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Theory of Nonlinear Optics

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1 Phenomenological representation of non-resonant nonlinearities

1.1 The polarization as the origin of nonlinearity in optics

We start with MAXWELL's equations in the optical domain without external charges, currents and no magnetization

$$\vec{\nabla} \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t} \qquad \vec{\nabla} \cdot \boldsymbol{D} = 0 \tag{1.1}$$

$$\vec{\nabla} \times \boldsymbol{B} = \mu_0 \frac{\partial \boldsymbol{D}}{\partial t} \qquad \vec{\nabla} \cdot \boldsymbol{B} = 0, \tag{1.2}$$

where $\boldsymbol{E}$ ist the electric field, $\boldsymbol{B}$ the magnetic induction and $\boldsymbol{D}$ the dielectric displacement which is defined as

$$\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}, \tag{1.3}$$

consisting of a vacuum contribution $\varepsilon_0 \boldsymbol{E}$ and the polarization $\boldsymbol{P}$ of the material. We note that MAXWELL's equations are linear in all components.

The polarization of the material $\boldsymbol{P}$ is the source of nonlinearities and is driven by the electromagnetic fields. It cannot be deduced from electrodynamics, but we need quantum mechanics/solid state physics. Furthermore it has to obey symmetries which will be discussed in this chapter.

We define the polarization $\boldsymbol{P}(\boldsymbol{r}, t)$ at a fixed point in space $\boldsymbol{r}$ and time t. In principle it depends on electromagnetic fields acting on all other places $\boldsymbol{r}'$ and other times t' as $\boldsymbol{P}(\boldsymbol{r}, t) = \boldsymbol{P}[\boldsymbol{E}(\boldsymbol{r}', t'), \boldsymbol{B}(\boldsymbol{r}', t')]$. However, we make several assumptions to simplify our calculations:

- There are only local interactions: $\boldsymbol{r} = \boldsymbol{r}'$. We explicitly exclude the propagation of a polarization in the material. This means the polarization of one unit of the material is influence by light acting on just that unit only. The reason for that is that nonlocal effects on atomic scales take place at $\sim 10^{-10}\,\mathrm{m}$ while the variations of the optical field are $\sim 10^{-6}\,\mathrm{m}$.
- *Causality*: The polarization cannot depend on fields from the future $t' \leq t$.
- No external action on the polarization within the memory time of the material. The polarization depends on time differences only $\boldsymbol{P}(\boldsymbol{r}, t) = \boldsymbol{P}[\boldsymbol{E}(\boldsymbol{r}, t-\tau), \boldsymbol{B}(\boldsymbol{r}, t-\tau)]$ with $\tau > 0$.
- No magnetic effects $\boldsymbol{P}(\boldsymbol{r}, t) = \boldsymbol{P}[\boldsymbol{E}(\boldsymbol{r}, t-\tau)]$

The above assumptions are well justified for conventional materials. Unfortunately, a further simplification is not straightforward.

1.2 The Taylor expansion of the polarization

The general idea ist that for non-resonant nonlinearities the interaction between photons via the material is *weak*. This implies that only a very limited number of photons hits one atom of the material within the relaxation time/memory of the polarization. We can classify the polarization with respect to the number of interacting photons

$$\boldsymbol{P}(t) = \boldsymbol{P}^{(1)}(t) + \boldsymbol{P}^{(2)}(t) + \boldsymbol{P}^{(3)}(t). \tag{1.4}$$

The linear polarization $\boldsymbol{P}^{(1)}(t)$ is induced by a single photon and is by far the dominant polarization. $\boldsymbol{P}^{(2)}(t)$ is the nonlinear polarization of quadratic/second order and is induced by two-photon processes.

a.) Linear polarization

We start by only considering one vector component $P_i^{(1)}(t)$ which is driven by all other vector components of the electric field E_j having interacted with the atom at all times $t-\tau_1$ in the past

$$P_i^{(1)}(t) = \varepsilon_0 \sum_{j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \, R_{ij}^{(1)}(\tau_1) E_j(t-\tau_1), \tag{1.5}$$

where $R_{ij}^{(1)}(\tau_1)$ is the linear response function. It is a time dependent second rank tensor which is decaying rapidly in time.

b.) Nonlinear polarization of second order

We can apply the previous procedure to higher order interactions in the same way. Now we assume that two photons have interacted at times $t-\tau_1$ and $t-\tau_2$ in the past with the atom. They both have to be taken into account leading to

$$P_i^{(2)}(t) = \varepsilon_0 \sum_{j,k=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2 \, R_{ijk}^{(2)}(\tau_1,\tau_2) E_j(t-\tau_1) E_k(t-\tau_2). \tag{1.6}$$

The response function $R_{ijk}^{(2)}$ is now a third rank tensor which depends on two times. In a similar way we can represent all higher order interactions $R^{(n)}$.

The response function $R^{(n)}$ has some special properties:

- It is real valued, because we have only considered real valued fields.
- It must obey causality corresponding $R^{(n)}(\tau_k < 0) = 0$.
- It must be symmetric with respect to an exchange of arguments $R_{i\ldots m\ldots k\ldots}^{(n)}(\ldots\tau_m\ldots\tau_k\ldots) = R_{i\ldots k\ldots m\ldots}^{(n)}(\ldots\tau_k\ldots\tau_m\ldots)$
- Has the the symmetries of the respective material.

Symmetry consideration for inversion symmetric materials

The latter property shall be demonstrated for a quadratic nonlinearity represented by $R^{(2)}_{ijk}(\tau_1,\tau_2)$. We assume the nonlinear material to have inversion symmetry as it is found in many crystals as e. g. Si, NaCl or in all amorphous materials, liquids or gases.

We introduce a formal inversion operator $\hat{\boldsymbol{J}}$ which does not act on material parameters of inversion symmetric materials $\hat{\boldsymbol{J}}R^{(2)}_{ijk}(\tau_1,\tau_2) = R^{(2)}_{ijk}(\tau_1,\tau_2)$ but performs a sign flip of vector components $\hat{\boldsymbol{J}}E_i = -E_i$ and $\hat{\boldsymbol{J}}P_i = -P_i$. We now apply the inversion operator to equation 1.6

$$\begin{aligned}
\hat{\boldsymbol{J}}[P^{(2)}_i(t)] &= \hat{\boldsymbol{J}}\left[\varepsilon_0 \sum_{i,j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2\, R^{(2)}_{ijk}(\tau_1,\tau_2) E_j(t-\tau_1)E_k(t-\tau_2)\right] \\
&= \varepsilon_0 \sum_{i,j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2\, \hat{\boldsymbol{J}}R^{(2)}_{ijk}(\tau_1,\tau_2)\hat{\boldsymbol{J}}E_j(t-\tau_1)\hat{\boldsymbol{J}}E_k(t-\tau_2) \\
-P^{(2)}_i(t) &= \varepsilon_0 \sum_{j,k=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2\, R^{(2)}_{ijk}(\tau_1,\tau_2)(-1)E_j(t-\tau_1)(-1)E_k(t-\tau_2)] \\
-P^{(2)}_i(t) &= P^{(2)}_i(t). \qquad (1.7)
\end{aligned}$$

This implies that $P^{(2)}_i(t)$ must be equal to zero for all times t which can only be fulfilled with $R^{(2)}_{ijk} = 0$. In a similar way we can prove that all *even* order nonlinearities must vanish in centrosymmetric and amorphous materials.

1.3 Polarization in frequency space

In optics we usually deal with monochromatic fields and are more interested in the spectral amplitudes than in the fast temporal evolution. Often it is required to quantify the nonlinear response on certain frequency components of the electric field.

a.) We start with the decomposition of the optical field into its frequency components via Fourier transform which are defined in this lecture as follows

$$\boldsymbol{E}(t) = \int_{-\infty}^{\infty} \mathrm{d}\omega\, \tilde{\boldsymbol{E}}(\omega)\mathrm{e}^{-\mathrm{i}\omega t} \quad \text{and} \quad \tilde{\boldsymbol{E}}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, \boldsymbol{E}(t)\mathrm{e}^{\mathrm{i}\omega t}. \qquad (1.8)$$

The temporal field $\boldsymbol{E}(t)$ is thus a sum of oscillating fields each having a spectral amplitude $\tilde{\boldsymbol{E}}(\omega)$. Note that $\boldsymbol{E}(t)$ is real valued which implies that $\tilde{\boldsymbol{E}}(\omega)$ is symmetric

$$\tilde{\boldsymbol{E}}(-\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, \boldsymbol{E}(t)\mathrm{e}^{-\mathrm{i}\omega t} = \left[\frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, \boldsymbol{E}(t)\mathrm{e}^{\mathrm{i}\omega t}\right]^* = \tilde{\boldsymbol{E}}^*(\omega). \qquad (1.9)$$

This means that the spectral amplitudes at negative frequencies $\tilde{\boldsymbol{E}}(-|\omega|)$ are nonzero, but strictly correlated to their positive counterparts and thus need not be considered separately.

b.) Now we can find an expression for $\tilde{\boldsymbol{P}}^{(1)}(\omega)$ by inserting (1.8) into (1.5)

$$\begin{aligned} P_i^{(1)}(t) &= \varepsilon_0 \sum_{j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \, R_{ij}^{(1)}(\tau_1) E_j(t-\tau_1) \\ &= \varepsilon_0 \sum_{j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \, R_{ij}^{(1)}(\tau_1) \int_{-\infty}^\infty \mathrm{d}\omega \, \tilde{E}_j(\omega) \mathrm{e}^{-\mathrm{i}\omega(t-\tau_1)} \\ &= \int_{-\infty}^\infty \mathrm{d}\omega \, \varepsilon_0 \sum_{j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \, R_{ij}^{(1)}(\tau_1) \mathrm{e}^{\mathrm{i}\omega\tau_1} \tilde{E}_j(\omega) \mathrm{e}^{-\mathrm{i}\omega t} = \int_{-\infty}^\infty \mathrm{d}\omega \, \tilde{P}_i^{(1)}(\omega) \mathrm{e}^{-\mathrm{i}\omega t}. \end{aligned} \tag{1.10}$$

$\tilde{P}_i^{(1)}(\omega)$ is the spectral amplitude of the *linear* polarization. It depends on fields oscillating with the same frequency in a linear way

$$\tilde{P}_i^{(1)}(\omega) = \varepsilon_0 \sum_{j=1}^{3} \left[\int_0^\infty \mathrm{d}\tau_1 \, R_{ij}^{(1)}(\tau_1) \mathrm{e}^{\mathrm{i}\omega\tau_1} \right] \tilde{E}_j(\omega) = \varepsilon_0 \sum_{j=1}^{3} \chi_{ij}^{(1)}(-\omega|\omega) \tilde{E}_j(\omega). \tag{1.11}$$

Here we introduce the complex valued linear susceptibility $\chi_{ij}^{(1)}(-\omega|\omega)$ which is the Fourier transform of the real valued response function

$$\chi_{ij}^{(1)}(-\omega|\omega) = \int_0^\infty \mathrm{d}\tau_1 \, R_{ij}^{(1)}(\tau_1) \mathrm{e}^{\mathrm{i}\omega\tau_1}. \tag{1.12}$$

The two arguments of $\chi_{ij}^{(1)}(-\omega|\omega)$ are introduced for consistency with the following notation of complex coefficients. The first argument represents the negative of the frequency of the induced polarization, where the second argument stands for the frequency of the electric field. Of course, in the linear case the frequency of the induced polarization and of the electric field coincide and therefore the first argument is usually omitted in the linear case. However, as we will see below such a simplification is no longer possible in the nonlinear case.

c.) Similarly as before we can perform a spectral decomposition of the quadratic polarization $\boldsymbol{P}^{(2)}(t)$ by inserting (1.8) into (1.6)

$$P_i^{(2)}(t) = \varepsilon_0 \sum_{i,j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2 \, R_{ijk}^{(2)}(\tau_1,\tau_2) E_j(t-\tau_1) E_k(t-\tau_2) \tag{1.13}$$

$$\begin{aligned} &= \varepsilon_0 \sum_{i,j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2 \, R_{ijk}^{(2)}(\tau_1,\tau_2) \int_{-\infty}^\infty \mathrm{d}\omega_1 \, \tilde{E}_j(\omega_1) \mathrm{e}^{-\mathrm{i}\omega_1(t-\tau_1)} \int_{-\infty}^\infty \mathrm{d}\omega_2 \, \tilde{E}_k(\omega_2) \mathrm{e}^{-\mathrm{i}\omega_2(t-\tau_2)} \\ &= \int_{-\infty}^\infty \mathrm{d}\omega_1 \int_{-\infty}^\infty \mathrm{d}\omega_2 \, \varepsilon_0 \sum_{i,j=1}^{3} \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2 \, R_{ijk}^{(2)}(\tau_1,\tau_2) \mathrm{e}^{\mathrm{i}(\omega_1\tau_1+\omega_2\tau_2)} \tilde{E}_j(\omega_1) \tilde{E}_k(\omega_2) \mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t}. \end{aligned}$$

Now we apply the Fourier transform to (1.13)

$$\begin{aligned}
\tilde{P}_i^{(2)}(\omega_p) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{\mathrm{i}\omega_p t} P_i^{(2)}(t) \\
&= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{\mathrm{i}\omega_p t} \int_{-\infty}^{\infty}\mathrm{d}\omega_1 \int_{-\infty}^{\infty}\mathrm{d}\omega_2 \Big[\\
&\qquad \varepsilon_0 \sum_{i,j=1}^{3} \int_0^{\infty}\mathrm{d}\tau_1 \int_0^{\infty}\mathrm{d}\tau_2\, R_{ijk}^{(2)}(\tau_1,\tau_2)\mathrm{e}^{\mathrm{i}(\omega_1\tau_1+\omega_2\tau_2)}\tilde{E}_j(\omega_1)\tilde{E}_k(\omega_2)\mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t}\Big] \\
&= \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{\mathrm{i}(\omega_p-\omega_1-\omega_2)t} \ldots \\
&\qquad \int_{-\infty}^{\infty}\mathrm{d}\omega_1 \int_{-\infty}^{\infty}\mathrm{d}\omega_2 \left[\varepsilon_0 \sum_{i,j=1}^{3} \int_0^{\infty}\mathrm{d}\tau_1 \int_0^{\infty}\mathrm{d}\tau_2\, R_{ijk}^{(2)}(\tau_1,\tau_2)\mathrm{e}^{\mathrm{i}(\omega_1\tau_1+\omega_2\tau_2)}\right]\tilde{E}_j(\omega_1)\tilde{E}_k(\omega_2).
\end{aligned} \tag{1.14}$$

We make use of the definition of the delta-distribution

$$\frac{1}{2\pi}\int_{-\infty}^{\infty}\mathrm{d}t\,\mathrm{e}^{\mathrm{i}(\omega_p-\omega_1-\omega_2)t} = \delta(\omega_p-\omega_1-\omega_2) \quad \text{with} \quad \int_{-\infty}^{\infty}\mathrm{d}x\,\delta(x)f(x) = f(x=0). \tag{1.15}$$

Inserting the delta-distribution and integrating results in

$$\begin{aligned}
\tilde{P}_i^{(2)}(\omega_p) &= \int_{-\infty}^{\infty}\mathrm{d}\omega_1 \int_{-\infty}^{\infty}\mathrm{d}\omega_2 \sum_{j,k=1}^{3} \chi_{ijk}^{(2)}(-\omega_p|\omega_1,\omega_2)\tilde{E}_j(\omega_1)\tilde{E}_k(\omega_2)\delta(\omega_p-\omega_1-\omega_2) \\
&= \int_{-\infty}^{\infty}\mathrm{d}\omega_1\,\varepsilon_0 \sum_{j,k=1}^{3} \chi_{ijk}^{(2)}(-\omega_p|\omega_1,\omega_p-\omega_1)\tilde{E}_j(\omega_1)\tilde{E}_k(\omega_p-\omega_1)
\end{aligned} \tag{1.16}$$

with the nonlinear susceptibility of second order

$$\chi_{ijk}^{(2)}(-\omega_p|\omega_1,\omega_2) = \int_0^{\infty}\mathrm{d}\tau_1 \int_0^{\infty}\mathrm{d}\tau_2\, R_{ijk}^{(2)}(\tau_1,\tau_2)\mathrm{e}^{\mathrm{i}(\omega_1\tau_1+\omega_2\tau_2)}. \tag{1.17}$$

Several comments are in order:

- Nonlinearities generate new frequencies in the induce polarization, which are converted to optical fields with new frequency components (new colors) by emission.
- The delta distribution $\delta(\omega_p-\omega_1-\omega_2)$, which we have finally integrated, ensures energy conservation.
- A photon of frequency ω_p can only be created, if its frequency is equal to the sum of the frequencies of the initial photons (including negative frequencies).

- Because the spectral amplitudes of the electric field can also have arguments with negative frequencies, equation (1.16) incorporates difference as well as sum frequency generation.

d.) Higher order nonlinear polarizations are introduced in the same way as to derive equation (1.16)

$$\tilde{P}_{\alpha_p}(n)(\omega_p) = \int_{-\infty}^{\infty} \mathrm{d}\omega_1 \ldots \int_{-\infty}^{\infty} \mathrm{d}\omega_n \, \varepsilon_0 \sum_{\alpha_1=1}^{3} \ldots \sum_{\alpha_n=1}^{3} \Big[\chi^{(n)}_{\alpha_p \alpha_1 \ldots \alpha_n}(-\omega_p|\omega_1,\ldots,\omega_n) \tilde{E}_{\alpha_1}(\omega_1) \ldots \tilde{E}_{\alpha_n}(\omega_n) \delta\big(\omega_p - \sum_{k=1}^{n} \omega_k\big) \Big] \tag{1.18}$$

with the nonlinear susceptibility

$$\chi^{(n)}_{\alpha_p \alpha_1 \ldots \alpha_n}(-\omega_p|\omega_1,\ldots,\omega_n) = \int_0^{\infty} \mathrm{d}\tau_1 \ldots \int_0^{\infty} \mathrm{d}\tau_n \, R^{(2)}_{\alpha_p \alpha_1 \ldots \alpha_n}(\tau_1,\ldots,\tau_n) \mathrm{e}^{\mathrm{i}(\omega_1 \tau_1)} \ldots \mathrm{e}^{\mathrm{i}(\omega_n \tau_n)}, \tag{1.19}$$

and $\omega_p = \sum_{i=1}^{n} \omega_i$ ensuring energy conservation.

Of course one of the frequencies found in the integral in equation (1.18) can be removed by performing the integration and evaluating the delta-distribution. We also want to list some properties of $\chi^{(n)}_{\alpha_p \alpha_1 \ldots \alpha_n}(-\omega_p|\omega_1,\ldots,\omega_n)$:

- Only defined/different from zero for $\omega_p = \omega_1 + \ldots + \omega_n$
- Arguments originating from the driving fields (frequencies and indices of polarization) can be exchanged together:
  $\chi^{(2)}_{\alpha_p \alpha_1 \ldots \alpha_k \ldots \alpha_l \ldots \alpha_n}(-\omega_p|\omega_1 \ldots \omega_k \ldots \omega_l \ldots \omega_n) = \chi^{(2)}_{\alpha_p \alpha_1 \ldots \alpha_l \ldots \alpha_k \ldots \alpha_n}(-\omega_p|\omega_1 \ldots \omega_l \ldots \omega_k \ldots \omega_n)$
- The susceptibility carries the symmetries of the respective material. The higher the symmetry of the material the lower the number of independent coefficients.
- A KRAMERS-KRONIG transformation can be performed with respect to one of the frequencies. It links real and imaginary parts of the coefficients as

  $$\chi^{(n)}_{\alpha_p \alpha_1 \ldots \alpha_n}(-\omega_p(\omega_0)|\omega_1,\ldots\omega_0\ldots\omega_n) = \frac{1}{\mathrm{i}\pi} \mathscr{P} \int_{-\infty}^{\infty} \mathrm{d}\varpi \, \frac{\chi^{(n)}_{\alpha_p \alpha_1 \ldots \alpha_n}(-\omega_p(\varpi)|\omega_1,\ldots\varpi\ldots\omega_n)}{\varpi - \omega_0}$$

  with $\mathscr{P}$ being Cauchy's principal value integral.

e.) Furthermore we want to take a look at the nonlinear interaction of several continuous waves (cw) which are discrete frequency components instead of a continuous spectrum

$$\boldsymbol{E}(t) = \frac{1}{2} \sum_{\lambda} \big(\boldsymbol{E}_\lambda \mathrm{e}^{-\mathrm{i}\omega_\lambda t} + \text{c.c.} \big). \tag{1.20}$$

The quantity $|\boldsymbol{E}_\lambda|$ is the peak amplitude of the electric field component with frequency ω_λ. Fourier spectrum of sum of cw-waves is equal to a sum of delta functions

$$\tilde{\boldsymbol{E}}(\omega) = \frac{1}{2} \sum_{\lambda} \big[\boldsymbol{E}_\lambda \delta(\omega - \omega_\lambda) + \boldsymbol{E}^*_\lambda \delta(\omega + \omega_\lambda) \big]. \tag{1.21}$$

The nonlinear polarization of n-th order induced by cw-waves is a sum of harmonic oscillations

$$\boldsymbol{P}^{(n)}(t) = \frac{1}{2}\sum_{\omega_p}\left(\boldsymbol{P}^{(n)}_{\omega_p} \mathrm{e}^{-\mathrm{i}\omega_p t} + \text{c.c.}\right). \tag{1.22}$$

We now want to determine the amplitudes of harmonic oscillations of the nonlinear polarization of n-th order induced by cw-waves $\boldsymbol{P}^{(n)}_{\omega_p}$ by following these steps:

1. Formal integration of equation (1.18) using the Fourier spectrum from (1.21).
2. Inserting the nonlinear coefficient $\chi^{(n)}_{\alpha_p\alpha_1\ldots\alpha_n}(-\omega_p|\omega_1,\ldots,\omega_n)$ defined in equation (1.19).
3. Evaluating the Fourier spectrum of nonlinear polarization of n-th order induced by cw-waves

$$\tilde{\boldsymbol{P}}^{(n)}(\omega) = \frac{1}{2}\sum_{\omega_p}\left(\boldsymbol{P}^{(n)}_{\omega_p}\delta(\omega-\omega_p) + \boldsymbol{P}^{(n)*}_{\omega_p}\delta(\omega+\omega_p)\right). \tag{1.23}$$

4. Evaluating permutation symmetry (incorporated into the K-coefficient).

The spectral amplitude of the nonlinear polarization can now be written as

$$P^{(n)}_{\omega_p\alpha_p} = \varepsilon_0 \sum_{\{\pm\omega_\lambda\}} K(-\omega_p|\pm\omega_1,\ldots\pm\omega_n) \sum_{\alpha_1\ldots\alpha_n=1}^{3} \chi^{(n)}_{\alpha_p\alpha_1\ldots\alpha_n}(-\omega_p|\pm\omega_1,\ldots\pm\omega_n) E^{(*)}_{1,\alpha_1} E^{(*)}_{n,\alpha_n} \tag{1.24}$$

- $\pm\omega_\lambda$: one of the frequencies of the interacting cw-waves, either positive or negative
- $E^{(*)}_{\lambda,\alpha_\lambda}$: vector component α_λ of the field amplitude $\boldsymbol{E}_\lambda$ of the continuous wave oscillating with ω_λ. In case of the negative frequency the complex conjugate field profile $E^*_{\lambda,\alpha_\lambda}$ is chosen.
- $\{\pm\omega_\lambda\}$: all combinations of interacting $\pm\omega_\lambda$ including negative frequencies, which generate ω_p, but no permutations.

The factor $K(-\omega_p|\pm\omega_1,\ldots\pm\omega_n)$ incorporates the effect of permutations. It includes all factors $\frac{1}{2}$ used to express the amplitudes of the electric fields and the polarization. We can generalize it in the following way:

$$K(-\omega_p|\pm\omega_1,\ldots\pm\omega_n) = 2^{l+m-n}p. \tag{1.25}$$

- n: order of the nonlinear interaction
- p: number of permutations of the $\pm\omega_\lambda$ (p is equal to $n!$ if all frequencies are different.)
- m: number of ω_λ being zero (For those the factor $\frac{1}{2}$ vanishes in the above expression (1.20) for the fields.)
- l: equal to 1 provided that ω_p is not zero, otherwise $l = 0$

1.4 Symmetries and conserved quantities

a.) Complete permutation symmetry

We want to extend the permutation symmetry of the electric fields represented by ω_λ towards the induced polarization:

$$\chi_{\alpha_p\alpha_1\ldots\alpha_k\ldots\alpha_n}(-\omega_p|\omega_1\ldots\omega_k\omega_n) = \chi_{\alpha_k\alpha_1\ldots\alpha_p\ldots\alpha_n}(\omega_k|\omega_1\ldots-\omega_p\omega_n). \tag{1.26}$$

This assumption is justified far away from resonance which means that nonlinear coefficients can be expressed by a Drude model

$$\chi^{(n)}_{\alpha_p\alpha_1\ldots\alpha_n}(-\omega_p|\omega_1\ldots\omega_n) \sim \frac{1}{\omega_0^2-\omega_p^2}\prod_{\lambda=1}^{n}\frac{1}{\omega_0^2-\omega_\lambda^2}, \tag{1.27}$$

where ω_0 is the resonance frequency of the material. The frequency of the polarization does not play an extraordinary role and can be mixed with the other components.

b.) Kleinmann symmetry (no frequency dependence)

The preconditions for the Kleinmann symmetry to hold are that we deal with frequencies in the transparent domain of the material which are far away from resonances ($\omega_0^2 \gg \omega_p^2$ and $\omega_0^2 \gg \omega_\lambda^2$). Then all frequency dependence can be neglected. Often Kleinmann symmetry is assumed because of the lack of more detailed knowledge.

For quadratic nonlinearities we can introduce a matrix notation based on field amplitudes of cw-fields defined in (1.20) and (1.21)

$$\begin{pmatrix} P_x^{(2)} \\ P_y^{(2)} \\ P_z^{(2)} \end{pmatrix} = 2\varepsilon_0 K(-\omega_p|\omega_1,\omega_2)\begin{pmatrix} d_{11}\ldots d_{16} \\ d_{21}\ldots d_{26} \\ d_{31}\ldots d_{36} \end{pmatrix}\begin{pmatrix} E_x(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_z(\omega_2) \\ E_y(\omega_1)E_z(\omega_2)+E_z(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_z(\omega_2)+E_z(\omega_1)E_x(\omega_2) \\ E_x(\omega_1)E_y(\omega_2)+E_y(\omega_1)E_x(\omega_2) \end{pmatrix}. \tag{1.28}$$

The d-coefficients are a very common representation of quadratic nonlinearities. They are orientation dependent (x- and y-axis usually aligned parallel to the crystal axis). We can also write down the K-factor for all cases possible:

- All frequencies different and nonzero: $K = 1$
- $\omega_1 = \omega_2$ or $\omega_1 = -\omega_2$: $K = \frac{1}{2}$
- $\omega_1 = 0$ or $\omega_2 = 0$: $K = 2$

c.) Conservation laws

For arbitrary orders of nonlinearities we can find conservation laws which are valid in the absence of dissipation or losses. We note W_λ as the energy flux of a field component of frequency ω_λ which points into a certain direction. Energy conservation means that $\sum_\lambda W_\lambda$ =const. For non-degenerate processes $|\omega_\alpha| \neq |\omega_\beta|$ and arbitrary α and β including the induced polarization we have photon flux conservation

$$\frac{W_\alpha}{\omega_\alpha} - \frac{W_\beta}{\omega_\beta} = \text{const.} \tag{1.29}$$

To successfully create a photon at ω_p a photon of each of the interacting waves must be absorbed (photon number difference remains conserved).

1.5 Cubic nonlinearities in isotropic materials

Cubic nonlinearities can usually be neglected if quadratic nonlinearities are present. Therefore they are only relevant if quadratic coefficients vanish (e. g. in inversion symmetric materials as glass) or if quadratically nonlinear processes are suppressed (e. g. due to the lack of phase matching). The susceptibility tensor

$$\chi^{(3)}_{ijkl}(-(\omega_1+\omega_2+\omega_3)|\omega_1,\omega_2,\omega_3) \tag{1.30}$$

possesses $3^4 = 81$ elements.

From now on will will only consider *isotropic* materials with the highest possible degree of symmetry, which means they have the minimum number of independent elements.

a.) All coefficients are equal $j = k = l$

These are coefficients such as $\chi^{(3)}_{xxxx}$ or $\chi^{(3)}_{yxxx}$. We assume that $\boldsymbol{E}$ points into the direction of equal coefficients

$$\boldsymbol{E} = (E_j + E_k + E_l)\hat{\mathbf{e}}_k.$$

Thus, the induced polarization must point into the same direction as $\boldsymbol{E}$ because the material has no internal direction. Since $\boldsymbol{E}||\boldsymbol{P}$ we have

$$\chi^{(3)}_{ikkk} = 0 \quad \text{for} \quad i \neq k \quad \text{and} \quad \chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz}. \tag{1.31}$$

The three non-vanishing elements must be equal as no direction is preferred. Thus we only have one independent element.

b.) One coefficient is different $j = k \neq l$

Here we assume that $\boldsymbol{E}$ points into the two relevant directions

$$\boldsymbol{E} = (E_j + E_k)\hat{\mathbf{e}}_k + E_l\hat{\mathbf{e}}_l.$$

The vector components of $\boldsymbol{P}^{(3)}$ at a fixed frequency are $P_i^{(3)} \sim \chi_{ikkl}^{(3)} E_j E_k E_l$. We assume that the material is rotated around the singular axis (l) by 180°. In the new coordinate system the electric field is

$$\boldsymbol{E}' = -(E_j + E_k)\hat{\mathbf{e}}_k' + E_l\hat{\mathbf{e}}_l'.$$

As the material is isotropic the nonlinear coefficient has not changed by rotation $\chi_{ikkl}^{(3)} = \chi_{ikkl}^{(3)'}$. The vector components of $\boldsymbol{P}^{(3)}$ in the rotated coordinate system are

$$P_i^{(3)'} \sim \chi_{ikkl}^{(3)}(-E_j)(-E_k)E_l = P_i^{(3)}. \tag{1.32}$$

This is only possible if $\boldsymbol{P}^{(3)}$ points along the axis of rotation. It follows that $\chi_{ikkl}^{(3)} = 0$ for $i \neq l$. Thus we have 18 non-vanishing coefficients (all permutations are taken into account, for different frequencies permutations give different coefficients)

$$\chi_{xyyx}^{(3)}(-(\omega_1+\omega_2+\omega_3)|\omega_1,\omega_2,\omega_3) \neq \chi_{xxyy}^{(3)}(-(\omega_1+\omega_2+\omega_3)|\omega_1,\omega_2,\omega_3). \tag{1.33}$$

However, we only have three independent coefficients e. g.

$$\chi_{xyyx}^{(3)} = \chi_{xzzx}^{(3)} = \chi_{yxxy}^{(3)} = \chi_{yzzy}^{(3)} = \chi_{zxxz}^{(3)} = \chi_{zyyz}^{(3)} \neq \chi_{xxyy}^{(3)}. \tag{1.34}$$

c.) All indices are different $j \neq k \neq l \neq j$

With the same arguments as above we can perform a 180° rotation of the material and the coordinate system around e. g. the j-direction and will find out that $\boldsymbol{P}^{(3)}$ must point into that direction. This we can do with every of direction as all are represented by a single field. As $\boldsymbol{P}^{(3)}$ can only point into a single direction it must vanish for that condition. All coefficients are zero.

Concluding our results a general $\chi^{(3)}$ tensor has **21** non-zero coefficients and only four independent coefficients. In the following we will introduce degeneracies in the interaction waves leading to Third harmonic generation and self-phase modulation.

1.5.1 Third harmonic generation (THG)

For this process we consider the case $j = k \neq l = i$ with the nonlinear coefficient

$$\chi_{ijkl}^{(3)}(-3\omega,\omega,\omega,\omega).$$

Since the arguments are equal the frequencies can be exchanged freely

$$\chi_{xxyy}^{(3)} = \chi_{xyxy}^{(3)} = \chi_{xyyx}^{(3)} = \ldots = \chi_{iikk}^{(3)}. \tag{1.35}$$

In total we have only two independent coefficients $\chi^{(3)}_{iikk}$ and $\chi^{(3)}_{iiii}$. In case of THG they are not really independent which will be shown in the following.

We assume that we can rotate the material in such a way that the electric field $\boldsymbol{E}$ points into the y- and z-direction

$$\boldsymbol{E} = \frac{E}{\sqrt{2}}\begin{pmatrix}0\\1\\1\end{pmatrix}.$$

The resulting components of the polarization are

$$\begin{aligned} P^{(3)}_y &= \varepsilon_0 K(-3\omega|\omega,\omega,\omega)\frac{E^2}{2}\left(\chi^{(3)}_{yzzy}+\chi^{(3)}_{yzyz}+\chi^{(3)}_{yyzz}+\chi^{(3)}_{yyyy}\right)E_y \\ P^{(3)}_z &= \varepsilon_0 K(-3\omega|\omega,\omega,\omega)\frac{E^2}{2}\left(\chi^{(3)}_{zyyz}+\chi^{(3)}_{zyzy}+\chi^{(3)}_{zzyy}+\chi^{(3)}_{zzzz}\right)E_z. \end{aligned} \tag{1.36}$$

For THG there are no frequency permutations $p = 1$ which implies $K = 1/4$ (see equation (1.25)). Then we have

$$\boldsymbol{P}^{(3)} = \varepsilon_0 K(-3\omega|\omega,\omega,\omega)\frac{E^2}{2}(3\chi^{(3)}_{yzzy}+\chi^{(3)}_{yyyy})\boldsymbol{E}. \tag{1.37}$$

The polarization is parallel to $\boldsymbol{E}$ which is no surprise, as the material is isotropic.

Now we assume that the material (and the coordinate system) is rotated in such a way that $\boldsymbol{E}$ points into the y-direction. This implies

$$\boldsymbol{P}^{(3)} = \varepsilon_0 K(-3\omega|\omega,\omega,\omega)E^2\chi^{(3)}_{yyyy}\boldsymbol{E}. \tag{1.38}$$

The relation between $\boldsymbol{P}^{(3)}$ and $\boldsymbol{E}$ must be independent of the rotation. Therefore we can equate (1.37) and (1.38)

$$\frac{3}{2}\chi^{(3)}_{yzzy}+\frac{1}{2}\chi^{(3)}_{yyyy} = \chi^{(3)}_{yyyy}. \tag{1.39}$$

This means in an isotropic material THG is determined by a single independent coefficient

$$\chi^{(3)}_{xxyy} = \chi^{(3)}_{xyxy} = \ldots = \chi^{(3)}_{\mathrm{THG}} \quad \text{and} \quad \chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz} = 3\chi^{(3)}_{\mathrm{THG}}. \tag{1.40}$$

We can generalize the total third harmonic polarization by first looking at a single component

$$\begin{aligned} P^{(3)}_y &= \varepsilon_0\frac{1}{4}(\chi^{(3)}_{yyyy}E_y^3+3\chi^{(3)}_{yzzy}E_yE_z^2+3\chi^{(3)}_{yxxy}E_yE_x^2) \\ &= \varepsilon_0\frac{3}{4}\chi^{(3)}_{\mathrm{THG}}\left(E_y^2+E_z^2+E_x^2\right)E_y \\ &= \varepsilon_0\frac{3}{4}\chi^{(3)}_{\mathrm{THG}}\boldsymbol{E}^2E_y. \end{aligned} \tag{1.41}$$

Thus the total third harmonic polarization is

$$\boxed{\boldsymbol{P}^{(3)} = \varepsilon_0\frac{3}{4}\chi^{(3)}_{\mathrm{THG}}\boldsymbol{E}^2\boldsymbol{E}}. \tag{1.42}$$

The third harmonic generated in an isotropic medium:

- points into the direction of the exciting field
- vanishes for circularly polarized light.

For left circularly polarized light: $\boldsymbol{E} = \frac{E}{\sqrt{2}}(\hat{\mathbf{e}}_x + \mathrm{i}\hat{\mathbf{e}}_y)$ which means $\boldsymbol{E}^2 = \frac{E^2}{2} - \frac{E^2}{2} = 0$.

We want to give an explanation for this phenomenon. In an isotropic medium the direction of polarization is entirely determined by the direction of the exciting field. In case of circularly polarized electric field the polarization can only rotate with the rotation speed of the fundamental harmonic, hence one rotation per optical cycle of the fundamental harmonic. Therefore, only polarization at fundamental harmonic frequency is generated.

1.5.2 Self phase modulation (SPM) or optical Kerr effect

Here we have the following nonlinear interaction: $\chi^{(3)}_{ijkl}(-\omega|\omega,\omega,-\omega)$.
The negative frequency $-\omega$ corresponds to the complex conjugate of the field $\boldsymbol{E}^*$. The nonlinear polarization and the driving field coincide in frequency. We observe a nonlinear phase modulation $\mathrm{Re}\,\chi^{(3)}_{ijkl}$ and losses $\mathrm{Im}\,\chi^{(3)}_{ijkl}$ of the initial wave only. We have more independent coefficients than for THG, e. g.

$$\chi^{(3)}_{xxyy}(-\omega|\omega,\omega,-\omega) \neq \chi^{(3)}_{xyyx}(-\omega|\omega,\omega,-\omega,). \tag{1.43}$$

The third order nonlinear polarization has the following form:

$$P^{(3)}_y = \varepsilon_0 \underbrace{K}_{3/4}\Big[2\Big(\chi^{(3)}_{yyxx}|E_x|^2E_y + \chi^{(3)}_{yyzz}|E_z|^2E_y\Big) + \chi^{(3)}_{yyyy}|E_y|^2E_y + \chi^{(3)}_{yxxy}E_x^2E_y^* + \chi^{(3)}_{yzzy}E_z^2E_y^*\Big]. \tag{1.44}$$

For SPM we have three frequency permutations ($p = 3$) and therefore $K = 3/4$ (see equation (1.25)). We use the same arguments as for THG

$$\chi^{(3)}_{yyyy} = \frac{1}{2}(\chi^{(3)}_{yzzy} + 2\chi^{(3)}_{yyzz} + \chi^{(3)}_{yyyy}) \quad \Rightarrow \quad \chi^{(3)}_{yyyy} = \chi^{(3)}_{yzzy} + 2\chi^{(3)}_{yyzz}. \tag{1.45}$$

$$\begin{aligned} P^{(3)}_y &= \varepsilon_0 K\Big[2\chi^{(3)}_{yyzz}(|E_x|^2 + |E_y|^2 + |E_z|^2)E_y + \chi^{(3)}_{yzzy}|E_y|^2E_y + \chi^{(3)}_{yzzy}(E_x^2 + E_z^2)E_y^*\Big] \\ &= \varepsilon_0 K\Big[2\chi^{(3)}_{yyzz}(|E_x|^2 + |E_y|^2 + |E_z|^2)E_y + \chi^{(3)}_{yzzy}(E_x^2 + E_y^2 + E_z^2)E_y^*\Big]. \end{aligned} \tag{1.46}$$

Neither x, y nor z direction are preferred:

$$\boldsymbol{P}^{(3)}(\omega) = \frac{3}{4}\varepsilon_0\Big(2\chi^{(3)}_{yyzz}|\boldsymbol{E}|^2\boldsymbol{E} + \chi^{(3)}_{yzzy}\boldsymbol{E}^2\boldsymbol{E}^*\Big). \tag{1.47}$$

We find two independent coefficients and want two discuss both nonlinear actions:

- First term: $|\boldsymbol{E}|^2\boldsymbol{E}$
  depends on the intensity only and is a pure phase modulation or a nonlinear loss

- Second term: $\boldsymbol{E}^2\boldsymbol{E}^*$
  is sensitive to the state of polarization of the electric field and completely vanishes for circularly polarized light as $\boldsymbol{E}^2$ (see end of section 1.5.1). It bears half of the contribution as the first term for linearly polarized light.

We can further simply the result for KLEINMANN symmetry (no frequency dependence): $\chi^{(3)}_{yzzy} = \chi^{(3)}_{yyzz}$. Only a single coefficient is left and we obtain

$$\boxed{\boldsymbol{P}^{(3)} = \varepsilon_0 \frac{3}{4}\chi^{(3)}_{yyzz}\left(2|\boldsymbol{E}|^2\boldsymbol{E} + \boldsymbol{E}^2\boldsymbol{E}^*\right)}. \tag{1.48}$$

Optical Kerr effect

Now we take linearly polarized light (e. g. $\boldsymbol{E} = E\hat{\mathbf{e}}_x$)

$$\boldsymbol{P}^{(3)} = \varepsilon_0 \frac{9}{4}\chi^{(3)}_{yyzz}|\boldsymbol{E}|^2\boldsymbol{E} = \varepsilon_0\Delta\chi \quad \text{with} \quad \Delta\chi = \frac{9}{4}\chi^{(3)}_{yyzz}|\boldsymbol{E}|^2. \tag{1.49}$$

The action of the nonlinear susceptibility can be expressed by a change of the refractive index $n = \sqrt{1+\chi}$ as

$$\Delta n = \sqrt{n_0^2 + \Delta\chi} - n_0 \approx \frac{\Delta\chi}{2n} = \frac{9}{8n_0}\chi^{(3)}_{yyzz}|\boldsymbol{E}|^2, \tag{1.50}$$

where n_0 is the refractive index in the absence of a nonlinear action. As there is no generally accepted definition of the amplitude of an oscillating field one prefers to refer to the intensity

$$I = \frac{\varepsilon_0}{2} c n_0 |\boldsymbol{E}|^2. \tag{1.51}$$

We can replace the electric field in (1.50) and find

$$\Delta n = n_2 I \quad \text{with} \quad n_2 = \frac{9}{4}\frac{\chi^{(3)}_{yyzz}}{\varepsilon_0 c n_0^2}. \tag{1.52}$$

We want to give some examples of nonlinear refractive indices:

$$\text{silica} \qquad n_2 = 2{,}3 \cdot 10^{-20}\,\frac{\text{m}^2}{\text{W}} \tag{1.53}$$

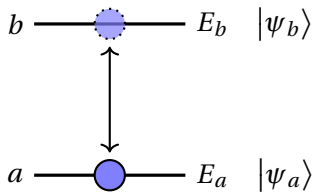
$$\text{Al}_{0.18}\text{Ga}_{0.82}As(1{,}5\,\mu\text{m}) \qquad n_2 = 2{,}1 \cdot 10^{-17}\,\frac{\text{m}^2}{\text{W}}. \tag{1.54}$$

2 Resonant nonlinearities - a quantum mechanical representation

In what follows we will shortly discuss the kind of nonlinear response, which shows up close to a resonance of the material. If the energy of the incident photons just roughly matches the energy of a quantum mechanical transition, the optical properties of the excited material may change dramatically as a function of the incident intensity. A Taylor expansion of the induced polarization with respect to powers of the acting electrical field as done in equation (1.4) is still applicable, however, the treatment applied in the last section is no longer useful. The expansion can usually not be restricted to a few orders and resulting coefficients turn out to be highly dispersive. Hence, a treatment taking into account the material response more seriously has to be applied.

2.1 Equations of motion of a two-level-system

We first want to revise what a two-level-system actually is: It is a quantum system with (at least) two energy levels a and b corresponding to distinct states $|\psi_a\rangle$ (excited state) and $|\psi_b\rangle$ (ground state) with energies E_a and E_b ($E_a > E_b$). The energy of photons exciting the ground state is $\hbar\omega_0 \approx E_a - E_b$.



Real quantum states have many (usually infinite) possible states. A two-level-system is singled out by the excitation. The dynamics of the system are determined by the Hamiltonian $\hat{H}$ for a single electron in an electromagnetic field

$$\hat{H} = \frac{1}{2m}(\hat{\boldsymbol{p}} + e\boldsymbol{A}_{\text{real}})^2 + V(\boldsymbol{r}). \tag{2.1}$$

The term $V(\boldsymbol{r})$ is the potential, which localizes the electrons. $\boldsymbol{A}_{\text{real}}$ is the vector potential, which represents the optical field by

$$\boldsymbol{E}_{\text{real}} = -\frac{\partial}{\partial t}\boldsymbol{A}_{\text{real}}. \tag{2.2}$$

The subscript *real* is used to emphasize that observable fields are real valued and that they contain components oscillating at $+\omega_0$ and $-\omega_0$. We consider the vector potential as *classical* which means that

- The strong field contains many photons
- quantum fluctuations can be neglected
- The field commutes with all operators
- The vector potential is weak compared to atomic fields $\Rightarrow \boldsymbol{A}^2_{\text{real}}$ is neglected.

We can now decompose the Hamiltonian into an unperturbed part $\hat{H}_0$ and a perturbation $\hat{W}$

$$\hat{H} = \underbrace{\frac{1}{2m}\hat{\boldsymbol{p}}^2 + V(\boldsymbol{r})}_{\hat{H}_0} + \underbrace{\frac{e}{m}\hat{\boldsymbol{p}}\boldsymbol{A}_{\text{real}}}_{\hat{W}} + \cancel{\frac{e^2}{2m}\boldsymbol{A}_{\text{real}}^2}. \tag{2.3}$$

The unperturbed part $\hat{H}_0$ is the Hamiltonian of the unperturbed two-level-system in absence of any electromagnetic field. It defines the energy levels E_a and follows the SCHRÖDINGER equation $\hat{H}_0 |\psi_a\rangle = E_a |\psi_a\rangle$, where $|\psi_{a/b}\rangle$ is a orthonormal basis of eigenstates $\langle\psi_a|\psi_b|\psi_a|\psi_b\rangle = \delta_{ab}$. The evolution is restricted to the two eigenstates, which means that the system is either in state a or b. The wave function of the whole system can be expressed as

$$|\psi(t)\rangle = a(t)|\psi_a\rangle + b(t)|\psi_b\rangle \quad \text{with} \quad |a(t)|^2 + |b(t)|^2 = 1, \tag{2.4}$$

where the second conditions follows from $\langle\psi(t)|\psi(t)|\psi(t)|\psi(t)\rangle = 1$.

The interaction operator $\hat{W}$ drives the nontrivial dynamics and induces changes. Using the time dependent SCHRÖDINGER equation leads to

$$\begin{aligned}
\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}|\psi(t)\rangle &= \hat{H}|\psi(t)\rangle \\
\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\big(a(t)|\psi_a\rangle + b(t)|\psi_b\rangle\big) &= \hat{H}(a(t)|\psi_a\rangle + b(t)|\psi_b\rangle) \\
\mathrm{i}\hbar|\psi_a\rangle\frac{\mathrm{d}}{\mathrm{d}t}a(t) + \mathrm{i}\hbar|\psi_b\rangle\frac{\mathrm{d}}{\mathrm{d}t}b(t) &= a(t)\hat{H}|\psi_a\rangle + b(t)\hat{H}|\psi_b\rangle.
\end{aligned} \tag{2.5}$$

Multiplying from the left with $\langle\psi_{a/b}|$ and using $\langle\psi_a|\psi_b|\psi_a|\psi_b\rangle = \delta_{ab}$ leads to

$$\begin{aligned}
\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}a &= E_a a + \langle\psi_a|\hat{W}|\psi_a\rangle a + \langle\psi_a|\hat{W}|\psi_b\rangle b \\
\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}b &= E_b b + \underbrace{\langle\psi_b|\hat{W}|\psi_b\rangle b + \langle\psi_b|\hat{W}|\psi_a\rangle a}_{\text{interaction elements}}.
\end{aligned} \tag{2.6}$$

Matrix elements of the interaction operator

We first calculate the symmetric elements:

$$\begin{aligned}
\langle\psi_a|\hat{W}|\psi_a\rangle &= \left\langle\psi_a|\frac{e}{m}\hat{\boldsymbol{p}}\boldsymbol{A}_{\text{real}}|\psi_a\right\rangle = \frac{e}{m}\boldsymbol{A}_{\text{real}}\langle\psi_a|\hat{\boldsymbol{p}}|\psi_a\rangle \\
&= \frac{e}{m}\boldsymbol{A}_{\text{real}}\frac{\hbar}{\mathrm{i}}\int \mathrm{d}V\,\psi_a^*(\boldsymbol{r})\vec{\boldsymbol{\nabla}}\psi_a(\boldsymbol{r}) \\
&= \frac{e}{m}\boldsymbol{A}_{\text{real}}\frac{\hbar}{2\mathrm{i}}\int \mathrm{d}V\,\vec{\boldsymbol{\nabla}}\big(\psi_a(\boldsymbol{r})\big)^2 = 0.
\end{aligned} \tag{2.7}$$

The last integral just gives us the values at the boundary. For a decaying wave functions the boundaries are zero, leading to vanishing interaction terms. In the last line we assume a real valued $\psi_a(\boldsymbol{r})$.

The second matrix element $\langle\psi_a|\hat{W}|\psi_b\rangle = \langle\psi_b|\hat{W}|\psi_a\rangle^*$ is responsible for transitions between the two states. It is usually nonzero , provided that an external field is present and both states have different parity. For the evaluation we need a helpful commutator relation

$$\begin{aligned}[\hat{\boldsymbol{r}}, \hat{H}_0] &= \hat{\boldsymbol{r}}\left[\frac{1}{2m}\hat{\boldsymbol{p}}^2 + V(\boldsymbol{r})\right] - \left[\frac{1}{2m}\hat{\boldsymbol{p}}^2 + V(\boldsymbol{r})\right]\hat{\boldsymbol{r}} \\ &= \frac{1}{2m}(\hat{\boldsymbol{r}}\hat{\boldsymbol{p}}^2 - \hat{\boldsymbol{p}}^2\hat{\boldsymbol{r}}) = \frac{1}{2m}([\hat{\boldsymbol{r}}, \hat{\boldsymbol{p}}]\hat{\boldsymbol{p}} + \hat{\boldsymbol{p}}[\hat{\boldsymbol{r}}, \hat{\boldsymbol{p}}]) = \frac{\mathrm{i}\hbar}{m}\hat{\boldsymbol{p}}. \end{aligned} \tag{2.8}$$

Now we can calculate the matrix element as

$$\begin{aligned}\langle\psi_a|\hat{W}|\psi_b\rangle &= \frac{e}{m}\boldsymbol{A}_{\mathrm{real}}\langle\psi_a|\hat{\boldsymbol{p}}|\psi_b\rangle = \frac{e}{m}\boldsymbol{A}_{\mathrm{real}}\left\langle\psi_a|\frac{m}{\mathrm{i}\hbar}(\hat{\boldsymbol{r}}\hat{H}_0 - \hat{H}_0\hat{\boldsymbol{r}})|\psi_b\right\rangle \\ &= \boldsymbol{A}_{\mathrm{real}}\frac{\mathrm{i}e}{\hbar}(E_a - E_b)\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle. \end{aligned} \tag{2.9}$$

The optical field couples to the states via the dipole moment of the transition

$$\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle = \int \mathrm{d}V\,\psi_a^*(\boldsymbol{r})\boldsymbol{r}\psi_b(\boldsymbol{r}). \tag{2.10}$$

In the following we want to transform a and b into macroscopic observables:

a.) Inversion $I = |a|^2 - |b|^2$

The inversion describes the state of excitation of the two-level-system. It can vary between –1 (no excitation) and 1 (maximum excitation). For its evolution we use equation (2.6)

$$\begin{aligned}\frac{\mathrm{d}I}{\mathrm{d}t} &= \frac{\mathrm{d}}{\mathrm{d}t}\left(|a|^2 - |b|^2\right) \overset{(2.4)}{=} \frac{\mathrm{d}}{\mathrm{d}t}\left(2|a|^2 - 1\right) = 2\frac{\mathrm{d}}{\mathrm{d}t}|a|^2 \\ &= 2a^*\frac{\mathrm{d}}{\mathrm{d}t}a + \text{c.c.} = \frac{2}{\mathrm{i}\hbar}\langle\psi_a|\hat{W}|\psi_b\rangle a^* b + \text{c.c.} \\ &= \frac{2e}{\hbar^2}\boldsymbol{A}_{\mathrm{real}}(E_a - E_b)\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle a^* b + \text{c.c.} \end{aligned} \tag{2.11}$$

Now we want to do a frequency analysis of ab^* for a weak perturbation $\hat{W}$ and a resonant excitation $(E_a - E_b)/\hbar \approx \omega_0$. If the energy of the state is much larger than the interaction energy the solutions of $a(t)$ and $b(t)$ using (2.6) are

$$a(t) \sim \exp\left(-\frac{\mathrm{i}E_a}{\hbar t}\right), \qquad b(t) \sim \exp\left(-\frac{\mathrm{i}E_b}{\hbar t}\right). \tag{2.12}$$

This leads to the following expression for ab^*

$$ab^* \sim \exp\left(-\mathrm{i}\frac{E_a - E_b}{\hbar}t\right) \approx \exp(-\mathrm{i}\omega_0 t), \tag{2.13}$$

which means that ab^* is related to the optical polarization.

b.) Polarization

The classical change of inversion is a result of absorbed optical power

$$\frac{\mathrm{d}I}{\mathrm{d}t} = 2\frac{\text{absorbed power per system}}{\text{energy required to excite a system}} = \frac{2}{E_a - E_b}\frac{1}{N}\boldsymbol{E}_{\text{real}}\frac{\partial}{\partial t}\boldsymbol{P}_{\text{real}}. \tag{2.14}$$

This can be understood by noting that power is Voltage times current, where $\boldsymbol{E}_{\text{real}}$ corresponds to the voltage and $\frac{\partial}{\partial t}\boldsymbol{P}_{\text{real}}$ to current. The latter one can be understood that the polarization describes a deviation of the electrons from their rest position in the lattice. A change of polarization describes the electron oscillations around their rest position and thus a current. Then $\boldsymbol{E}_{\text{real}}\frac{\partial}{\partial t}\boldsymbol{P}_{\text{real}}$ is the absorbed power per volume. It contains fast parts (oscillations with $\approx 2\omega_0$), however, they average out or can be neglected. In the following we only consider the slow/almost stationary parts.

We decompose the fields into fast parts oscillating with the carrier frequency of the optical field ω_0 and slowly varying amplitudes

$$\begin{aligned}
\boldsymbol{P}_{\text{real}}(t) &= \frac{1}{2}\left[\boldsymbol{P}(t)\exp(-\mathrm{i}\omega_0 t) + \boldsymbol{P}^*(t)\exp(\mathrm{i}\omega_0 t)\right] \\
\boldsymbol{E}_{\text{real}}(t) &= \frac{1}{2}\left[\boldsymbol{E}(t)\exp(-\mathrm{i}\omega_0 t) + \boldsymbol{E}^*(t)\exp(\mathrm{i}\omega_0 t)\right] \\
\boldsymbol{A}_{\text{real}}(t) &= \frac{1}{2}\left[\boldsymbol{A}(t)\exp(-\mathrm{i}\omega_0 t) + \boldsymbol{A}^*(t)\exp(\mathrm{i}\omega_0 t)\right].
\end{aligned} \tag{2.15}$$

The amplitudes are assumed to be slow that their time derivatives are negligible compared with the derivatives of the fast-varying exponents

$$|\omega_0 \boldsymbol{A}(t)| \gg \left|\frac{\partial}{\partial t}\boldsymbol{A}(t)\right| \Rightarrow \boldsymbol{A}(t) \overset{(2.2)}{\approx} \frac{1}{\mathrm{i}\omega_0}\boldsymbol{E}(t) \tag{2.16}$$

$$|\omega_0 \boldsymbol{P}(t)| \gg \left|\frac{\partial}{\partial t}\boldsymbol{P}(t)\right| \Rightarrow \frac{\partial}{\partial t}\boldsymbol{P}_{\text{real}}(t) \approx -\frac{\mathrm{i}\omega_0}{2}\left[\boldsymbol{P}(t)\exp(-\mathrm{i}\omega_0 t) - \boldsymbol{P}^*(t)\exp(\mathrm{i}\omega_0 t)\right]. \tag{2.17}$$

Then we can formulate the *classical* evolution of inversion

$$\begin{aligned}
\frac{\mathrm{d}I}{\mathrm{d}t} &\overset{(2.14)}{=} \frac{2}{E_a - E_b}\frac{1}{N}\boldsymbol{E}_{\text{real}}\frac{\partial}{\partial t}\boldsymbol{P}_{\text{real}} \\
&= \frac{2}{E_a - E_b}\frac{1}{N}\frac{\boldsymbol{E}^*(t)}{2}\frac{-\mathrm{i}\omega_0}{2}\boldsymbol{P}(t) + \text{c.c.} + \text{fast oscillations} \\
&= \frac{\omega}{2\mathrm{i}(E_a - E_b)N}\boldsymbol{E}^*(t)\boldsymbol{P}(t) + \text{c.c.} + \text{fast oscillations}.
\end{aligned} \tag{2.18}$$

Similarly we can formulate the *quantum* evolution of inversion

$$\begin{aligned}
\frac{\mathrm{d}I}{\mathrm{d}t} &\overset{(2.11)}{=} \frac{2e}{\hbar^2}\boldsymbol{A}_{\text{real}}(E_a - E_b)\left\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\right\rangle a^* b + \text{c.c.} \\
&\overset{(2.13)}{\underset{(2.15)}{=}} \frac{e}{\hbar^2}\frac{\mathrm{i}}{\omega_0}(E_a - E_b)\left\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\right\rangle \boldsymbol{E}^*(t)\exp(\mathrm{i}\omega_0 t)ab^* + \text{c.c.} + \text{fast oscillations}.
\end{aligned} \tag{2.19}$$

If we now compare the slow parts containing $\boldsymbol{E}^*(t)$ in the classical and quantum formulations

$$\frac{e}{\hbar^2}\frac{\mathrm{i}}{\omega_0}(E_a - E_b)\left\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\right\rangle \boldsymbol{E}^*(t)\exp(\mathrm{i}\omega_0 t)ab^* \approx \frac{\omega_0}{2\mathrm{i}(E_a - E_b)N}\boldsymbol{E}^*(t)\boldsymbol{P}(t). \tag{2.20}$$

The slowly varying envelope of the optical polarization is then

$$\boldsymbol{P} = -2eN\underbrace{\left(\frac{E_a - E_b}{\hbar\omega_0}\right)^2}_{\approx 1}\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle\, ab^* \exp(\mathrm{i}\omega_0 t). \tag{2.21}$$

Here we assume a resonant excitation which means that the frequency detuning $\Delta\omega$ is small

$$\Delta\omega = \frac{E_a - E_b}{\hbar} - \omega \ll \omega_0. \tag{2.22}$$

The total or real valued polarization then is

$$\boldsymbol{P}_{\text{real}} = \frac{1}{2}\left[\boldsymbol{P}(t)\exp(-\mathrm{i}\omega_0 t) + \text{c.c.}\right] = -eN\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle\, ab^* + \text{c.c.} \tag{2.23}$$

Now we also want to discuss the dynamics of the nonlinear polarization by using (2.6)

$$\begin{aligned}
\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}(ab^*) &= \left(\mathrm{i}\hbar\frac{\mathrm{d}a}{\mathrm{d}t}\right)b^* - a\left(\mathrm{i}\hbar\frac{\mathrm{d}b}{\mathrm{d}t}\right)^* \\
&= (E_a - E_b)ab^* - \langle\psi_a|\hat{W}|\psi_b\rangle\left(|a|^2 - |b|^2\right) \\
&= (E_a - E_b)ab^* - \boldsymbol{A}_{\text{real}}\frac{\mathrm{i}e}{\hbar}(E_a - E_b)\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle\, I \\
&\approx (E_a - E_b)ab^* - \frac{1}{2\mathrm{i}\omega_0}\left[\boldsymbol{E}(t)\mathrm{e}^{-\mathrm{i}\omega_0 t} - \boldsymbol{E}^*(t)\mathrm{e}^{\mathrm{i}\omega_0 t}\right]\frac{\mathrm{i}e}{\hbar}(E_a - E_b)\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle\, I \\
&= (E_a - E_b)ab^* - \frac{e(E_a - E_b)}{2\hbar\omega_0}\left[\boldsymbol{E}(t)\mathrm{e}^{-\mathrm{i}\omega_0 t} - \boldsymbol{E}^*(t)\mathrm{e}^{\mathrm{i}\omega_0 t}\right]\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle\, I.
\end{aligned} \tag{2.24}$$

The driving term of ab^* with the wrong frequency ($\sim \mathrm{e}^{+\mathrm{i}\omega_0 t}$) is neglected. The evolution of the polarization can be derived by using (2.24), (2.21) and (2.22)

$$\mathrm{i}\frac{\mathrm{d}\boldsymbol{P}}{\mathrm{d}t} = \Delta\omega\boldsymbol{P} + \frac{Ne^2}{\hbar}I\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle(\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle\boldsymbol{E}). \tag{2.25}$$

For an isotropic medium with linear polarization of the electric field, the polarization $\boldsymbol{P}$ is parallel to $\boldsymbol{E}$ which leads to

$$\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle(\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle\boldsymbol{E}) = |\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}. \tag{2.26}$$

Then the final set of equations can be summarized to

Equations of motion

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{P} = \Delta\omega\boldsymbol{P} + \frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}I \tag{2.27}$$

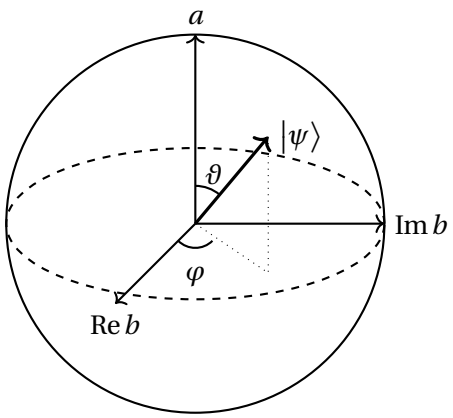
$$\frac{\mathrm{d}I}{\mathrm{d}t} = \frac{1}{2\mathrm{i}\hbar N}\boldsymbol{E}^*\boldsymbol{P} + \text{c.c.} \tag{2.28}$$

Note that up to now the equations of motion contain no damping, because we treated the optical field classically thus omitting spontaneous emission. Equations (2.27) and (2.28) are strictly valid only in time domains which are small compared with classical relaxation times.

Conservation law of Bloch equations and the Bloch sphere

As stated before we can describe the state of the two-level-system as

$$\left|\psi(t)\right\rangle = a(t)\left|\psi_a\right\rangle + b(t)\left|\psi_b\right\rangle. \tag{2.29}$$



Furthermore we have the conservation of the norm $|a(t)|^2 + |b(t)|^2 = 1$ and no interest in absolute phases which means we can set a to a real value. Now we may plot the space of the two-level state as a function of a, $\mathrm{Re}(b)$ and $\mathrm{Im}(b)$ on the surface of the Bloch-sphere (sphere with radius 1).

We can now express the norm in terms of slowly varying envelope of the polarization and inversion

$$\begin{aligned}(|a|^2+|b|^2)^2 &= |a|^4 + |b|^4 - 2|a|^2|b|^2 + 4|a|^2|b|^2 = (|a|^2-|b|^2)^2 + 4|ab^*|^2 \\ &= I^2 + \frac{1}{e^2N^2|\left\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\right\rangle|^2}|\boldsymbol{P}|^2 = 1\end{aligned} \tag{2.30}$$

and find this conserved quantity connecting inversion and polarization. Here we used

$$\boldsymbol{P} = -2eN\left\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\right\rangle ab^* \exp(\mathrm{i}\omega_0 t).$$

Inversion and the scalar component of the normalized slowly varying envelope of the polarization are on the surface of the Bloch sphere.

2.2 Rabi oscillations

In what follows we solve the system of equations (2.27) and (2.28) for a linearly polarized optical field oscillating with the frequency of the transition $\Delta\omega = 0$ with constant amplitude, but a step-like envelope (rectangular pulse)

$$\boldsymbol{E}(t) = E_0\hat{\mathbf{e}}\,\Theta(t)\,\Theta(t_1 - t), \tag{2.31}$$

where $\Theta(t)$ is the HEAVISIDE step function. We assume that for the initial conditions ($t = -\infty$) the system is purely in the ground state $I = -1$ and $\boldsymbol{P} = 0$.

a.) At first we look at $t < 0$ with $\boldsymbol{E} = 0$. Here we find

$$\frac{\mathrm{d}}{\mathrm{d}t}I = 0 \Rightarrow I = -1, \qquad \frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{P} = 0 \Rightarrow \boldsymbol{P} = 0. \tag{2.32}$$

The system remains in its ground state.

b.) Now we look at $0 < t < t_1$ with $|\boldsymbol{E}| = E_0 = \text{const.}$ (real valued). We can decompose the polarization into its real and imaginary parts

$$\boldsymbol{P} = [P_R(t) + \mathrm{i}P_i(t)]\hat{\mathbf{e}}. \tag{2.33}$$

Using this we find

$$\begin{aligned}
&\frac{\mathrm{d}}{\mathrm{d}t}P_R = 0 \Rightarrow P_R(t) = 0 \\
&\frac{\mathrm{d}}{\mathrm{d}t}P_i = -\frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2 E_0 I \qquad (2.34)\\
&\frac{\mathrm{d}}{\mathrm{d}t}I = \frac{E_0}{\hbar N}P_i. \qquad (2.35)
\end{aligned}$$

Differentiating the third equation we find an equation for a harmonic motion

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}I + \Omega^2 I = 0 \qquad \boxed{\Omega = \frac{e}{\hbar}E_0|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|} \quad \text{Rabi frequeny.} \tag{2.36}$$

The whole system performs oscillations with the Rabi frequency Ω. The solution of the system is then

$$I(t) = -\cos(\Omega t), \qquad P_i = \frac{\hbar N}{E_0}\frac{\mathrm{d}I}{\mathrm{d}t} = \frac{\hbar N}{E_0}\Omega\sin(\Omega t). \tag{2.37}$$

c.) The last part is $t > t_1$ with $\boldsymbol{E} = 0$. Here we find again

$$\begin{aligned}
&\frac{\mathrm{d}}{\mathrm{d}t}P_i = 0 \Rightarrow P_i(t) = \frac{\hbar N}{E_0}\Omega\sin(\Omega t_1) \quad \text{stationary envelope} \\
&\frac{\mathrm{d}}{\mathrm{d}t}I = -\cos(\Omega t_1). \qquad (2.38)
\end{aligned}$$

The real valued polarization is then

$$\boldsymbol{P}_{\text{real}}(t) = \mathrm{i}\frac{\hat{\mathbf{e}}}{2}\left[P_i(t)\exp(-\mathrm{i}\omega_0 t) - P_i^*(t)\exp(\mathrm{i}\omega_0 t)\right]. \tag{2.39}$$

After the end of the excitation the polarization keeps on oscillating forever. But, any real system is damped, an effect we have omitted by describing the optical field classically. Hence above solution is only valid in a time range short compared with the relaxation time, which can range from 100 fs in solids up to ms in dilute gases.

We conclude the results from the solution:

- Polarization and electric field need not to be proportional.
- The maximum strength of the polarization need not depend on the electric field strength (see equation (2.37)).
- The polarization can be present, although the optical field is absent.

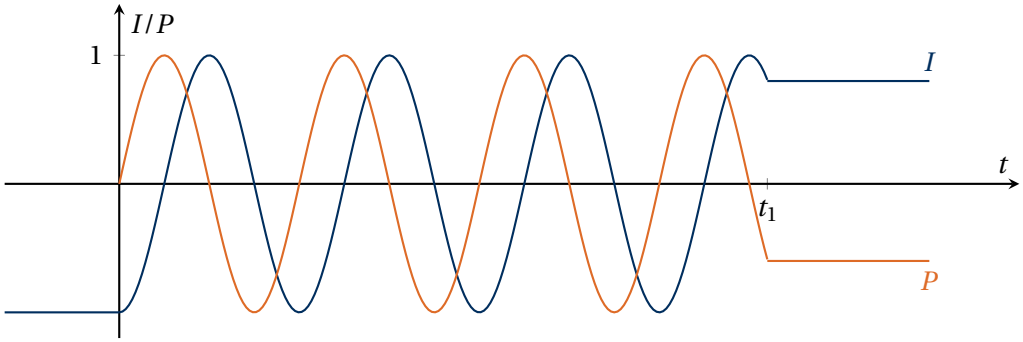


Fig. 1: Visualization of the the inversion (blue) and polarization (orange) for an electric field present at $0 \leq t \leq t_1$.

- Any kind of susceptibility becomes meaningless in the time range of Rabi-oscillations.
- A two-level-system can be brought to complete inversion by a single very short laser pulse.
- For a correctly tailored optical pulse the system can finally return to the ground state which means that no energy is absorbed (transparent medium). This behaviour can be reproduced for more complex pulse shapes leading to self-induced-transparency-solitons.

2.3 The influence of damping

Damping is always present in real world systems. It is caused by the coupling to a continuum of modes, which could be modes of the electromagnetic radiation, phonons or plasmons. Its exact calculation requires a quantum mechanical description of the respective field. Here we just introduce some phenomenological constants into respective evolution equations.

Damped equations of motion

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{P} = \left(\Delta\omega - \frac{\mathrm{i}}{T_2}\right)\boldsymbol{P} + \frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}I \tag{2.40}$$

$$\frac{\mathrm{d}I}{\mathrm{d}t} = \frac{1}{2\mathrm{i}\hbar N}(\boldsymbol{E}^*\boldsymbol{P} - \boldsymbol{E}\boldsymbol{P}^*) - \frac{I+1}{T_0}, \tag{2.41}$$

where T_2 describes the relaxation time of the polarization and T_0 the life time of the upper state. Note that the relaxation term of the inversion was chosen such that I relaxes towards the equilibrium state -1.

We can describes different scenarios using these relaxations constants:

- Free atom/atom in a trap:
  Here, relaxation is due to spontaneous emission with rather long relaxation times (ms). The relaxation is strictly correlated $T_2 = 2T_0$.

- Dense systems:
  These are gases under normal pressure, liquids or solids which will be considered in the following. They show a strong mutual interaction with many decay channels. The polarization is dominantly affected by so-called “dephasing”. Therefore T_2 is orders of magnitude shorter than T_0, e. g. in a direct semiconductor: $T_2 \sim 100\,\mathrm{fs}$, $T_0 \sim \mathrm{ns}$.

We can now distinguish different cases according to the actual pulse duration T_p:

a.) $T_p < T_2 < T_0$ (coherent regime)

During the optical pulse we observe Rabi-oscillations. After the optical pulse the polarization still oscillates, but decays. The system finally relaxes to the ground state.

b.) $T_2 < T_p < T_0$

Here the dephasing of the polarization is faster than the actual pulse propagates through the medium. This means the Rabi oscillations have already disappeared and the evolution of $\boldsymbol{P}(t)$ follows that of $\boldsymbol{E}(t)$. We can estimate the absolute values of the terms in equation (2.40) as

$$\left|\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{P}\right| \approx \left|\frac{\boldsymbol{P}}{T_p}\right| \ll \left|\frac{\boldsymbol{P}}{T_2}\right|. \tag{2.42}$$

We can now neglect the time derivative of $\boldsymbol{P}(t)$ which leads to an algebraic equation for the polarization

$$\begin{aligned} 0 &= \left(\Delta\omega - \frac{\mathrm{i}}{T_2}\right)\boldsymbol{P} + \frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}I \\ \Rightarrow \boldsymbol{P}(t) &= -\frac{\frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2}{\Delta\omega - \frac{\mathrm{i}}{T_2}} I\boldsymbol{E}(t). \end{aligned} \tag{2.43}$$

The polarization is determined by the actual optical field. Now we can also find the effective inversion dependent susceptibility

$$\boldsymbol{P}(t) = \varepsilon_0\chi(t)\boldsymbol{E}(t) \quad \text{with} \quad \chi(I) = -\underbrace{\frac{Ne^2}{\varepsilon_0\hbar}\frac{|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2}{\Delta\omega - \frac{\mathrm{i}}{T_2}}}_{=:\chi_0} I. \tag{2.44}$$

The polarization has lost its independence. It has become *enslaved*. We can now also describe the evolution of the inversion

$$\begin{aligned} \frac{\mathrm{d}I}{\mathrm{d}t} &= \frac{1}{2\mathrm{i}\hbar N}(\boldsymbol{E}^*\boldsymbol{P} - \boldsymbol{E}\boldsymbol{P}^*) - \frac{I+1}{T_0} = \frac{\varepsilon_0}{\hbar N}\mathrm{Im}(\chi(I))|\boldsymbol{E}|^2 - \frac{I+1}{T_0} \\ &\text{with} \quad \mathrm{Im}(\chi(I)) = -\frac{Ne^2}{\varepsilon_0\hbar}\frac{|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2}{\Delta\omega^2 + \frac{1}{T_2^2}}\frac{1}{T_2}I \sim I. \end{aligned} \tag{2.45}$$

We can rewrite the differential equation as a rate equation of $\mathrm{Im}[\chi(t)]$ by multiplying (2.45) with the prefactor of $\mathrm{Im}[\chi(t)]$

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathrm{Im}(\chi) = \underbrace{-\frac{\varepsilon_0}{\hbar N}\mathrm{Im}(\chi_0)\,\mathrm{Im}(\chi)|\boldsymbol{E}|^2}_{(1)} \underbrace{-\frac{\mathrm{Im}(\chi)-\mathrm{Im}(\chi_0)}{T_0}}_{(2)}, \tag{2.46}$$

with χ_0 being the susceptibility in the ground state. The first term (1) describes absorption ($\mathrm{Im}\,\chi_0 > 0$) induced bleaching of the transition by reducing $\mathrm{Im}\,\chi$. It switches off, when $\mathrm{Im}\,\chi \to 0$. The second term (2) describes the relaxation towards the ground state.

The complete susceptibility induced by the two-level-system is then

$$\chi = \frac{\Delta\omega^2 + \frac{1}{T_2^2}}{\Delta\omega - \frac{\mathrm{i}}{T_2}} T_2\,\mathrm{Im}(\chi). \tag{2.47}$$

Finally lets look at the situation when the optical field is constant after the Rabi oscillations have decayed ($t \to \infty$):

- We are in a steady state with $\mathrm{Im}\,\chi > 0$.
- Almost zero inversion ($I \to \infty$) can be reached where the system is almost transparent. A complete transparency is impossible, as absorption is switched off close to transparency. Carrier relaxation represented by T_0 prevents the system from reaching zero inversion.
- A state of positive inversion cannot be attained.

c.) $T_2 < T_0 \ll T_p$ (inversion enslaved)

In this situation the time derivative the the rate equations can be neglected $\frac{\mathrm{d}I}{\mathrm{d}t} = 0 \Rightarrow \frac{\mathrm{d}}{\mathrm{d}t}\mathrm{Im}(\chi) = 0$. The imaginary part of the susceptibility is a direct function of the acting optical field. Using equation (2.46) we find

$$\begin{aligned}
\mathrm{Im}(\chi) &= \frac{\mathrm{Im}(\chi_0)}{1+\frac{T_0\varepsilon_0}{\hbar N}\mathrm{Im}(\chi_0)|\boldsymbol{E}|^2} && \text{replace with intensity} \\
&= \frac{\mathrm{Im}(\chi_0)}{1+\frac{2T_0}{c\hbar N n}\mathrm{Im}(\chi_0)I_0} && I_0 = \frac{\varepsilon_0}{2}cn|\boldsymbol{E}|^2 \\
&= \frac{\mathrm{Im}(\chi_0)}{1+\frac{I_0}{I_S}} && \text{saturation intensity } I_S = \frac{N\hbar c\, n}{2T_0\,\mathrm{Im}(\chi_0)}.
\end{aligned} \tag{2.48}$$

Real and imaginary part of the susceptibility are proportional to each other (c. f. equation (2.47))

$$\chi(\omega) = \frac{\chi_0(\omega)}{1+\frac{I_0}{I_S}} \qquad I_S = \frac{N\hbar c\, n}{2T_0\,\mathrm{Im}(\chi_0)}. \tag{2.49}$$

This is the simplest formulation describing the response of an optical transition. It is only valid for time scales much longer than all relaxation times of the material. It describes a single transition, to which the optical field is resonant. We only require two quantities for the whole description:

- The ground state susceptibility $\chi_0(\omega)$ of the two-level-system (usually only a small fraction of the complete susceptibility)
- The saturation intensity I_S.

Equation (2.48) is not equivalent to the expressions derived in chapter (1.5.2) (see e. g. equation (1.52)). It is not only highly dispersive, but also a Taylor expansion with respect to the optical intensity yielding such quantities as e. g. n_2 will only converge in a limited range of intensities. If the expansion is done at $I_0 = 0$

$$\chi \approx \chi_0 - \chi_0 \frac{I_0}{I_S} \tag{2.50}$$

it will diverge for intensities higher than I_s and will give wrong results much earlier.

3 Nonlinear optics and field propagation - a plane wave approach

In the last chapter we have learned about the nonlinear response of the material. Together with MAXWELL's equations we can now in principle determine any kind of field propagation in the presence of a nonlinear response. Unfortunately, the complete set of equations including all nonlinear interaction is much too difficult to be solved in a self-consistent way. Hence, approximations have to be made which are usually based on the assumption that the nonlinear response of the material is small compared with the linear one and that some peculiar features of the geometry allow for further simplifications. A few examples of such treatments will be presented in that chapter.

3.1 Perturbation theory for nonlinear solutions

First we start with some *assumptions*:

- *Monochromatic fields*: The fields consist of a single frequency ω. If required, e. g. for frequency conversion processes, several discrete frequencies are taken into account. $\boldsymbol{E}_{\text{real}}, \boldsymbol{B}_{\text{real}}, \boldsymbol{P}_{\text{real}}$ take the same form namely

$$\boldsymbol{E}_{\text{real}}(\boldsymbol{r}, t) = \frac{1}{2}\left(\boldsymbol{E}(\boldsymbol{r})\mathrm{e}^{-\mathrm{i}\omega t} + \boldsymbol{E}^*(\boldsymbol{r})\mathrm{e}^{\mathrm{i}\omega t}\right). \tag{3.1}$$

- The material polarization at a particular frequency is split into a linear and a nonlinear part representing the linear part with the dielectric tensor $\hat{\varepsilon}$

$$\boldsymbol{P}(\boldsymbol{r}) = \boldsymbol{P}^{\mathrm{L}}(\boldsymbol{r}) + \boldsymbol{P}^{\mathrm{NL}}(\boldsymbol{r}) = \varepsilon_0(\hat{\varepsilon}(\boldsymbol{r}) - \mathbb{1})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}^{\mathrm{NL}}(\boldsymbol{r}). \tag{3.2}$$

- We further assume a linearly homogeneous, but not yet isotropic system

$$\hat{\varepsilon}(\omega, \boldsymbol{r}) = \hat{\varepsilon}(\omega). \tag{3.3}$$

  Remark: An index profile as it is typical for waveguides can be treated in a similar manner by taking waveguide modes instead of plane waves into account.

- Small perturbation due to nonlinear action: $|\boldsymbol{P}^{\mathrm{NL}}(\boldsymbol{r})| \ll |\varepsilon_0\hat{\varepsilon}(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r})|$.

We want to briefly state MAXWELL's equations for monochromatic fields

$$\vec{\boldsymbol{\nabla}} \times \boldsymbol{E} = \mathrm{i}\omega\boldsymbol{B}, \quad \vec{\boldsymbol{\nabla}} \times \boldsymbol{B} = -\mathrm{i}\omega\mu_0(\varepsilon_0\hat{\varepsilon}\boldsymbol{E} + \boldsymbol{P}^{\mathrm{NL}}), \quad \vec{\boldsymbol{\nabla}} \cdot \left(\varepsilon_0\hat{\varepsilon}\boldsymbol{E} + \boldsymbol{P}^{\mathrm{NL}}\right) = 0, \quad \vec{\boldsymbol{\nabla}} \cdot \boldsymbol{B} = 0.$$

Taking the curl on FARADAY's law leads to the wave equation

$$\begin{aligned} \vec{\boldsymbol{\nabla}}\left(\vec{\boldsymbol{\nabla}} \cdot \boldsymbol{E}\right) - \Delta\boldsymbol{E} &= \frac{\omega^2}{c^2}\left(\hat{\varepsilon}\boldsymbol{E} + \frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}\right) \quad \text{with} \quad \varepsilon_0\mu_0 = \frac{1}{c^2} \\ \underbrace{\left(\Delta - \vec{\boldsymbol{\nabla}}\mathrm{div} + \frac{\omega^2}{c^2}\hat{\varepsilon}\right)}_{\text{linear operator}}\boldsymbol{E} &= \underbrace{-\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}}_{\text{small perturbation}} . \end{aligned} \tag{3.4}$$

First we want to look at solutions $\boldsymbol{E}^{\mathrm{L}}(\boldsymbol{r})$ of the linear system. Then we can write $\boldsymbol{E}^{\mathrm{L}}(\boldsymbol{r})$ as an eigenvector of the eigenvalue problem

$$\hat{\varepsilon}^{-1}(\Delta - \vec{\boldsymbol{\nabla}}\mathrm{div})\boldsymbol{E}^{\mathrm{L}}(\boldsymbol{r}) = -\frac{\omega^2}{c^2}\boldsymbol{E}^{\mathrm{L}}(\boldsymbol{r}) \quad \text{with eigenvalue} \quad -\frac{\omega^2}{c^2}. \tag{3.5}$$

For a homogeneous infinite system we find the solution $\boldsymbol{E}^{\mathrm{L}}(\boldsymbol{r}) = \boldsymbol{E}^{\boldsymbol{k}}\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}$. This is the starting point to determine the nonlinear solution via perturbation theory.

Remark: One might be tempted to drop the term $\vec{\boldsymbol{\nabla}}\cdot\boldsymbol{E}^{\mathrm{L}}$, but this is only possible in homogeneous isotropic media. In anisotropic media as they are typical for a quadratic nonlinear response, the term $\vec{\boldsymbol{\nabla}}\cdot\boldsymbol{E}^{\mathrm{L}}$ is not necessarily zero and longitudinal electric fields may occur

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{pmatrix} \Rightarrow \vec{\boldsymbol{\nabla}}\cdot(\hat{\varepsilon}\boldsymbol{E}^{\mathrm{L}}) = \varepsilon_x\frac{\partial E_x}{\partial x} + \varepsilon_y\frac{\partial E_y}{\partial y} + \varepsilon_z\frac{\partial E_z}{\partial z} = 0$$

$$\not\Rightarrow \vec{\boldsymbol{\nabla}}\cdot\boldsymbol{E}^{\mathrm{L}} = \frac{\partial E_x}{\partial x} + \frac{\partial E_y}{\partial y} + \frac{\partial E_z}{\partial z} = 0. \tag{3.6}$$

We want to state requirements on nonlinearly induced processes (e. g. frequency conversion):

- The spatial scales are much longer compared with linear process (e. g. conversion lengths $\gg$ wavelength)
- The complex amplitudes of linear solutions may change but not their structure (e. g. polarization).

The general structure of a nonlinear solution is

$$\boldsymbol{E}(\boldsymbol{r}) = \boldsymbol{E}^{\boldsymbol{k}}u(\boldsymbol{r})\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}). \tag{3.7}$$

In what follows we discuss several cases for which further assumptions can be made.

3.2 Propagation in z-direction only

In many relevant case (e. g. frequency conversion) very broad collimated beams, which travel into a single direction are used and any dependence on transverse coordinates (x, y) can be neglected or taken into account parametrically only. We assume for our solution to take the form

$$\boldsymbol{E}(\boldsymbol{r}) = \boldsymbol{E}^{\beta}u(z)\exp\bigl(\mathrm{i}\beta z\bigr). \tag{3.8}$$

The related magnetic induction is obtained by the linear solution $\boldsymbol{E}^{\beta}\exp\bigl(\mathrm{i}\beta z\bigr)$ via the curl

$$\vec{\boldsymbol{\nabla}} \times (\boldsymbol{E}^{\beta}\exp\bigl(\mathrm{i}\beta z\bigr)) = (\mathrm{i}\beta\hat{\mathbf{e}}_z) \times (\boldsymbol{E}^{\beta}\exp\bigl(\mathrm{i}\beta z\bigr)) = \mathrm{i}\omega\boldsymbol{B}^{\beta}\exp\bigl(\mathrm{i}\beta z\bigr)$$

$$\boldsymbol{B}^{\beta} = \frac{\beta}{\omega}\hat{\mathbf{e}}_z \times \boldsymbol{E}^{\beta}. \tag{3.9}$$

Now we want to insert our ansatz (3.8) into (3.4) where we use that $u(z)$ is a slowly varying envelope with

$$\left|\frac{\mathrm{d}}{\mathrm{d}z}u(z)\right| \ll |\beta u(z)|. \tag{3.10}$$

This leads us to

$$\begin{aligned}
&\left(\Delta - \vec{\nabla}\mathrm{div} + \frac{\omega^2}{c^2}\hat{\varepsilon}\right)[\boldsymbol{E}^{\boldsymbol{k}}u(\boldsymbol{r})\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})] \\
&= \left[(\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)\frac{\mathrm{d}^2}{\mathrm{d}z^2} + \frac{\omega^2}{c^2}\hat{\varepsilon}\boldsymbol{E}^\beta\right](u(z)\exp(\mathrm{i}\beta z)) \\
&= u(z)\left[(\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)\frac{\mathrm{d}^2}{\mathrm{d}z^2} + \frac{\omega^2}{c^2}\hat{\varepsilon}\boldsymbol{E}^\beta\right]\exp(\mathrm{i}\beta z) + (\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)\exp(\mathrm{i}\beta z)\left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} + 2\mathrm{i}\beta\frac{\mathrm{d}}{\mathrm{d}z}\right)u(z) \\
&= u(z)\underbrace{\left(\Delta - \vec{\nabla}\mathrm{div} + \frac{\omega^2}{c^2}\hat{\varepsilon}\right)\boldsymbol{E}^\beta}_{=0\text{ linear solution}}\exp(\mathrm{i}\beta z) + (\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)\exp(\mathrm{i}\beta z)\frac{\mathrm{d}}{\mathrm{d}z}\left(\frac{\mathrm{d}}{\mathrm{d}z} + 2\mathrm{i}\beta\right)u(z) \\
&= (\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)\exp(\mathrm{i}\beta z)\frac{\mathrm{d}}{\mathrm{d}z}\left(\frac{\mathrm{d}}{\mathrm{d}z} + 2\mathrm{i}\beta\right)u(z) = -\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}.
\end{aligned} \tag{3.11}$$

Assuming the amplitude to vary slowly we can simplify

$$\left(\frac{\mathrm{d}}{\mathrm{d}z} + 2\mathrm{i}\beta\right)u(z) \approx 2\mathrm{i}\beta u(z) \tag{3.12}$$

and multiplying a suitable vector from the left

$$\begin{aligned}
(\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)\exp(\mathrm{i}\beta z)2\mathrm{i}\beta\frac{\mathrm{d}}{\mathrm{d}z}u(z) &= -\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}} \\
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}u(z) &= -\frac{\omega^2}{c^2}\frac{\exp(-\mathrm{i}\beta z)}{2\beta\varepsilon_0}\frac{(\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)^*}{|\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z|^2}\boldsymbol{P}^{\mathrm{NL}}.
\end{aligned} \tag{3.13}$$

As the amplitude $u(z)$ has currently no unit, we change the unit to $\sqrt{\frac{\mathrm{W}}{\mathrm{m}^2}}$ that the squared amplitude represents the intensity flowing in z-direction

$$a(z) = \sqrt{\left\langle s_z^\beta\right\rangle}u(z) \tag{3.14}$$

with $\left\langle s_z^\beta\right\rangle$ being the z-component of the time averaged POYNTING vector of the linear solution which we want to calculate in the following:

$$\begin{aligned}
\left\langle s_z^\beta\right\rangle &= \frac{1}{2\mu_0}\mathrm{Re}(\boldsymbol{E}\times\boldsymbol{B}^*)_z \\
&= \frac{1}{2\mu_0}\mathrm{Re}[\boldsymbol{E}^\beta\cancel{\exp(\mathrm{i}\beta z)}\times\boldsymbol{B}^{\beta*}\cancel{\exp(-\mathrm{i}\beta z)}]_z \\
&= \frac{1}{2\mu_0}\mathrm{Re}\left[\boldsymbol{E}^\beta\times\left(\frac{\beta}{\omega}\hat{\mathbf{e}}_z\times\boldsymbol{E}^{\beta*}\right)\right]_z = \frac{1}{2\mu_0}\frac{\beta}{\omega}\mathrm{Re}\left[|\boldsymbol{E}^\beta|^2\hat{\mathbf{e}}_z - \boldsymbol{E}^\beta(\boldsymbol{E}^{\beta*}\hat{\mathbf{e}}_z)\right]_z \\
&= \frac{1}{2\mu_0}\frac{\beta}{\omega}\left[|\boldsymbol{E}^\beta|^2 - |E_z^\beta|^2\right] = \frac{1}{2\mu_0}\frac{\beta}{\omega}\left|\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z\right|^2.
\end{aligned} \tag{3.15}$$

We can summarize our findings of the nonlinearly driven evolution of a mode in z-direction

Evolution in z-direction

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}u(z) = -\frac{\omega^2}{c^2}\frac{1}{2\beta\varepsilon_0}\frac{(\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)^*}{|\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z|^2}\boldsymbol{P}^{\mathrm{NL}}\exp\left(-\mathrm{i}\beta z\right) \tag{3.16}$$

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}a(z) = -\sqrt{\frac{\omega^3}{8\beta\varepsilon_0 c^2}}\frac{(\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z)^*}{|\boldsymbol{E}^\beta - E_z^\beta\hat{\mathbf{e}}_z|}\boldsymbol{P}^{\mathrm{NL}}\exp\left(-\mathrm{i}\beta z\right). \tag{3.17}$$

3.3 Diffraction in linearly isotropic media

To describe effects of a nonlinearly induced change of the beam profile one has to take into account the effect of transverse coordinates to account for diffraction, self-focusing and spatial soliton formation. To simplify the analysis, we restrict to isotropic media, which are most relevant for cubic nonlinearities.

The starting point is again equation (3.4) with a scalar valued dielectric function $\hat{\varepsilon} = \varepsilon$

$$\left(\Delta - \vec{\boldsymbol{\nabla}}\mathrm{div} + \frac{\omega}{c^2}\varepsilon\right)\boldsymbol{E} = -\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}. \tag{3.18}$$

Again we make the assumption of a weak nonlinear action $|\boldsymbol{P}^{\mathrm{NL}}| \ll |\varepsilon_0\varepsilon\boldsymbol{E}|$ and homogeneity $\vec{\boldsymbol{\nabla}}\varepsilon = 0$ and use the first of MAXWELL's equations

$$\begin{aligned} 0 = \vec{\boldsymbol{\nabla}}\cdot\boldsymbol{D} &= \vec{\boldsymbol{\nabla}}\cdot\left[\varepsilon_0\varepsilon\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}^{\mathrm{NL}}\right] \\ &\approx \vec{\boldsymbol{\nabla}}\cdot\left[\varepsilon_0\varepsilon\boldsymbol{E}(\boldsymbol{r})\right] = \varepsilon_0\varepsilon\vec{\boldsymbol{\nabla}}\cdot\boldsymbol{E}(\boldsymbol{r}) = 0. \end{aligned} \tag{3.19}$$

Then we can simplify (3.18) to

$$\left(\Delta + \frac{\omega}{c^2}\frac{1}{\varepsilon}\right)\boldsymbol{E} = -\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}. \tag{3.20}$$

Propagation mainly in z-direction

The field shape is determined by the respective linear wave $\boldsymbol{E}^{\mathrm{L}}(\boldsymbol{r}) = \boldsymbol{E}^\beta\exp\left(\mathrm{i}\beta z\right)$ with $\beta^2 = \varepsilon\frac{\omega^2}{c^2}$. The nonlinear wave takes the form

$$\boldsymbol{E}(\boldsymbol{r}) = \boldsymbol{E}^\beta u(x,y,z)\exp\left(\mathrm{i}\beta z\right) \quad \text{with} \quad \left|\frac{\partial u}{\partial z}\right| \ll |\beta\, u(x,y,z)|, \tag{3.21}$$

where $u(x,y,z)$ is the slowly varying envelope. $\boldsymbol{E}^\beta$ is the polarization of a linear wave propagating in z-direction which is transverse (no z-component).

Derivation of the evolution of $u(x,y,z)$

We can find the evolution equation by inserting (3.21) into (3.20)

$$\begin{aligned}
&\left(\Delta+\frac{\omega^2}{c^2}\varepsilon\right)[\boldsymbol{E}^\beta u(x,y,z)\exp(\mathrm{i}\beta z)]\\
&=\boldsymbol{E}^\beta\exp(\mathrm{i}\beta z)\left(\cancel{-\beta^2}+2\mathrm{i}\beta\frac{\partial}{\partial z}+\frac{\partial^2}{\partial z^2}+\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+\cancel{\frac{\omega^2}{c^2}\varepsilon}\right)u(x,y,z)\\
&=\boldsymbol{E}^\beta\exp(\mathrm{i}\beta z)\left[\frac{\mathrm{d}}{\mathrm{d}z}\left(\cancel{\frac{\mathrm{d}}{\mathrm{d}z}}+2\mathrm{i}\beta\right)+\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right]u(x,y,z)\\
&=\boldsymbol{E}^\beta\exp(\mathrm{i}\beta z)\left[2\mathrm{i}\beta\frac{\mathrm{d}}{\mathrm{d}z}+\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right]u(x,y,z)=-\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}.
\end{aligned}\tag{3.22}$$

We again assumed a slow varying amplitude. Similarly to section 3.2 we can multiply a suitable vector from the left and rearrange

$$\left[\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}+\frac{1}{2\beta}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)\right]u(x,y,z)=-\frac{\omega^2}{c^2}\frac{1}{2\beta\varepsilon_0}\exp(-\mathrm{i}\beta z)\frac{\boldsymbol{E}^{\beta*}}{|\boldsymbol{E}^\beta|^2}\boldsymbol{P}^{\mathrm{NL}}.\tag{3.23}$$

Furthermore we can apply the transformation $a=\sqrt{\left\langle s_z^\beta\right\rangle}u$ to arrive at intensities based on the z-component of the time averaged POYNTING vector of the linear solution $\left\langle s_z^\beta\right\rangle$ (c. f. (3.15))

$$\left\langle s_z^\beta\right\rangle=\frac{1}{2\mu_0}\frac{\beta}{\omega}|\boldsymbol{E}^\beta|^2.\tag{3.24}$$

Note that E_z^β is zero for isotropic media. We now obtain the evolution equation for nonlinearity driven diffraction

evolution for diffraction

$$\left[\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}+\frac{1}{2\beta}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)\right]a(x,y,z)=-\sqrt{\frac{\omega^3}{8\beta\varepsilon_0c^2}}\frac{\boldsymbol{E}^{\beta*}}{|\boldsymbol{E}^\beta|}\boldsymbol{P}^{\mathrm{NL}}(z)\exp(-\mathrm{i}\beta z).\tag{3.25}$$

3.4 Pulse evolution in linearly isotropic media

To describe effects of a nonlinearly induced change of a pulse profile one has to take into account a finite frequency spectrum and cannot restrict to monochromatic waves anymore to account for dispersive spreading, self-compression and temporal soliton formation. To simplify the analysis, we restrict to isotropic media and a propagation into a single direction thus neglecting diffractive spreading.

We start with equation (3.20) for monochromatic waves propagating in an isotropic medium with $\hat{\varepsilon}=\varepsilon$ under the action of a weak nonlinear action at a fixed frequency

$$\left(\Delta+\frac{\omega}{c^2}\varepsilon\right)\boldsymbol{E}(\omega,z)=-\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}(\omega,z).\tag{3.26}$$

As most of the quantities used in the further derivation are frequency dependent, we restrict to a fixed frequency ω_0 being in the middle of the investigated spectrum and representing the carrier frequency of the pulse.

We assume that the field shape is similar to that of a linear wave propagating at ω_0 for all relevant frequency components $\boldsymbol{E}^{\mathrm{L}}(\omega_0,\boldsymbol{r}) = \boldsymbol{E}^{\beta_0}\exp\left(\mathrm{i}\beta_0 z\right)$ with $\beta_0^2 = \varepsilon(\omega_0)\frac{\omega_0^2}{c^2}$. The nonlinear wave has the form

$$\boldsymbol{E}(\omega,\boldsymbol{r}) = \boldsymbol{E}^{\beta_0}u(\omega,z)\exp\left(\mathrm{i}\beta_0 z\right) \quad \text{with} \quad |\frac{\partial u}{\partial z}| \ll |\beta\, u(\omega,z)| \tag{3.27}$$

with $\boldsymbol{E}^{\beta_0}$ being the polarization of a linear (transverse) wave propagating in z-direction at ω_0. $u(\omega,z)$ is again the slowly varying envelope. The spectrum is so narrow that the phase evolution along z is still well represented by β_0.

Derivation of the evolution of $u(\omega,z)$

We can find the evolution equation by inserting equation (3.27) into (3.26) and using the slowly-varying-envelope-approach

$$\begin{aligned}\left(\Delta+\frac{\omega^2}{c^2}\varepsilon(\omega)\right)\boldsymbol{E}(\omega,z) &\approx \boldsymbol{E}^{\beta_0}\mathrm{e}^{\mathrm{i}\beta_0 z}\left[2\mathrm{i}\beta_0\frac{\partial}{\partial z}+\frac{\omega^2}{c^2}\varepsilon(\omega)-\beta_0^2\right]u(\omega,z)\\ &= -\frac{\omega^2}{c^2}\frac{1}{\varepsilon_0}\boldsymbol{P}^{\mathrm{NL}}(\omega,z).\end{aligned} \tag{3.28}$$

Now we can multiply a suitable vector from the left and rearrange

$$\Big[\mathrm{i}\frac{\partial}{\partial z}+\underbrace{\frac{1}{2\beta_0}\left(\frac{\omega^2}{c^2}\varepsilon(\omega)-\beta_0^2\right)}_{(1)}\Big]u(\omega,z) = -\frac{\omega^2}{c^2}\frac{1}{2\beta_0\varepsilon_0}\mathrm{e}^{-\mathrm{i}\beta_0 z}\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}_0^{\beta}|^2}\boldsymbol{P}^{\mathrm{NL}}(\omega,z). \tag{3.29}$$

We prepare for a FOURIER-back-transform under the assumption that $\beta(\omega)=\frac{\omega^2}{c^2}\varepsilon(\omega)$ is close to β_0

$$(1) = \frac{\beta(\omega)+\beta_0}{2\beta_0}(\beta(\omega)-\beta_0) \approx \beta(\omega)-\beta_0 = \frac{1}{v_g}(\omega-\omega_0)+\frac{\mathrm{GVD}}{2}(\omega-\omega_0)^2+\ldots \tag{3.30}$$

where we use the expressions of group velocity v_g and group velocity dispersion GVD

$$v_g = \left[\frac{\partial}{\partial\omega}\beta(\omega)\Big|_{\omega_0}\right]^{-1}, \qquad \mathrm{GVD} = \frac{\mathrm{d}^2}{\mathrm{d}\omega^2}\beta(\omega)\Big|_{\omega_0}. \tag{3.31}$$

Now we integrate the different spectral components/monochromatic waves of the pulse

$$\begin{aligned}\boldsymbol{E}_{\mathrm{real}}(z,t) &= \int_0^\infty \mathrm{d}\omega\,\frac{1}{2}\left[\boldsymbol{E}(z,\omega)\mathrm{e}^{-\mathrm{i}\omega t}+\mathrm{c.c.}\right] = \mathrm{Re}\int_0^\infty \mathrm{d}\omega\left[\boldsymbol{E}(z,\omega)\mathrm{e}^{-\mathrm{i}\omega t}\right]\\ &= \mathrm{Re}\left[\int_0^\infty \mathrm{d}\omega\,\boldsymbol{E}(z,\omega)\mathrm{e}^{-\mathrm{i}(\omega-\omega_0)t}\mathrm{e}^{-\mathrm{i}\omega_0 t}\right].\end{aligned} \tag{3.32}$$

Now we can insert our ansatz (3.27) and find

$$\begin{aligned}\boldsymbol{E}_{\text{real}}(z,t) &= \text{Re}\left[\boldsymbol{E}^{\beta_0}\int_0^\infty \underbrace{\text{d}\omega\, u(\omega,z)\text{e}^{-\text{i}(\omega-\omega_0)t}}_{\text{Fourier transform of SVE}}\underbrace{\text{e}^{-\text{i}(\beta_0 z\omega_0 t)}}_{\text{carrier wave}}\right]\\ &= \text{Re}\left[\boldsymbol{E}^{\beta_0}\tilde{u}(z,t)\text{e}^{\text{i}(\beta_0 z-\omega_0 t)}\right],\end{aligned} \tag{3.33}$$

where we have the FOURIER transform of the slowly varying envelope as

$$\tilde{u}(z,t) = \int_0^\infty \text{d}\omega\, u(\omega,z)\text{e}^{-\text{i}(\omega-\omega_0)t}. \tag{3.34}$$

Now we can apply the FOURIER transform (3.34) on equation (3.29) using approximation (3.30)

$$\begin{aligned}&\int_0^\infty \text{d}\omega\left[\text{i}\frac{\partial}{\partial z}+\frac{1}{v_g}(\omega-\omega_0)+\frac{\text{GVD}}{2}(\omega-\omega_0)^2+\ldots\right]u(\omega,z)\text{e}^{-\text{i}(\omega-\omega_0)t}\\ &=\left[\text{i}\frac{\partial}{\partial z}+\frac{\text{i}}{v_g}\frac{\partial}{\partial t}-\frac{\text{GVD}}{2}\frac{\partial^2}{\partial t^2}+\ldots\right]\tilde{u}(t,z)\\ &=\frac{1}{2\beta_0\varepsilon_0 c^2}\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\int_0^\infty \text{d}\omega\left(-\omega^2\boldsymbol{P}^{\text{NL}}(\omega,z)\text{e}^{-\text{i}\omega t}\right)\text{e}^{-\text{i}(\beta_0 z-\omega_0 t)}\\ &=\frac{1}{2\beta_0\varepsilon_0 c^2}\left[\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\frac{\partial^2}{\partial t^2}\tilde{\boldsymbol{P}}^{\text{NL}}(z,t)\right]\text{e}^{-\text{i}(\beta_0 z-\omega_0 t)}.\end{aligned} \tag{3.35}$$

Remark: $\tilde{\boldsymbol{P}}^{\text{NL}}(z,t)$ is not the real valued nonlinear polarization, but its complex counterpart, the real part of which is the direct measurable polarization. It contains the positive frequency part only, but not its complex conjugate counterpart on the negative frequency side.

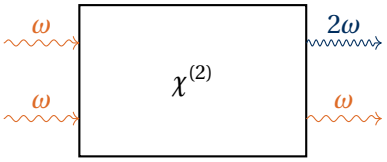
Again we can apply the transformation $a(z,t) = \sqrt{\left\langle s_z^{\beta_0}\right\rangle}\tilde{u}(z,t)$ to express the evolution in terms of intensities. As before $\left\langle s_z^{\beta_0}\right\rangle$ is the z-component of the time averaged POYNTING vector of the linear solution at the center frequency of the pulse ω_0. Again, we arrive at the evolution equation for the slowly varying envelope of a pulse

evolution for a slowly varying pulse envelope

$$\left[\text{i}\frac{\partial}{\partial z}+\frac{\text{i}}{v_g}\frac{\partial}{\partial t}-\frac{\text{GVD}}{2}\frac{\partial^2}{\partial t^2}+\ldots\right]a(t,z) = \frac{1}{\sqrt{8\beta_0 c^2\varepsilon_0\omega_0}}\left[\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\frac{\partial^2}{\partial t^2}\tilde{\boldsymbol{P}}^{\text{NL}}(z,t)\right]\text{e}^{-\text{i}(\beta_0 z-\omega_0 t)}. \tag{3.36}$$

4 Field evolution for quadratic nonlinearities

The main subject of this chapter will be *Second Harmonic generation* (SHG). The basic principle is that a fundamental harmonic (FH) wave at ω is frequency doubled to a second harmonic (SH) wave oscillating at 2ω.



The geometry consists of plane waves propagating in z-direction (no transverse structure). Note that the orientation of the coordinates is defined by the propagation direction and not by the nonlinear crystal. Because typical tensors of the nonlinear response are given with respect to the crystal axis, an additional coordinate transformation might be required.

4.1 Equations of motion

The relevant types of nonlinear polarization are the following:

$$\begin{aligned} P_i^{(2)}(\omega) &= \varepsilon_0 K(-\omega|2\omega,-\omega) \sum_{j,k=1}^{3} \chi_{ijk}^{(2)}(-\omega|2\omega,-\omega) E_{2\omega_j} E_{\omega_k}^* \quad \text{with} \quad K(-\omega|2\omega,-\omega) = 1 \\ P_i^{(2)}(2\omega) &= \varepsilon_0 K(-2\omega|\omega,\omega) \sum_{j,k=1}^{3} \chi_{ijk}^{(2)}(-2\omega|\omega,\omega) E_{\omega_j} E_{\omega_k} \quad \text{with} \quad K(-2\omega|\omega,\omega) = \frac{1}{2}. \end{aligned} \tag{4.1}$$

The interacting waves are one at FH frequency and one at SH frequency. We adapt (3.17) and obtain

$$\begin{aligned} \text{FH} \qquad & \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} a_\omega(z) = -\sqrt{\frac{\omega^3}{8\beta(\omega)\varepsilon_0 c^2}} \frac{\boldsymbol{E}_\perp^{\omega *}}{|\boldsymbol{E}_\perp^{\omega *}|} \boldsymbol{P}^{\mathrm{NL}}(z,\omega) \exp\bigl(-\mathrm{i}\beta(\omega) z\bigr) \\ \text{SH} \qquad & \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} a_{2\omega}(z) = -\sqrt{\frac{\omega^3}{\beta(2\omega)\varepsilon_0 c^2}} \frac{\boldsymbol{E}_\perp^{2\omega *}}{|\boldsymbol{E}_\perp^{2\omega *}|} \boldsymbol{P}^{\mathrm{NL}}(z,2\omega) \exp\bigl(-\mathrm{i}\beta(2\omega) z\bigr). \end{aligned} \tag{4.2}$$

Here $\boldsymbol{E}_\perp^{\omega/2\omega} = \boldsymbol{E}^{\omega/2\omega} - E_z^{\omega/2\omega}\hat{\mathbf{e}}_z$ is the transverse field polarization at FH/SH frequency.

The total electric field driving the nonlinear polarization can be found by using (3.8), (3.14) and (3.15)

$$\begin{aligned} \boldsymbol{E}_\omega(\boldsymbol{r}) &= \boldsymbol{E}^\omega u_\omega(z) \mathrm{e}^{\mathrm{i}\beta(\omega)z} = \boldsymbol{E}^\omega \frac{a_\omega(z)}{\sqrt{\langle s_z^\omega \rangle}} \mathrm{e}^{\mathrm{i}\beta(\omega)z} \\ &= \frac{\boldsymbol{E}^\omega}{|\boldsymbol{E}_\perp^\omega|} \sqrt{\frac{2\omega}{\varepsilon_0 c^2 \beta(\omega)}} a_\omega(z) \mathrm{e}^{\mathrm{i}\beta(\omega)z} \\ \boldsymbol{E}_{2\omega}(\boldsymbol{r}) &= \frac{\boldsymbol{E}^{2\omega}}{|\boldsymbol{E}_\perp^{2\omega}|} \sqrt{\frac{4\omega}{\varepsilon_0 c^2 \beta(2\omega)}} a_{2\omega}(z) \mathrm{e}^{\mathrm{i}\beta(2\omega)z}. \end{aligned} \tag{4.3}$$

We can now insert the fields (4.3) and the polarization (4.1) into the evolution equations (4.2)

$$\begin{aligned}
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}a_\omega(z) &= -\sqrt{\frac{\omega^3}{8\beta(\omega)c^2\varepsilon_0}}\frac{\mathrm{e}^{-\mathrm{i}\beta(\omega)z}}{|\boldsymbol{E}_\perp^\omega|}\sum_{i=x,y}E_i^{\omega *}P_i^{(2)}(\omega)\\
&= -\sqrt{\frac{\omega^3}{8\beta(\omega)c^2}}\frac{\mathrm{e}^{-\mathrm{i}\beta(\omega)z}}{|\boldsymbol{E}_\perp^\omega|}\sum_{i=x,y}E_i^{\omega *}\sum_{j,k=1}^{3}\chi_{ijk}^{(2)}(-\omega|2\omega,-\omega)E_{2\omega_j}E_{\omega_k}^*\\
&= -\sqrt{\frac{\omega^3}{8\beta(\omega)c^2}}\frac{\mathrm{e}^{-\mathrm{i}\beta(\omega)z}}{|\boldsymbol{E}_\perp^\omega|}\sum_{i=x,y}E_i^{\omega *}\sum_{j,k=1}^{3}\chi_{ijk}^{(2)}(-\omega|2\omega,-\omega)\\
&\quad\cdot\frac{E_j^{2\omega}}{|\boldsymbol{E}_\perp^{2\omega}|}\sqrt{\frac{4\omega}{\varepsilon_0c^2\beta(2\omega)}}a_{2\omega}(z)\mathrm{e}^{\mathrm{i}\beta(2\omega)z}\frac{E_k^{\omega *}}{|\boldsymbol{E}_\perp^\omega|}\sqrt{\frac{2\omega}{\varepsilon_0c^2\beta(\omega)}}a_\omega(z)^*\mathrm{e}^{-\mathrm{i}\beta(\omega)z}\\
&= -\mathrm{e}^{\mathrm{i}[\beta(2\omega)-2\beta(\omega)]z}\underbrace{\sqrt{\frac{\omega^5}{\beta(\omega)^2\beta(2\omega)\varepsilon_0c^6}}\sum_{i,j,k=1}^{3}\chi_{ijk}^{(2)}\frac{E_{\perp i}^{\omega *}}{|\boldsymbol{E}_\perp^\omega|}\frac{E_k^{\omega *}}{|\boldsymbol{E}_\perp^\omega|}\frac{E_j^{2\omega *}}{|\boldsymbol{E}_\perp^{2\omega}|}}_{=:\chi_{\mathrm{FH}}^{(2)}}a_{2\omega}a_\omega^*.
\end{aligned}\tag{4.4}$$

Analogously we find the evolution equation for $a_{2\omega}(z)$

$$\begin{aligned}
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}a_\omega(z) &= -\sqrt{\frac{\omega^3}{\beta(2\omega)c^2\varepsilon_0}}\frac{\mathrm{e}^{-\mathrm{i}\beta(2\omega)z}}{|\boldsymbol{E}_\perp^{2\omega}|}\sum_{i=x,y}E_i^{2\omega *}P_i^{(2)}(2\omega)\\
&= -\sqrt{\frac{\omega^3}{4\beta(2\omega)c^2}}\frac{\mathrm{e}^{-\mathrm{i}\beta(2\omega)z}}{|\boldsymbol{E}_\perp^{2\omega}|}\sum_{i=x,y}E_i^{2\omega *}\sum_{j,k=1}^{3}\chi_{ijk}^{(2)}(-2\omega|\omega,\omega)E_{\omega_j}E_{\omega_k}\\
&= -\mathrm{e}^{-\mathrm{i}[\beta(2\omega)-2\beta(\omega)]z}\underbrace{\sqrt{\frac{\omega^5}{\beta(\omega)^2\beta(2\omega)\varepsilon_0c^6}}\sum_{i,j,k=1}^{3}\chi_{ijk}^{(2)}\frac{E_{\perp i}^{2\omega *}}{|\boldsymbol{E}_\perp^{2\omega}|}\frac{E_k^{\omega}}{|\boldsymbol{E}_\perp^{\omega}|}\frac{E_j^{\omega}}{|\boldsymbol{E}_\perp^{\omega}|}}_{=:\chi_{\mathrm{SH}}^{(2)}}a_\omega^2(z).
\end{aligned}\tag{4.5}$$

For further simplifications we introduce effective nonlinear coefficients of fundamental and second harmonic as

$$\begin{aligned}
\chi_{\mathrm{FH}}^{(2)} &= \frac{\omega^2}{\beta(\omega)c^3}\sqrt{\frac{\omega}{\beta(2\omega)\varepsilon_0}}\sum_{i,j,k=1}^{3}\chi_{ijk}^{(2)}(-\omega|2\omega,-\omega)\frac{E_{\perp i}^{\omega *}}{|\boldsymbol{E}_\perp^\omega|}\frac{E_k^{\omega *}}{|\boldsymbol{E}_\perp^\omega|}\frac{E_j^{2\omega *}}{|\boldsymbol{E}_\perp^{2\omega}|}\\
\chi_{\mathrm{SH}}^{(2)} &= \frac{\omega^2}{\beta(\omega)c^3}\sqrt{\frac{\omega}{\beta(2\omega)\varepsilon_0}}\sum_{i,j,k=1}^{3}\chi_{ijk}^{(2)}(-2\omega|\omega,\omega)\frac{E_{\perp i}^{2\omega *}}{|\boldsymbol{E}_\perp^{2\omega}|}\frac{E_k^{\omega}}{|\boldsymbol{E}_\perp^{\omega}|}\frac{E_j^{\omega}}{|\boldsymbol{E}_\perp^{\omega}|}.
\end{aligned}\tag{4.6}$$

Now we assume that no further fields are involved and energy is conserved (Manley-Rowe relations) which leads to the conservation of energy flux

$$\begin{aligned}
0 &= \frac{\mathrm{d}}{\mathrm{d}z}\left(|a_\omega|^2+|a_{2\omega}|^2\right) = \mathrm{i}a_\omega^*\left(-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}a_\omega\right)+\mathrm{i}a_{2\omega}^*\left(-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}a_{2\omega}\right)+\mathrm{c.c.}\\
&= \mathrm{i}a_\omega^*\left(\mathrm{e}^{-\mathrm{i}\Delta\beta z}\chi_{\mathrm{FH}}^{(2)}a_{2\omega}(z)a_\omega^*(z)\right)+\mathrm{i}a_{2\omega}^*\left(\mathrm{e}^{\mathrm{i}\Delta\beta z}\chi_{\mathrm{SH}}^{(2)}a_\omega^2(z)\right)+\mathrm{c.c.}\\
&= \mathrm{i}\mathrm{e}^{-\mathrm{i}\Delta\beta z}\chi_{\mathrm{FH}}^{(2)}a_{2\omega}(z)a_\omega^{*2}(z)-\mathrm{i}\mathrm{e}^{-\mathrm{i}\Delta\beta z}\chi_{\mathrm{SH}}^{(2)*}a_{2\omega}a_\omega^{*2}(z)+\mathrm{c.c.}\\
&= -2\,\mathrm{Im}\left[\mathrm{e}^{-\mathrm{i}\Delta\beta z}a_{2\omega}a_\omega^{*2}(\chi_{\mathrm{FH}}^{(2)}-\chi_{\mathrm{SH}}^{(2)*})\right].
\end{aligned}\tag{4.7}$$

Here we introduced the *phase mismatch*

$$\Delta\beta = 2\beta(\omega) - \beta(2\omega). \tag{4.8}$$

Note that (4.7) must hold for every z. This can only be fulfilled by

$$\chi^{(2)}_{\mathrm{FH}} = \chi^{(2)*}_{\mathrm{SH}} =: \chi^{(2)}_{\mathrm{eff}}. \tag{4.9}$$

Now we may perform a phase transformation of the SH amplitude

$$a(z) = a_\omega(z) \qquad b(z) = a_{2\omega}(z)\mathrm{e}^{-\mathrm{i}\Delta\beta z} \tag{4.10}$$

which gives us the final set of evolution equations

Evolution equations for SHG

$$\text{FH:}\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}a \qquad + \chi^{(2)}_{\mathrm{eff}} a^* b = 0 \tag{4.11}$$

$$\text{SH:}\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z}b - \Delta\beta b + \chi^{(2)*}_{\mathrm{eff}}\, a^2 = 0. \tag{4.12}$$

4.2 Normalization

We now aim towards a further simplification of the evolution equations for SHG which might provide us with simple estimations of relevant scales. For that we introduce dimensionless quantities:

$$z = Z_0 Z, \quad a = \sqrt{I_0}A \quad \text{and} \quad b = \sqrt{I_0}B\mathrm{e}^{\mathrm{i}\varphi}, \tag{4.13}$$

where Z_0 is the *characteristic length*, I_0 the *relevant intensity level* and φ a constant phase term. We insert these definitions into (4.11) and (4.12) and find

$$\text{FH:}\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}A \qquad + Z_0\chi^{(2)}_{\mathrm{eff}}\sqrt{I_0}\mathrm{e}^{\mathrm{i}\varphi}A^*B = 0 \tag{4.14}$$

$$\text{SH:}\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}B - \Delta\beta Z_0 B + Z_0\chi^{(2)}_{\mathrm{eff}}\sqrt{I_0}\mathrm{e}^{-\mathrm{i}\varphi}A^2 = 0. \tag{4.15}$$

Now we can actually choose the normalization constants Z_0 and I_0 in such a way, that all constants become unity

$$Z_0 = \frac{1}{|\Delta\beta|}, \quad \chi^{(2)}_{\mathrm{eff}}\mathrm{e}^{\mathrm{i}\varphi}\frac{\sqrt{I_0}}{|\Delta\beta|} = 1. \tag{4.16}$$

Z_0 now describes length scales for which the FH and SH waves run out of phase. For a perfectly phase-matched crystal this length scale goes to infinity. For the intensity we obtain

$$I_0 = \frac{1}{|Z_0\chi^{(2)}_{\mathrm{eff}}|} = \left|\frac{\Delta\beta}{\chi^{(2)}_{\mathrm{eff}}}\right|^2. \tag{4.17}$$

The characteristic induces a noticeable nonlinear action (it is reduced by a growing nonlinear coefficient, but growing with the squared mismatch). This explains why second harmonic generation is such a rare phenomenon. Particular care has to be taken to reduce the mismatch. Otherwise required power levels grow to infinity.

The phase φ is chosen as

$$\varphi = -\arg\left(\chi^{(2)}_{\text{eff}}\right). \tag{4.18}$$

It removes the phase of the nonlinear coefficient. This means the phase of the coefficient of the quadratic nonlinearity has no physical relevance.
Remark: Normalization is not a unique procedure. For example, in case of vanishing mismatch the sample length is much smaller than the inverse mismatch therefore becoming the relevant scale.

The final set of normalized equations can be summarized to

Normalized evolution equations for SHG

$$\text{FH:}\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}A + A^* B = 0 \tag{4.19}$$

$$\text{SH:}\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}B - \sigma B + A^2 = 0. \tag{4.20}$$

$\sigma = \operatorname{sgn}(\Delta\beta)$ is called the *scaled mismatch* and can have values $-1, 0, +1$. The original fields for a given $A(Z)$ and $B(Z)$ are

$$\begin{aligned} \boldsymbol{E}_{\text{real}}(z,t) = \sqrt{I_0}\,\mathrm{Re}\Bigg[& \sqrt{\frac{2\omega}{\beta(\omega)\varepsilon_0 c^2}}\frac{\boldsymbol{E}^{\omega}}{|\boldsymbol{E}^{\omega}_{\perp}|}A\left(Z=\frac{z}{Z_0}\right)\mathrm{e}^{\mathrm{i}\beta(\omega)z-\mathrm{i}\omega t} \\ & + \sqrt{\frac{4\omega}{\beta(2\omega)\varepsilon_0 c^2}}\frac{\boldsymbol{E}^{2\omega}}{|\boldsymbol{E}^{2\omega}_{\perp}|}B\left(Z=\frac{z}{Z_0}\right)\mathrm{e}^{2\mathrm{i}\beta(\omega)z-2\mathrm{i}\omega t+\mathrm{i}\varphi}\Bigg]. \end{aligned} \tag{4.21}$$

4.3 Conservation laws - the Hamiltonian

We have already enforced energy conservation onto our set of equations

$$Q = |A|^2 + |B|^2 = \text{const.} \tag{4.22}$$

A second conserved quantity is the Hamiltonian of the system given by

$$H = \sigma B^* B - A^2 B^* - A^{*2} B. \tag{4.23}$$

We bring meaning to this Hamiltonian by considering that B^* corresponds to the creation of a photon at the SH, whereas B describes the destruction of a photon. The same applies for A. Then the first term describes the destruction of generated second harmonic light by destructive interference in the mismatched case. The second term incorporates the SHG by the annihilation of two photons from the FH wave creating a photon of the SH wave. The last term describes down conversion of the SH wave into two photons of the FH wave.

We now prove that the Hamiltonian is indeed conserved:

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}Z}H &= \sigma B^* \frac{\mathrm{d}}{\mathrm{d}z}B - A^2\frac{\mathrm{d}}{\mathrm{d}z}B^* - 2AB^*\frac{\mathrm{d}}{\mathrm{d}z}A + \text{c.c.} \qquad (4.24)\\ &= \mathrm{i}\sigma B^*(-\sigma B + A^2) + \mathrm{i}A^2(-\sigma B + A^2)^* - 2\mathrm{i}AB^*A^*B + \text{c.c.}\\ &= -\mathrm{i}\sigma|B|^2 + \mathrm{i}\sigma|A|^4 - 2\mathrm{i}|A|^2|B|^2 + \cancel{\mathrm{i}\sigma B^* A^2} - \cancel{\mathrm{i}\sigma B^* A^2} + \text{c.c.}\\ &= 0. \qquad (4.25)\end{aligned}$$

Since the other terms are purely imaginary they will cancel with they complex conjugated terms. The quantum relation of the conserved quantities is the conserved norm of the wave function which is equivalent to total intensity and the expectation value of the Energy (Hamiltonian operator) which is equivalent to the Hamiltonian.

4.4 General solution

Now we want to find a general solution for the set of normalized evolution equations for SHG. We start with a transformation of complex variables into real ones by separating amplitudes and phases

$$A(Z) = |A(Z)|\mathrm{e}^{\mathrm{i}\varphi_A(Z)} \quad \text{and} \quad B(Z) = |B(Z)|\mathrm{e}^{\mathrm{i}\varphi_B(Z)} \tag{4.26}$$

giving us four independent variables. We can insert this ansatz (4.26) into equations (4.19) and (4.20) and divide by $\mathrm{e}^{\mathrm{i}\varphi_A(Z)}$ and $\mathrm{e}^{\mathrm{i}\varphi_B(Z)}$ which yields

$$\begin{aligned}&\text{FH} \quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}|A(Z)| - |A(Z)|\frac{\partial}{\partial Z}\varphi_A(Z) + |A(Z)|\cdot|B(Z)|\mathrm{e}^{\mathrm{i}\varphi_B(Z)-2\mathrm{i}\varphi_A(Z)} = 0\\ &\text{SH} \quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}|B(Z)| - |B(Z)|\frac{\partial}{\partial Z}\varphi_B(Z) - \sigma|B(Z)| + |A(Z)|^2\mathrm{e}^{2\mathrm{i}\varphi_A(Z)-\mathrm{i}\varphi_B(Z)} = 0.\end{aligned} \tag{4.27}$$

Now we only consider the imaginary part of equation (4.27)

$$\begin{aligned}&\text{FH} \quad \frac{\mathrm{d}}{\mathrm{d}Z}|A(Z)| + |A(Z)|\cdot|B(Z)|\sin\big(\Delta\varphi(Z)\big) = 0\\ &\text{SH} \quad \frac{\mathrm{d}}{\mathrm{d}Z}|B(Z)| - |A(Z)|^2\sin\big(\Delta\varphi(Z)\big) = 0,\end{aligned} \tag{4.28}$$

where $\Delta\varphi(Z) = \varphi_B(Z) - 2\varphi_A(Z)$ is the phase difference between SH and FH fields. We want to investigate the evolution equation of the phase difference as well by using the real part of equation (4.27)

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}Z}\Delta\varphi(Z) &= \frac{\partial}{\partial Z}\varphi_B(Z) - 2\frac{\partial}{\partial Z}\varphi_A(Z)\\ &= -\sigma + \left(\frac{|A(Z)|^2}{|B(Z)|} - 2|B(Z)|\right)\cos\big(\Delta\varphi(Z)\big). \qquad (4.29)\end{aligned}$$

In all coherent systems without a fixed time reference the absolute phase does not matter and only a phase difference plays a role. Thus we only have three independent real valued variables $\Delta\varphi(Z), |A(Z)|, |B(Z)|$.

We can reduce the number of free variables further by using the energy conservation (4.22)

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}Z}|B(Z)| &= \big(Q-|B(Z)|^2\big)\sin\big(\Delta\varphi(Z)\big)\\ \frac{\mathrm{d}}{\mathrm{d}Z}\Delta\varphi(Z) &= -\sigma+\left(\frac{Q-|B(Z)|^2}{|B(Z)|}-2|B(Z)|\right)\cos\big(\Delta\varphi(Z)\big).\end{aligned} \tag{4.30}$$

We can once more reduce the number of free variables by using the conserved Hamiltonian (4.23)

$$\begin{aligned}H &= \sigma|B(Z)|^2-2|A(Z)|^2|B(Z)|\cos\big(\Delta\varphi(Z)\big)\\ &= \sigma|B(Z)|^2-2(Q-|B(Z)|^2)|B(Z)|\cos\big(\Delta\varphi(Z)\big).\end{aligned} \tag{4.31}$$

We can use this result to extract and eliminate $\sin\big(\Delta\varphi(Z)\big)=\sqrt{1-\cos^2(\Delta\varphi(Z))}$ from equation (4.30)

$$\frac{\mathrm{d}}{\mathrm{d}Z}|B| = (Q-|B|^2)\sin\big(\Delta\varphi\big) = \pm(Q-|B|^2)\sqrt{1-\left(\frac{H-\sigma|B|^2}{2|B|(Q-|B|^2)}\right)^2}. \tag{4.32}$$

Now we transform to the SH power $\mathscr{I}=|B|^2$ by multiplying equation (4.32) with $2|B|$ because $\frac{\mathrm{d}\mathscr{I}}{\mathrm{d}Z}=2|B|\frac{\mathrm{d}|B|}{\mathrm{d}Z}$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}Z}\mathscr{I} &= \pm(Q-\mathscr{I})\sqrt{4\mathscr{I}-4\mathscr{I}\left(\frac{H-\sigma\mathscr{I}}{2\sqrt{\mathscr{I}}(Q-\mathscr{I})}\right)^2}\\ &= \pm\sqrt{4\mathscr{I}(Q-\mathscr{I})^2-(H-\sigma\mathscr{I})^2}.\end{aligned} \tag{4.33}$$

The formal solution can be obtained via integration. Using the initial conditions for given SH power at $Z=0:\mathscr{I}(0)=\mathscr{I}_0$

$$\int_{\mathscr{I}_0}^{\mathscr{I}(L)}\frac{\mathrm{d}\mathscr{I}}{\sqrt{4\mathscr{I}(Q-\mathscr{I})^2-(H-\sigma\mathscr{I})^2}} = \pm\int_0^L \mathrm{d}Z = \pm L. \tag{4.34}$$

A practicable solution of equation (4.34) still requires solving the integral and inverting the resulting function. This finally leads to so-called elliptic functions.

A nonlinear system with N real valued variables requires $N-1$ conserved quantities or symmetries for an analytical solution. The existence of conserved quantities is essential for an analytical solution.

4.5 Phase-matched SHG

Now we want to find solutions of (4.34) for the case of complete phase matching ($\sigma=0$) and no SH power at $Z=0$

$$|B(Z=0)|^2=\mathscr{I}_0=0. \tag{4.35}$$

Looking at the conserved quantities we find that the total energy is $Q = |A(Z=0)|^2$ and the Hamiltonian is simply zero $H = 0$ by comparing with (4.23). This simplifies (4.34) dramatically and leads to

$$\int_0^{\mathscr{I}(L)} \frac{\mathrm{d}\mathscr{I}}{2(Q-\mathscr{I})\sqrt{\mathscr{I}}} = \pm L,$$
$$\Rightarrow \frac{1}{2\sqrt{Q}} \ln\left(\frac{\sqrt{Q}+\sqrt{\mathscr{I}}}{\sqrt{Q}-\sqrt{\mathscr{I}}}\right)\Bigg|_0^{\mathscr{I}(L)} = \pm L. \tag{4.36}$$

Inverting the results gives us the following expression

SH power in phase matched case

$$\mathscr{I}(L) = Q \tanh^2\left(\sqrt{Q}L\right). \tag{4.37}$$

We conclude that total conversion is only reached for infinity propagation length. The conversion efficiency $\eta = \frac{\mathscr{I}}{Q}$ depends only on the product $\sqrt{Q}L$. The required power scales inverse to the squared propagation length (see normalization in chapter 4.2).

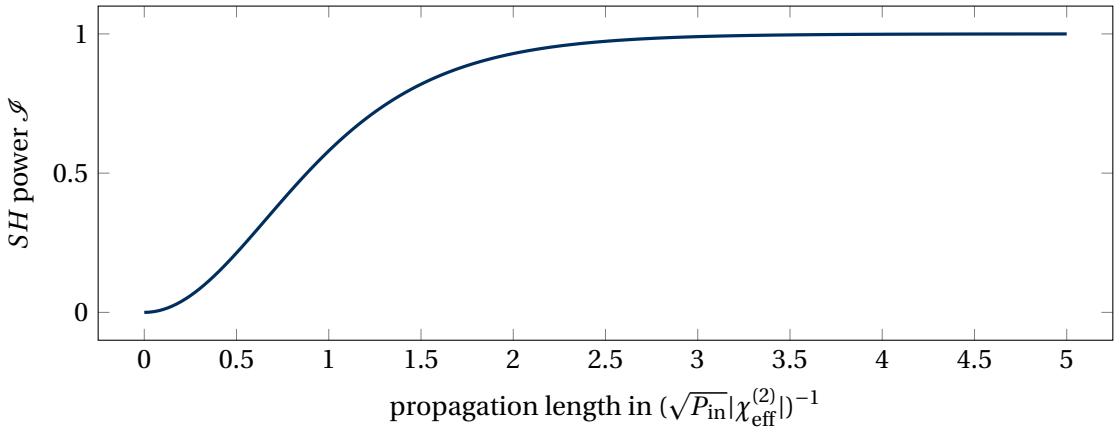


Fig. 2: Up-conversion in the phase-matched case.

4.6 Down conversion (phase matched case)

A general property of Hamiltonian systems is the reversibility of their evolution. If SH generation is possible, as discussed in the previous chapter, also the opposite process called parametric down conversion should be observed.

We assume the phase-matched case $\sigma = 0$ and all initial power to be in the SH wave

$$|B(Z=0)|^2 = \mathscr{I}_0 = Q \quad \text{and} \quad |A(Z=0)|^2 = 0. \tag{4.38}$$

The Hamiltonian (4.23)

$$H = \sigma|B(Z)|^2 - 2(Q-|B(Z)|^2)|B(Z)|\cos\big(\Delta\varphi(Z)\big) = 0. \tag{4.39}$$

Only the initial value of $\mathscr{I}$ ($\mathscr{I} = Q$ instead of $\mathscr{I} = 0$) has changed compared with SHG. The derivation is analogous to section 4.5 but we have changed the initial conditions

$$\frac{1}{2\sqrt{Q}} \ln\left(\frac{\sqrt{Q}+\sqrt{\mathscr{I}}}{\sqrt{Q}-\sqrt{\mathscr{I}}}\right)\Bigg|_{Q}^{\mathscr{I}(L)} = \pm L. \tag{4.40}$$

This leads to some problems. The function on the left side of equation (4.40) becomes infinity on the lower boundary. The intensity of the SH wave can only deviate from Q, if the propagation length L tends to infinity. This is consistent with an inversion of the SH process discussed above. The latter one also needs an infinite propagation length to obtain complete conversion.

A direct solution of equation (4.36) yields

$$\frac{\mathrm{d}}{\mathrm{d}Z}\mathscr{I} = \pm 2(Q-\mathscr{I})\sqrt{\mathscr{I}} \Rightarrow \mathscr{I}(L) = Q. \tag{4.41}$$

The system can stay in a state with all the power in the SH field and formally requires an infinite propagation length to obtain noticeable down-conversion.

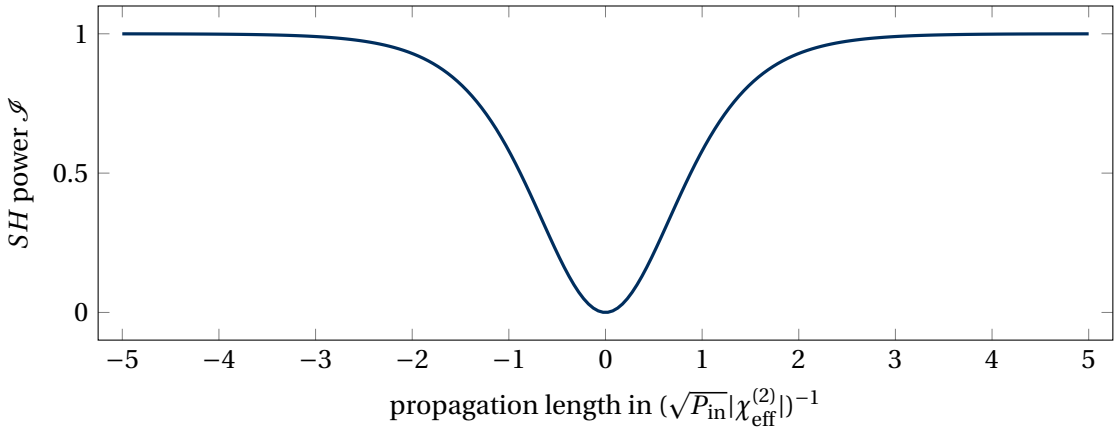


Fig. 3: Down-conversion for the phase-matched case. The down conversion process starts from quantum fluctuations.

In real systems any small perturbation (e. g. quantum noise) starts the down-conversion process after a finite length. In the phase-matched case complete down-conversion is obtained (inverse behaviour to SHG) and SHG starts (see previous chapter).

4.7 Stability of the second harmonic field

We have already noticed that down-conversion is started by field noise. To investigate that phenomenon in more detail we perform a stability analysis. We take the results from section 4.2

$$\begin{aligned} \text{FH:}\quad & \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}A \qquad + A^* B = 0 \\ \text{SH:}\quad & \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}B - \sigma B + A^2 = 0. \end{aligned} \tag{4.42}$$

We want to look for stationary solutions $B_0 = \sqrt{Q}\mathrm{e}^{-\mathrm{i}\sigma Z}$ and $A_0 = 0$ with small fluctuations $\delta A(Z)$ and $\delta B(Z)$

$$A(Z) = \delta A(Z)\mathrm{e}^{-\mathrm{i}\frac{\sigma}{2}Z} \quad \text{and} \quad B(Z) = \left(\sqrt{Q} + \delta B(Z)\right)\mathrm{e}^{-\mathrm{i}\sigma Z}. \tag{4.43}$$

Then the evolution equations for the small perturbations become

$$\begin{aligned} &\text{FH:} \quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}\delta A + \frac{\sigma}{2}\delta A + \delta A^*\left(\sqrt{Q} + \cancel{\delta B}\right) = 0 \\ &\text{SH:} \qquad\quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z}\delta B \qquad\quad + \cancel{\delta A^2} \qquad\quad = 0. \end{aligned} \tag{4.44}$$

We assume δA and δB to be small, so that quadratic terms can be neglected. Then the SH perturbation δB remains in first order approximation a constant (as long as $\delta A(Z)$ remains small). Now we apply $\left(-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} + \frac{\sigma}{2}\right)$ to the first part of (4.44)

$$\begin{aligned} 0 &= \left(-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} + \frac{\sigma}{2}\right)\left[\left(\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}Z} + \frac{\sigma}{2}\right)\delta A + \delta A^*\sqrt{Q}\right] \\ &= \left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} + \left(\frac{\sigma}{2}\right)^2\right)\delta A + \sqrt{Q}\left[\left(\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} + \frac{\sigma}{2}\right)\delta A\right]^* \\ &\overset{(4.44)}{=} \left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} + \left(\frac{\sigma}{2}\right)^2 - Q\right)\delta A. \end{aligned} \tag{4.45}$$

the general solution of this equation can be expressed in terms of exponential functions

$$\delta A(Z) = \delta A_+ \exp\left(\sqrt{Q - \left(\frac{\sigma}{2}\right)^2}Z\right) + \delta A_- \exp\left(-\sqrt{Q - \left(\frac{\sigma}{2}\right)^2}Z\right). \tag{4.46}$$

The two coefficients $\delta A_\pm$ are fixed and determined by the initial fluctuations $\delta A(Z=0)$ and their first derivative

$$\frac{\mathrm{d}}{\mathrm{d}Z}\delta A(Z)\bigg|_{Z=0} = \mathrm{i}\frac{\sigma}{2}\delta A(Z=0) + \mathrm{i}\sqrt{Q}\delta A^*(Z=0). \tag{4.47}$$

Now we can use this and the initial conditions

$$\delta A(Z=0) = \delta A_+ + \delta A_- \quad \text{and} \quad \frac{\mathrm{d}}{\mathrm{d}Z}\delta A(Z=0) = \sqrt{Q - \left(\frac{\sigma}{2}\right)^2}\left[\delta A_+ - \delta A_-\right] \tag{4.48}$$

to find an expression for $\delta A_\pm$

$$\delta A_\pm = \left(\frac{1}{2} \pm \mathrm{i}\frac{\sigma/2}{\sqrt{4Q - \sigma^2}}\right)\delta A(Z=0) \pm \mathrm{i}\frac{\sqrt{Q}}{\sqrt{4Q-\sigma^2}}\delta A^*(Z=0). \tag{4.49}$$

Now we can distinguish two cases:

a.) SH power is below a threshold $Q \leq \left(\frac{\sigma}{2}\right)^2$

This occurs for a mismatched sample ($\sigma = \pm 1$) only: $Q \leq \frac{1}{4}$. The arguments in the exponential terms become imaginary

$$\delta A(Z) = \delta A_+ \exp\left(\mathrm{i}\sqrt{\frac{1}{4} - Q}Z\right) + \delta A_- \exp\left(-\mathrm{i}\sqrt{\frac{1}{4} - Q}Z\right). \tag{4.50}$$

This leads to an oscillatory behaviour. The initial perturbation is conserved (which is typical for Hamiltonic systems). We do not observe growth of the SH wave or further down-conversion. The system is (marginally) stable.

b.) SH power is above a threshold $Q > \left(\frac{\sigma}{2}\right)^2$

Then the fluctuations will grow exponentially

$$\delta A(Z) = \delta A_+ \exp\left(\sqrt{Q - \left(\frac{\sigma}{2}\right)^2}Z\right) + \delta A_- \underbrace{\exp\left(-\sqrt{Q - \left(\frac{\sigma}{2}\right)^2}Z\right)}_{\approx 0 \text{ for large } Z}. \tag{4.51}$$

Inserting the equation for the coefficients $\delta A_\pm$ we find for large Z

$$\left[\left(\frac{1}{2} + \mathrm{i}\frac{\sigma/2}{\sqrt{4Q - \sigma^2}}\right)\delta A(Z=0) + \mathrm{i}\frac{\sqrt{Q}}{\sqrt{4Q-\sigma^2}}\delta A^*(Z=0)\right]\exp\left(\sqrt{Q - \left(\frac{\sigma}{2}\right)^2}Z\right). \tag{4.52}$$

We observe that this solution is unstable. Also the initial perturbation δA *and* its complex conjugate δA^* are amplified (so-called parametric amplifier).

We also want to discuss the case of large SH power with $4Q \gg \sigma^2 = 1$. Then we can approximate $\delta A(Z)$ as

$$\delta A(Z) \approx \frac{1}{2}\left(\delta A(Z=0) + \mathrm{i}\delta A^*(Z=0)\right)\mathrm{e}^{\sqrt{Q}Z}. \tag{4.53}$$

The phase of the amplified perturbation for large Z is

$$\begin{aligned}\varphi &= \arctan\left(\frac{\mathrm{Im}[\delta A(Z=0) + \mathrm{i}\delta A^*(Z=0)]}{\mathrm{Re}[\delta A(Z=0) + \mathrm{i}\delta A^*(Z=0)]}\right) \\ &= \arctan\left(\frac{\mathrm{Im}[\delta A(Z=0)] + \mathrm{Re}[\delta A(Z=0)]}{\mathrm{Re}[\delta A(Z=0)] + \mathrm{Im}[\delta A(Z=0)]}\right) = \arctan(1) = \frac{\pi}{4}.\end{aligned} \tag{4.54}$$

This means that after a certain propagation length the phase of the perturbation is fixed and independent of $\delta A(Z=0)$. This means the phase fluctuations (even of quantum origin) are suppressed which leads to squeezed (non-classical) light.

Lastly we want to mention the critical intensity for down conversion in real world units $Q < \frac{1}{4}$

$$I_{\text{thresh.}} = \left(\frac{\sigma}{2}\right)^2 I_0 = \frac{1}{4}\left|\frac{\Delta\beta}{\chi^{(2)}_{\text{eff}}}\right|^2. \tag{4.55}$$

4.8 Visualization of the field dynamics using the Hamiltonian

We aim for a demonstration of the field dynamics in an intuitive way. We start again with the conserved quantities, namely the Hamiltonian H given in (4.31) and the total intensity Q given by (4.22). Then we can find another conserved quantity

$$H'(\sigma,|B|,\Delta\varphi) = \frac{H}{Q^{3/2}} = \underbrace{\frac{\sigma}{\sqrt{Q}}}_{=\sigma'}\underbrace{\frac{|B(z)|^2}{Q}}_{=:|B'|} - 2\left(1-\frac{|B(Z)|^2}{Q}\right)\frac{|B(z)|}{\sqrt{Q}}\cos\big(\Delta\varphi(Z)\big)$$

$$H'(\sigma',|B'|,\Delta\varphi) = \sigma'|B'|^2 - 2(1-|B'|^2)|B'|\cos\big(\Delta\varphi\big), \tag{4.56}$$

where $|B'|$ is the scaled amplitude of the SH field with $0 \le |B|' \le 1$, $\Delta\varphi$ the phase difference between SH and FH fields and σ' the rescaled mismatch.

The system now evolves along the lines/contours of fixed rescaled Hamiltonian H'. The conserved direction of motion on each contour line is determined by

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}Z}\Delta\varphi(Z) &= -\sigma + \left(\frac{Q}{|B(Z)|} - 3|B(Z)|\right)\cos\big(\Delta\varphi(Z)\big)\\ \frac{1}{\sqrt{Q}}\frac{\mathrm{d}}{\mathrm{d}Z}\Delta\varphi(Z) &= -\sigma' + \left(\frac{1}{|B'|} - 3|B'|\right)\cos\big(\Delta\varphi(Z)\big).\end{aligned} \tag{4.57}$$

We can draw the contour lines for several values of the Hamiltonian H' and different mismatches in figure 4.

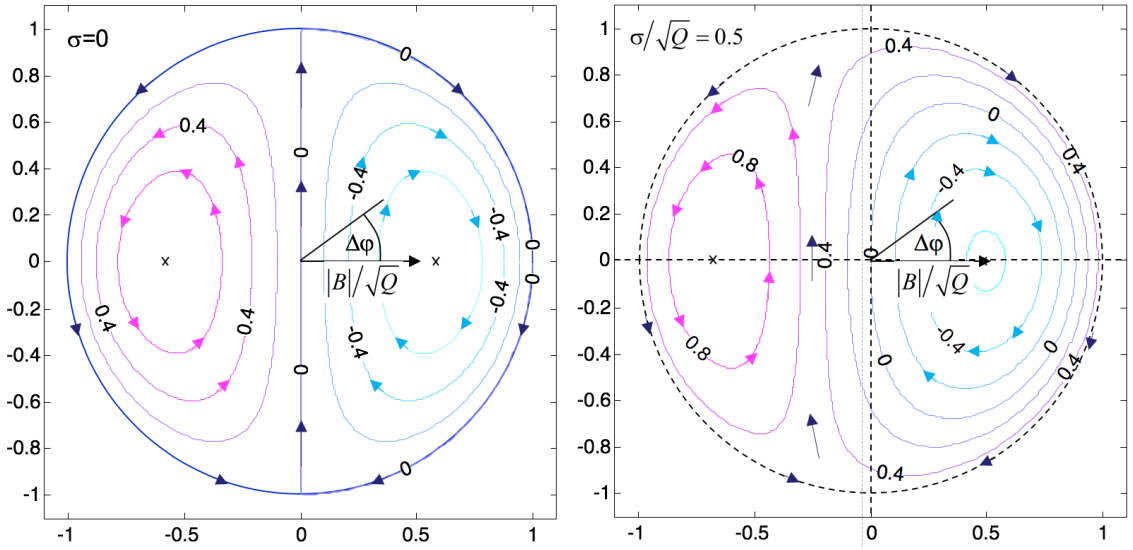


Fig. 4: Contour plots of the scaled Hamiltonian H' for zero mismatch (left) and $\sigma' = 0.5$ (right). The radial coordinate $|B'|$ describes the scaled SH power. For our initial conditions we start at contour lines going through the origin.

For SHG we find a contour line of $H' = 0$ which starts at the origin (at zero SH power). For a zero mismatch 4 (left) we see that $|B'|$ approaches 1 (complete conversion) after infinite time. However, if we introduce a mismatch 4 (right) then the inversion is incomplete, because the

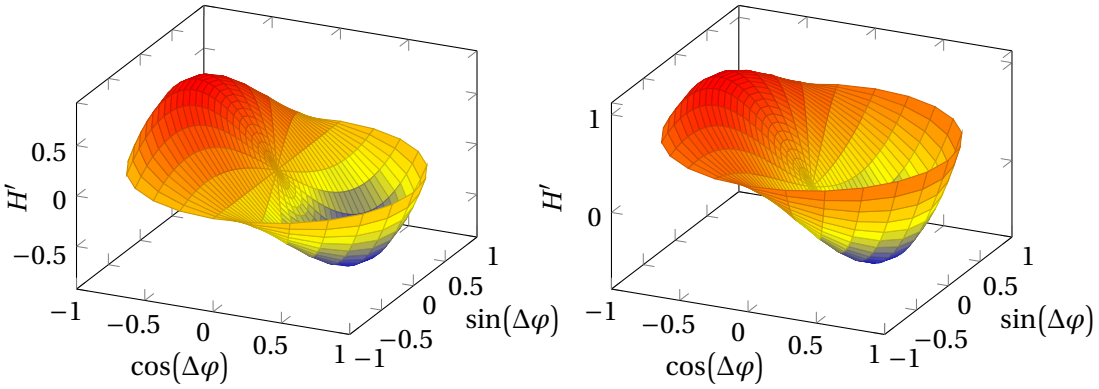


Fig. 5: 3D visualization of the scaled Hamiltonian for zero mismatch (left) and $\sigma' = 0.5$ (right).

system evolves on a contour line which never reaches the border of $|B'| = 1$. We observe a periodic motion corresponding to up- and down-conversion.

If we start outside the origin (e. g. simultaneously inject FH and SH fields), the phase between SH and FH fields matters. We see a periodic motion around the minimum or maximum of the scaled Hamiltonian. If we start at one of the extrema of H' we will have a stationary state consisting of SH and FH fields.

The scaled mismatch $\sigma' = \frac{\sigma}{\sqrt{Q}}$ controls the dynamics of the system. Increasing the power corresponds to a reduction of mismatch.

4.9 Low-depletion-limit

We now want to consider a system with a *large mismatch* with the unscaled equations (4.12). We will find a low conversion efficiency, which means that the FH intensity is nearly constant (undepleted pump approximation). Then

$$a(z) \approx \sqrt{I_{\mathrm{FH}}} = \text{const.} \tag{4.58}$$

Then we may remove $a(z)$ from the equation of the SH field (4.12) yielding

$$\text{SH:} \quad \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} b - \Delta\beta b + \chi_{\mathrm{eff}}^{(2)*} I_{\mathrm{FH}} = 0. \tag{4.59}$$

Using the initial condition of no SH intensity $b(z=0) = 0$ we find the solution

$$b(z) = -\frac{\chi_{\mathrm{eff}}^{(2)*}}{\Delta\beta} I_{\mathrm{FH}} \left(\exp\left(-\mathrm{i}\Delta\beta z\right) - 1\right). \tag{4.60}$$

The intensity of the SH wave is then

$$\begin{aligned} I_{\mathrm{SH}} = |b(z)|^2 &= \frac{|\chi_{\mathrm{eff}}^{(2)*}|^2}{\Delta\beta^2} I_{\mathrm{FH}}^2 \left|\mathrm{e}^{\mathrm{i}\frac{\Delta\beta z}{2}}\right|^2 \left|\mathrm{e}^{-\mathrm{i}\frac{\Delta\beta z}{2}} - \mathrm{e}^{\mathrm{i}\frac{\Delta\beta z}{2}}\right|^2 \\ &= 4|\chi_{\mathrm{eff}}^{(2)*}|^2 I_{\mathrm{FH}}^2 \left(\frac{\sin\left(\frac{\Delta\beta}{2} z\right)}{\Delta\beta}\right)^2. \end{aligned} \tag{4.61}$$

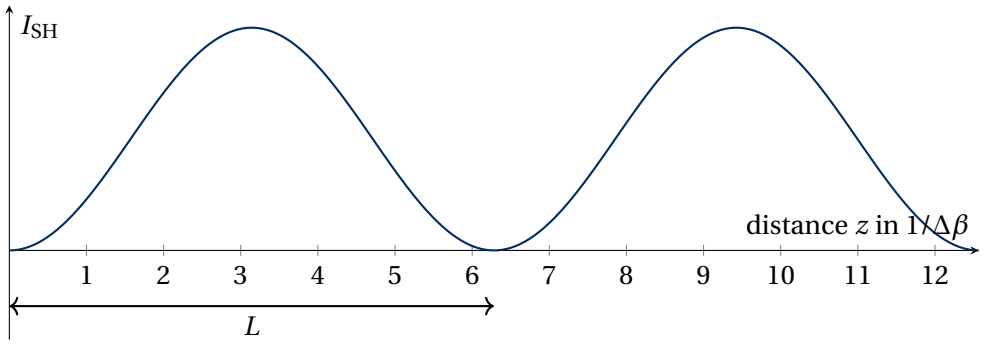


Fig. 6: SH power in the undepleted pump approximation with $a(z) = \text{const}$. L describes the oscillation period of SH-generation.

We observe (figure 6) that the intensity decays quickly with growing mismatch and oscillates periodically with a period of

$$L = \frac{2\pi}{|\Delta\beta|} = \left|\frac{2\pi}{\beta(2\omega) - 2\beta(\omega)}\right| = \left|\frac{1}{\frac{n_{SH}}{\lambda_{SH}} - 2\frac{n_{FH}}{\lambda_{FH}}}\right| = \left|\frac{\lambda_{FH}\lambda_{SH}}{\lambda_{FH}n_{SH} - 2\lambda_{SH}n_{FH}}\right|$$
$$= \left|\frac{\lambda_{SH}}{n_{FH} - n_{SH}}\right|, \tag{4.62}$$

where n_{FH}, n_{SH} are the refractive indices at FH/SH frequency and $\lambda_{FH}, \lambda_{SH}$ the vacuum wavelength of the FH/SH wave.

5 Diffraction, dispersion and nonlinearities

Now we want to discuss effects of further (transverse) coordinates which leads us to partial differential equations instead of ODEs. Usually we will find no analytical solutions. However, we need the transverse coordinates in order to account for diffraction.

5.1 Beam evolution in isotropic Kerr materials

We make several assumptions concerning the field propagation:

- The propagation takes place mainly in z-direction.
- We use a linearly polarized monochromatic electric field.
- The Kerr medium is isotropic: $\beta = n(\omega)\frac{\omega}{c}$.

We use the evolution equation for nonlinearity driven diffraction that was derived in chapter 3.3 (equation (3.25))

$$\left[\mathrm{i}\frac{\partial}{\partial z} + \frac{1}{2\beta}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\right] a(x,y,z) = -\sqrt{\frac{\omega^3}{8\beta\varepsilon_0 c^2}}\frac{\boldsymbol{E}^{\beta *}}{|\boldsymbol{E}^{\beta}|}\boldsymbol{P}^{\mathrm{NL}}(z)\exp\left(-\mathrm{i}\beta z\right). \tag{5.1}$$

The monochromatic field implies that no Third-Harmonic Generation takes place and we only have to deal with self-phase modulation. Since the electric field is linearly polarized, the nonlinearity induces a refractive index change $\Delta n = n_2 I$ (I-intensity). Then the nonlinear induced polarization takes the form

$$\boldsymbol{P}^{\mathrm{NL}}(z) \sim n_2 |a(x,y,z)|^2 a(x,y,z)\exp\left(\mathrm{i}\beta z\right). \tag{5.2}$$

The nonlinearly induced change of the propagation constant is then

$$\Delta\beta = \Delta n\frac{\omega}{c} = n_2|a(x,y,z)|^2\frac{\omega}{c}. \tag{5.3}$$

Now we can include the effect of the nonlinear polarization as a change of the propagation constant $\Delta\beta$

$$\left[\mathrm{i}\frac{\partial}{\partial z} + \frac{1}{2\beta}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) + \frac{\omega}{c}n_2|a(x,y,z)|^2\right] a(x,y,z) = 0. \tag{5.4}$$

Now we again perform the normalization process with $z = Z_0 Z, x = W_0 X, y = W_0 Y$ and $a(x,y,z) = \sqrt{I_0}A(X,Y,Z)$

$$\left[\mathrm{i}\frac{\partial}{\partial Z} + \frac{1}{2}\frac{Z_0}{\beta W_0^2}\left(\frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2}\right) + \frac{\omega}{c}n_2 Z_0 I_0 |A(X,Y,Z)|^2\right] A(X,Y,Z) = 0. \tag{5.5}$$

We can now choose the normalization constants in such a way that the prefactors become one

$$\begin{aligned} \frac{Z_0}{\beta W_0^2} = 1 \qquad &\Rightarrow \quad Z_0 = \beta W_0^2 = \frac{2\pi n W_0^2}{\lambda_0} \\ \frac{\omega}{c} n_2 Z_0 I_0 = 1 \quad &\Rightarrow \quad I_0 = \frac{\lambda_0}{2\pi} \frac{1}{|n_2| Z_0} = \frac{\lambda_0}{2\pi} \frac{\lambda_0}{2\pi n |n_2| W_0^2} = \frac{1}{n|n_2|} \left(\frac{\lambda_0}{2\pi W_0} \right)^2 . \end{aligned} \tag{5.6}$$

We see that W_0 is a free scaling parameter, so we choose it e. g. as the initial beam width. This means that the principal effects we discuss will not depend on the beam width.

Since we chose W_0 to be the beam width its advantageous to introduce the total beam power

$$P = \int_{-\infty}^{\infty} \mathrm{d}x \int_{-\infty}^{\infty} \mathrm{d}y \, |a(x,y,z)|^2 = W_0^2 I_0 \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \, |A(X,Y,Z)|^2, \tag{5.7}$$

with $P_0 = W_0^2 I_0 = \dfrac{1}{n|n_2|} \left(\dfrac{\lambda_0}{2\pi} \right)^2$ as a new scaling quantity.

Note that P_0 only depends on material properties and wavelengths, not the beam width. Qualitative changes are now to be expected to depend on the beam power in relation to P_0, but not on the beam width, as it can be scaled away.

The final equation we have to solve is a nonlinear 2+1 dimensional SCHRÖDINGER equation (NLS)

$$\left[\mathrm{i} \frac{\partial}{\partial z} + \frac{1}{2} \left(\frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2} \right) + \kappa |A|^2 \right] A = 0 \quad \text{with} \quad \kappa = \mathrm{sgn}(n_2) = \pm 1. \tag{5.8}$$

However, this equation has no general solution (not integrable). The nonlinearity creates an effective potential $\kappa |A|^2 \sim -V$. For $\kappa = +1 (n_2 > 0)$ high intensities correspond to potential dips.

Conserved quantities

For our further discussions we want to investigate the conserved quantities. We start with

the total power P

$$
\begin{aligned}
P &= P_0 \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \, |A(X,Y,Z)|^2 \\
\frac{\mathrm{d}P}{\mathrm{d}z} &= P_0 \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left[A^* \frac{\partial}{\partial Z} A + \text{c.c.} \right] \\
&= P_0 \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(A^* \mathrm{i} \left[\frac{1}{2} \left(\frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2} \right) + |A|^2 \right] A + \text{c.c.} \right) \\
&\quad \int_{-\infty}^{\infty} A^* \frac{\partial^2}{\partial X^2} A = \underbrace{\left(A^* \frac{\partial}{\partial X} A \right)}_{\to 0} \Bigg|_{-\infty}^{\infty} - \int_{-\infty}^{\infty} \mathrm{d}X \frac{\partial}{\partial X} A^* \frac{\partial}{\partial X} A \\
&= P_0 \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(-\frac{\mathrm{i}}{2} \frac{\partial}{\partial X} A^* \frac{\partial}{\partial X} A - \frac{\mathrm{i}}{2} \frac{\partial}{\partial Y} A^* \frac{\partial}{\partial Y} A + \mathrm{i}|A|^4 + \text{c.c.} \right) \\
&= P_0 \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(-\frac{\mathrm{i}}{2} \left| \frac{\partial}{\partial X} A \right|^2 - \frac{\mathrm{i}}{2} \left| \frac{\partial}{\partial Y} A \right|^2 + \mathrm{i}|A|^4 + \text{c.c.} \right) = 0.
\end{aligned} \tag{5.9}
$$

The last part cancels with its complex conjugate.

The second conserved quantity is the Hamiltonian which is given by

$$
H = \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \Big(\underbrace{\left| \frac{\partial A}{\partial X} \right|^2 + \left| \frac{\partial A}{\partial Y} \right|^2}_{\text{kinetic energy}} - \underbrace{\kappa |A|^4}_{\substack{\text{self-attraction} \\ \text{self-repulsion}}} \Big). \tag{5.10}
$$

The second term corresponds to a potential energy since $|A|^4$ can be interpreted as a multiplication of particle density $\sim |A|^2$ times Potential $\sim |A|^2$. We can proof that the Hamiltonian is conserved in a similar way as for the total power.

Second moment

A third quantity we want to introduce is called the *second moment* defined by

$$
S(Z) = \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \, (X^2 + Y^2) |A(X,Y,Z)|^2. \tag{5.11}
$$

Despite the other quantities the second moment is not conserved. It is a measure of the beam width $\langle W(Z) \rangle = \sqrt{\frac{S(Z)}{Q}}$.

We can use the definition of $S(Z)$ to derive its evolution equation. However, we only want to mention the result:

$$
\boxed{\frac{\mathrm{d}^2}{\mathrm{d}Z^2} S = 4H} \quad \text{conserved.} \tag{5.12}
$$

In the following we want to distinguish two cases:

a.) $$H = \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(\left| \frac{\partial A}{\partial X} \right|^2 + \left| \frac{\partial A}{\partial Y} \right|^2 - \kappa |A|^4 \right) > 0.$$

We fulfill this case always for $\kappa = -1 (n_2 < 0)$. Here n_2 corresponds to a defocussing nonlinearity. The case $\kappa = +1$ also works for prevailing kinetic energy (e. g. curved phase fronts). Then we find

$$\frac{\mathrm{d}^2}{\mathrm{d}Z^2} S = 4H > 0. \tag{5.13}$$

The solutions of this equations are parabolas which diverge for $Z \to \infty$

$$\langle W(Z) \rangle = \sqrt{\frac{S(Z)}{Q}} \to \infty. \tag{5.14}$$

We can also draw the solutions as done in figure 7.

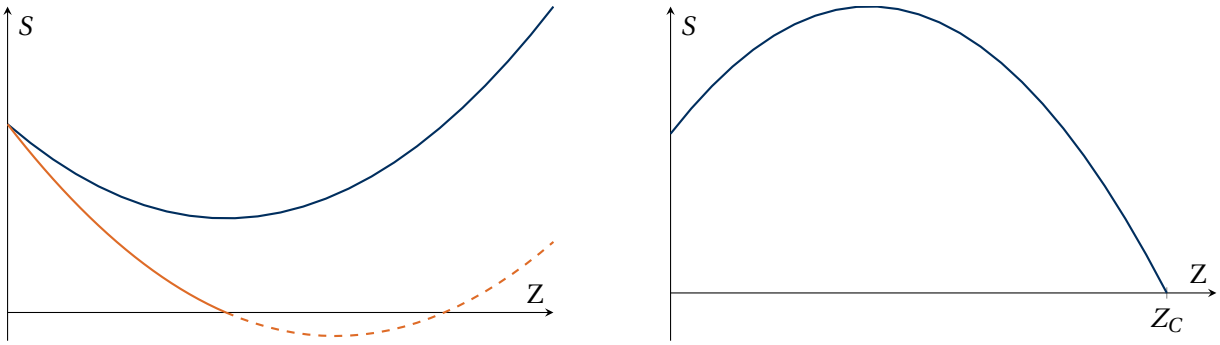


Fig. 7: Solutions of the second moment $S(Z)$ for $H > 0$ (left) and $H < 0$ (right). Since $S(Z) > 0$ per definition, the evolution path is not possible after $S(Z) = 0$.

b.) $$H = \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(\left| \frac{\partial A}{\partial X} \right|^2 + \left| \frac{\partial A}{\partial Y} \right|^2 - \kappa |A|^4 \right) < 0.$$

This case only works for $\kappa = +1 (n_2 > 0)$. Using the evolution equation of the second moment we find that the beam will collapse within a finite propagation distance

$$S(Z) = 0 \Rightarrow W(Z) = 0 \Rightarrow |A|^2 \to \infty. \tag{5.15}$$

The collapse is unphysical because of zero width (paraxial approximation fails) and infinite intensities (power expansion of $\boldsymbol{P}^{\mathrm{NL}}$ cannot be stopped after the third order). However, the evolution is often very well described up to the damage threshold of the material.

Furthermore we want to calculate the critical power for self-focusing for $\kappa = 1$. We assume that the initial field has a fixed beam profile $f(X, Y)$ but a varying amplitude: $A(X, Y, Z =$

$0) = A_0 f(X,Y)$. Then the Hamiltonian can be written as

$$H = \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(\left| \frac{\partial A}{\partial X} \right|^2 + \left| \frac{\partial A}{\partial Y} \right|^2 - |A|^4 \right) = |A_0|^2 h_{\mathrm{kin}} - |A_0|^4 h_{\mathrm{int}} \tag{5.16}$$

$$\text{with} \quad h_{\mathrm{kin}} = \frac{1}{2} \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \left(\left| \frac{\partial f}{\partial X} \right|^2 + \left| \frac{\partial f}{\partial Y} \right|^2 \right) \quad \text{and} \quad h_{\mathrm{int}} = \frac{1}{2} \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \, |f|^4.$$

Both h_{kin} and h_{int} are only shape dependent. We can easily find the critical power as

$$H = |A_0|^2 h_{\mathrm{kin}} - |A_0|^4 h_{\mathrm{int}} < 0 \quad \text{for} \quad |A_0|^2 > |A_{\mathrm{crit}}| = \frac{h_{\mathrm{kin}}}{h_{\mathrm{int}}}. \tag{5.17}$$

Every beam collapses above a certain (profile dependent) power level. The beam profile with the minimum power for collapse is called the *Townes soliton*. It is a metastable (quasi-stationary) state between spreading and self-focusing. It is cylindrically symmetric, has no analytical solution and thus must be numerically determined. It looks similar to a Gaussian profile (except of a simple exponential decay in the tails). Numerically we find Q_{Townes} as

$$Q_{\mathrm{Townes}} = \int_{-\infty}^{\infty} \mathrm{d}X \int_{-\infty}^{\infty} \mathrm{d}Y \, |A_{\mathrm{Townes}}(X,Y,Z)|^2 = 5.85043. \tag{5.18}$$

In real world units, the critical power for collapse would be

$$P_{\mathrm{critical}} = Q_{\mathrm{Townes}} P_0 = 5.85043 \frac{1}{n|n_2|} \left(\frac{\lambda_0}{2\pi} \right)^2. \tag{5.19}$$

For fused silica with $n = 1.45$, $n_2 = 2{,}4 \cdot 10^{-20} \, \frac{\mathrm{m}^2}{\mathrm{W}}$ and a wavelength of $\lambda_0 = 800\,\mathrm{nm}$ we find

$$P_{\mathrm{critical}} = 2{,}8\,\mathrm{MW}. \tag{5.20}$$

For a Gaussian profile the critical power is increased by 2 %.

5.2 Pulse evolution in a two-level medium

Lastly we want to consider the pulse evolution in a two-level-system. First we have to make some assumptions:

- The propagation is entirely in z-direction (no diffraction).
- The medium is isotropic: $\beta_0 = n(\omega_0) \frac{\omega_0}{c}$.
- The two-level-system is not damped (pulse duration $\ll$ damping time).
- The electric field is linearly polarized.
- Group velocity dispersion (GVD) of the background medium can be neglected.

The polarization of the two-level medium is

$$\boldsymbol{P}^{\mathrm{TLS}}_{\mathrm{real}}(z,t) = \frac{1}{2}\left[\boldsymbol{P}^{\mathrm{TLS}}(z,t)\mathrm{e}^{\mathrm{i}(\beta_0 z-\omega_0 t)} + \mathrm{c.c.}\right], \tag{5.21}$$

where $\boldsymbol{P}^{\mathrm{TLS}}(z,t)$ is the slowly varying envelope of the polarization of the two-level-system. The total electric field is

$$\boldsymbol{E}_{\mathrm{real}}(z,t) = \frac{1}{2}\left[\boldsymbol{E}(z,t)\mathrm{e}^{\mathrm{i}(\beta_0 z-\omega_0 t)} + \mathrm{c.c.}\right] \tag{5.22}$$

$$\text{with } \boldsymbol{E}(z,t) = \boldsymbol{E}^{\beta_0}\tilde{u}(z,t) = \boldsymbol{E}^{\beta_0}\frac{a(z,t)}{\sqrt{\left\langle s_z^{\beta_0}\right\rangle}} = \boldsymbol{E}^{\beta_0}\frac{a(z,t)}{\sqrt{\frac{\varepsilon_0 c^2}{2}\frac{\beta_0}{\omega_0}|\boldsymbol{E}^{\beta_0}|^2}} = \frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\sqrt{\frac{2}{\varepsilon_0 c n(\omega_0)}}a(z,t).$$

The evolution equation for the slowly varying envelope of a pulse is (see (3.36))

$$\begin{aligned}
\left[\mathrm{i}\frac{\partial}{\partial z} + \frac{\mathrm{i}}{v_g}\frac{\partial}{\partial t}\right]a(t,z) &= \frac{1}{\sqrt{8\beta_0 c^2\varepsilon_0\omega_0}}\left[\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\frac{\partial^2}{\partial t^2}\tilde{\boldsymbol{P}}^{\mathrm{NL}}(z,t)\right]\mathrm{e}^{-\mathrm{i}(\beta_0 z-\omega_0 t)} \\
&= \frac{1}{\sqrt{8\beta_0 c^2\varepsilon_0\omega_0}}\left[\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\frac{\partial^2}{\partial t^2}\left(\boldsymbol{P}^{\mathrm{TLS}}(z,t)\mathrm{e}^{\mathrm{i}(\beta_0 z-\omega_0 t)}\right)\right]\mathrm{e}^{-\mathrm{i}(\beta_0 z-\omega_0 t)} \\
&= \frac{1}{\sqrt{8\beta_0 c^2\varepsilon_0\omega_0}}\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\left(-\omega_0^2 - 2\mathrm{i}\omega_0\cancel{\frac{\partial}{\partial t}} + \cancel{\frac{\partial^2}{\partial t^2}}\right)\boldsymbol{P}^{\mathrm{TLS}}(z,t) \\
&\approx -\sqrt{\frac{\omega_0^2}{8nc\varepsilon_0}}\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|^2}\boldsymbol{P}^{\mathrm{TLS}}(z,t).
\end{aligned} \tag{5.23}$$

Now we use the evolution equations of the polarization for zero detuning $\hbar\omega_0 = E_a - E_b$ (equations (2.27) and (2.28))

$$\begin{aligned}
\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{P}^{\mathrm{TLS}} &= \frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}I \\
\frac{\mathrm{d}I}{\mathrm{d}t} &= \frac{1}{2\mathrm{i}\hbar N}\boldsymbol{E}^*\boldsymbol{P}^{\mathrm{TLS}} + \mathrm{c.c.}
\end{aligned} \tag{5.24}$$

Again we introduce a normalization $z = Z_0 Z$, $t = T_0 T$, $a(Z,T) = \sqrt{\hat{I}_0}A(Z,T)$ and

$$\boldsymbol{P}^{\mathrm{TLS}}(z,t) = -\mathrm{i}\frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\hat{P}_0 P(Z,T). \tag{5.25}$$

Then equation (5.23) becomes

$$\left[\mathrm{i}\frac{\partial}{\partial z} + \frac{\mathrm{i}}{v_g}\frac{Z_0}{T_0}\frac{\partial}{\partial T}\right]A(Z,T) = -\frac{Z_0}{\sqrt{\hat{I}_0}}\sqrt{\frac{\omega_0^2}{8nc\varepsilon_0}}\frac{\boldsymbol{E}^{\beta_0 *}}{|\boldsymbol{E}^{\beta_0}|}\left[-\mathrm{i}\frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\hat{P}_0 P(Z,T)\right]. \tag{5.26}$$

Inserting all the expressions for $\boldsymbol{P}^{\mathrm{TLS}}$ (5.25) and $\boldsymbol{E}$ (5.22) into the evolution equations we find

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}T}\left[-\mathrm{i}\frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\hat{P}_0 P(Z,T)\right] = T_0\sqrt{\hat{I}_0}\frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\left[\frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\sqrt{\frac{2}{\varepsilon_0 c n(\omega_0)}}A(Z,T)\right]I \tag{5.27}$$

$$\frac{\mathrm{d}I}{\mathrm{d}T} = \frac{T_0\sqrt{\hat{I}_0}}{2\mathrm{i}\hbar N}\left[\frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\sqrt{\frac{2}{\varepsilon_0 c n(\omega_0)}}A(Z,T)\right]^*\left[-\mathrm{i}\frac{\boldsymbol{E}^{\beta_0}}{|\boldsymbol{E}^{\beta_0}|}\hat{P}_0 P(Z,T)\right] + \mathrm{c.c.} \tag{5.28}$$

We choose our scale parameters in such a way, that the constants vanish. This leads us to the following conditions:

$$\begin{aligned}
&\overset{(5.26)}{\Rightarrow} \frac{Z_0}{v_g T_0} = 1, \qquad \frac{Z_0}{\sqrt{\hat{I}_0}} \sqrt{\frac{\omega_0^2}{8nc\varepsilon_0}} \hat{P}_0 = 1, \\
&\overset{(5.27)}{\Rightarrow} \frac{T_0\sqrt{\hat{I}_0}}{\hat{P}_0} \frac{Ne^2}{\hbar} |\langle \psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2 \sqrt{\frac{2}{\varepsilon_0 c n(\omega_0)}} = 1, \\
&\overset{(5.28)}{\Rightarrow} \frac{T_0\sqrt{\hat{I}_0}}{\hbar N} \hat{P}_0 \sqrt{\frac{2}{\varepsilon_0 c n(\omega_0)}} = 1.
\end{aligned} \tag{5.29}$$

Some algebraic rearrangements result in the following solutions for the scale parameters with $\mu := |\langle \psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|$:

$$\hat{P}_0 = N e \mu, \quad T_0 = \frac{1}{e\mu}\sqrt{\frac{2nc\hbar\varepsilon_0}{v_g \omega_0 N}}, \quad \hat{I}_0 = \frac{1}{4}\hbar\omega_0 v_g N. \tag{5.30}$$

The final set of normalized equation now looks like

Normalized equations in a two-level-system

$$\frac{\partial}{\partial T} P(Z,T) = A I \tag{5.31a}$$

$$\frac{\partial}{\partial T} I(Z,T) = -\frac{1}{2}\left(A^* P + A P^*\right) \tag{5.31b}$$

$$P(Z,T) = \left(\frac{\partial}{\partial Z} + \frac{\partial}{\partial T}\right) A(Z,T). \tag{5.31c}$$

We use the initial conditions $\lim_{T\to-\infty} P(Z,T) = 0$, $\lim_{T\to-\infty} A(Z,T) = 0$ and $\lim_{T\to-\infty} Z(Z,T) = -1$ where the system is in the ground state.

We try to find solution using conserved quantities. One of these is

$$|P|^2 + I^2 = 1 \tag{5.32}$$

$$\frac{\partial}{\partial T}\left(|P|^2 + I^2\right) = P^* \frac{\partial P}{\partial T} + P \frac{\partial P^*}{\partial T} + 2I \frac{\partial I}{\partial T} = P^* A I + P A^* I + 2I\left[-\frac{1}{2}(A^* P + A P^*)\right] = 0.$$

In the following we restrict to real valued solutions for A, P, I which simplifies (5.31b) to

$$\frac{\partial}{\partial T} I(Z,T) = -A(Z,T) P(Z,T). \tag{5.33}$$

Since (5.32) looks similar to the trigonometric PYTHAGOREAN, we introduce a substitution

$$P(Z,T) = \sin\Theta(Z,T), \quad I(Z,T) = -\cos\Theta(Z,T), \quad A(Z,T) = -\frac{\partial\Theta(Z,T)}{\partial T}. \tag{5.34}$$

This inherently fulfils the conservation law $|P|^2+I^2=1$, but also equations (5.31a) and (5.31b). An evaluation of equation (5.31c) leads us to

$$\left(\frac{\partial}{\partial Z}+\frac{\partial}{\partial T}\right)\frac{\partial}{\partial T}\Theta(Z,T)=-\sin\Theta(Z,T). \tag{5.35}$$

For further simplification we transform into a moving spatial frame $\tilde{Z}=2Z-T$ which changes the derivatives accordingly

$$\frac{\partial}{\partial Z}\Rightarrow 2\frac{\partial}{\partial\tilde{Z}},\quad \frac{\partial}{\partial T}\Rightarrow\frac{\partial}{\partial T}-\frac{\partial}{\partial\tilde{Z}},\quad \left(\frac{\partial}{\partial Z}+\frac{\partial}{\partial T}\right)\frac{\partial}{\partial T}\Rightarrow\left(\frac{\partial^2}{\partial T^2}+\frac{\partial^2}{\partial\tilde{Z}^2}\right). \tag{5.36}$$

Using this, equation (5.35) transforms into the *Sine-Gordon equation*

$$\left(\frac{\partial^2}{\partial\tilde{Z}^2}+\frac{\partial^2}{\partial T^2}\right)\Theta(\tilde{Z},T)=\sin\Theta(\tilde{Z},T) \tag{5.37}$$

which is actually solvable. We want to briefly state its physical origins:

- Pendulum under the action of gravity:
  $\frac{\mathrm{d}^2}{\mathrm{d}T^2}\Theta(T)+\sin\Theta(T)=0$
- Set of coupled pendula under the action of gravity:
  $$\frac{\mathrm{d}^2}{\mathrm{d}T^2}\Theta(Z_i,T)+\sin\Theta(Z_i,T)=\underbrace{[\Theta(Z_{i+1},T)-\Theta(Z_i,T)]+[\Theta(Z_{i-1},T)-\Theta(Z_i,T)]}_{\sim\frac{\partial^2}{\partial\tilde{Z}^2}\Theta(T,\tilde{Z})}.$$

The sine-Gordon equation is integrable, however, it requires an infinite number of conserved quantities. It bears conserved multi-soliton solutions, e. g. first order solitons like

$$\Theta(\tilde{Z},T)=4\arctan\left[\exp\left(\pm\frac{\tilde{Z}-vT}{\sqrt{1-v^2}}+\delta\right)\right], \tag{5.38}$$

where $0\le v<1$ is the velocity of the soliton. We observe that the pendula make a full turn in angular space changing their phase from 0 to 2π (*kink*, the case of + sign) or from 2π to 0 (*anti-kink*, the case of – sign). For our description this means that the inversion goes from –1 to +1 and returns to –1. The given solution is displayed in figure 8.

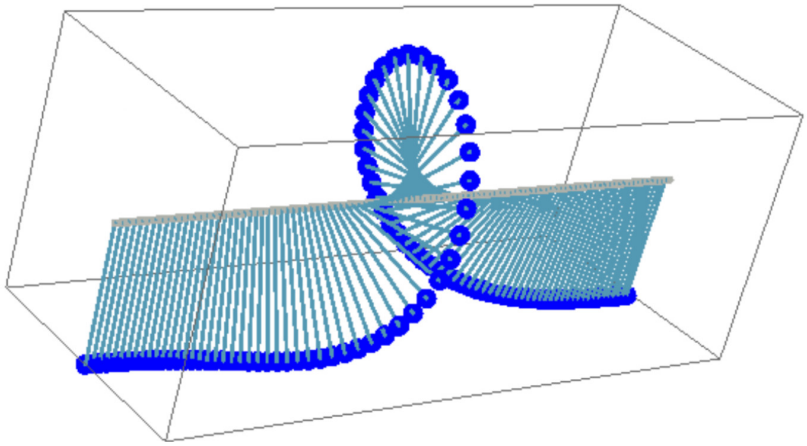


Fig. 8: Basic single soliton solution, simulated in *Mathematica* as systems of spring-coupled torsional pendula, see V.G. Ivancevic and T.T. Ivancevig, JGSP 31(2013) 1–56