatorname{sinc}^2\frac{\Delta kL}{2} \geq \frac{1}{2} \tag{2.37}$$
[*cf.* Eq. (2.28)].
- *Acceptance angle*, i.e. the range of angles over which the phase-matching condition holds for a constant wavelength. It is also defined in accordance with Eq. (2.37).

Problem 2.23 Show that the phase-matching bandwidth and the group velocity mismatch are related according to
$$\Delta\lambda \approx 0.88\frac{\lambda^2}{cL}\frac{1}{\text{GVM}}. \tag{2.38}$$

Note 2.21 The dependence of the refractive index on the wavelength (sometimes also on the temperature), is sometimes written in the form of empirical *Sellmeyer equations.* For example, for BBO the Sellmeyer equations are as follows:

$$n_O^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2 \tag{2.39}$$
$$n_E^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2. \tag{2.40}$$

Problem 2.24 Derive the general expression for the acceptance angle for SHG in the $2O \to E$ configuration in a uniaxial crystal. The ordinary and extraordinary refractive indices are known, as well as the phase matching angle θ.
Hint: Use Eq. (2.34).

Problem 2.25 For SHG in a BBO crystal (see Problem 2.22) of length $L = 5$ mm, perform the following calculations.

a) Find the group velocity mismatch.

b) Find the FWHM of the phase matching band (in units of wavelength).

c) Find the acceptance angle.

d) Find the walk-off angle for both beams.

e) The power of the fundamental beam is $P = 100$ mW. Estimate the power of generated second harmonic assuming optimal beam geometry if $d_{\text{eff}} = 1.4$ pm/V.

Problem 2.26 The optimal beam configuration according to Boyd and Kleinman (Note 2.11) implies that the beam should be focused into the crystal with a certain angular aperture, which is proportional to $L^{-1/2}$ (check this!). On the other hand, the phase-matching acceptance angle is proportional to L^{-1}. Therefore, for the crystal length exceeding a certain critical value, the focusing strength will be limited by phase matching rather than the optimal intensity. Estimate this critical length for the SHG in BBO in the configuration of Problem 2.22.

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[5] FWHM = full width at half-maximum

Note 2.22 Second-order nonlinear processes in which the two lower energy waves have the same polarization (e.g. both ordinary or both extraordinary) are called *Type I* processes. If the polarizations are orthogonal, the nonlinear process is *Type II*.

Problem 2.27 Lithium triborate (LBO) is a biaxial crystal with the following parameters:

λ, μm	n_x	n_y	n_z
0.8	1.570	1.596	1.611
0.4	1.590	1.619	1.636

You need to generate the second harmonic of laser light at wavelength $\lambda = 800$ nm. Analyze all three principal planes of this crystal and determine all possible Type I phase matching configurations in terms of polarization combinations and beam angles (θ and φ).

Note 2.23 *(Spontaneous) parametric down-conversion (PDC or SPDC)* is the second-order nonlinear process in which photons comprising a *pump* laser beam propagating through a nonlinear crystal split into pairs of photons of lower energy. The quantum nature of PDC is inverse to that of SFG, but unlike SFG, PDC is a pure quantum process; it has no classical explanation. The phase-matching requirements for PDC are analogous to SFG: we must have, at the same time, $\omega_3 = \omega_1 + \omega_2$ and $\vec{k}_3 = \vec{k}_1 + \vec{k}_2$. If $\omega_1 = \omega_2$ or $\vec{k}_1 = \vec{k}_2$, PDC is called, respectively, *spectrally* or *spatially degenerate*.

Problem 2.28 You need to implement parametric down-conversion in frequency-degenerate, but spatially non-degenerate type I $e \to o + o$ configuration with a pump wavelength of 532 nm in a BBO crystal. The three waves are located in the same meridional plane (same φ), but propagate at different polar angles ($\theta_1 < \theta_3 < \theta_2$). The generated photons must be emitted at angle $\theta_2 - \theta_1 = 6^\circ$. Find the polar angle θ_3 of the pump wave vector which enables phase matching for such a process.

Note 2.24 Because the ordinary index of refraction does not change significantly around the direction of the pump, the photons will be emitted not only inside the meridional plane, but along the surface of the *cone* with the axis along the pump wavevector and a 6° apex angle. In each pair, the two photons will be emitted along diametrally opposite lines on the side surface of the cone.

Problem 2.29 Solve the coupled-wave equation (2.21) for SFG in the nondepleted regime in a *periodically poled* nonlinear material, in which the orientation of the $\chi^{(2)}$ tensor is periodically inverted with the *poling period*

$$\Lambda = \frac{2\pi}{\Delta k} \tag{2.41}$$

and a 50% duty cycle. Plot the sum-frequency intensity $I_3(z)$ as a function of the propagation distance for $0 \le z < 4\Lambda$. In the same graph, also plot[6] $I_3(z)$ for (1) the same material with the same Δk, but without periodic poling and (2) the same material, without periodic poling, with $\Delta k = 0$. Show that on average $I_3(z)$ behaves similarly to that in a non-poled material with $\Delta k = 0$ and $d'_{\text{eff}} = \frac{2}{\pi} d_{\text{eff}}$.

Note 2.25 The technique described in Problem 2.29 is called *quasi phase matching (QPM)*. It has many advantages compared to critical phase matching:

- polarizations and direction of the waves can be chosen to utilize the highest tensor element of $\chi^{(2)}$ in the given crystal;
- QPM allows generation of almost any wavelength by choosing the proper combination of the reversal period and crystal temperature;
- weaker dependence on the orientation greatly increases the phase matching bandwidth and the acceptance angle, which permits stronger focusing (*cf.* Problem 2.26).

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[6] For full credit, your picture should correctly show the comparative behavior of the three functions around $z = 0$.

Periodic poling is implemented by applying spatially periodic high voltage to a crystal for several hours, which leads to reorientation of the crystal domains. This technology has so far been developed only for a few nonlinear materials and there are limitations on the thickness of the crystals that can be manufactured, which limits their application. Furthermore, periodically poled crystals are substantially more expensive than regular ones.

Problem 2.30 You need to generate second harmonic of continuous-wave laser light at wavelength $\lambda = 800$ nm in a periodically poled potassium titanyl phosphate (PPKTP) crystal of length $L = 5$ mm. Light propagates along the x axis and is polarized along the z axis. Find the necessary information about the crystal on the Internet and answer the following questions.

a) What is the poling period required for phase matching?

b) Find the FWHM of the phase matching band. **Hint:** Eq. (2.38) cannot be used here.

c) Find the acceptance angle.

d) The power of the fundamental beam is $P = 100$ mW. Find the power of generated second harmonic assuming optimal beam geometry.

Problem 2.31 You need to implement parametric down-conversion in PPKTP in the Type II spectrally and spatially degenerate configuration. The pump light at $\lambda = 400$ nm propagates along the x axis and is polarized along the y axis. Find the necessary poling period.