

Photonics II - Lectures

Lecturer: Rainer Kaltenbaek

Faculty of Mathematics and Physics
University of Ljubljana
IQOQI Vienna
Austrian Academy of Sciences

Ljubljana, Summer Semester 2020

1 Modulating light

In Photonics I, you heard about how light is generated, how it is focused, how it interferes, how its polarization can be modified, and how light is detected. In the following sections, we will discuss how we can modulate light by using electric or magnetic fields or by using sound waves or liquid crystals. In order to discuss these concepts, we will first quickly introduce a few concepts from nonlinear optics.

1.1 A brief introduction to Nonlinear optics

1.1.1 Susceptibility

Later we will discuss the modulation of optical signals by using electric or magnetic fields. Since (classical) light can be described as an electromagnetic wave, an electromagnetic modulation of light is effectively a nonlinear effect. By nonlinear optics we refer to effects where electromagnetic waves modify the optical properties of a material. A well-known example of a nonlinear optical effect is second-harmonic generation (SHG), where a beam of light with photons of frequency ω is incident on a nonlinear crystal, and part of that incident light is frequency “upconverted” to 2ω . For example, the incident light could have 780 nm and the outgoing wavelength 390 nm. We will discuss this effect in more detail later on.

To describe nonlinear effects more accurately, it is convenient to consider the polarization $P(t)$ of a material system in the presence of an electric field strength $E(t)$. For simplicity, we are only talking about scalar fields at the moment, not about vector fields.

If there are no nonlinear effects, the polarization will be in a linear relation to the electric field:

$$P(t) = \epsilon_0 \chi^{(1)} E(t), \quad (1.1)$$

where ϵ_0 is the electric permittivity of vacuum, and the proportionality constant $\chi^{(1)}$ is called the linear susceptibility. If there are nonlinear effects, one can generalize equation 1.1 as follows:

$$\begin{aligned} P(t) &= \epsilon_0 [\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots] \\ &= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + \dots \end{aligned} \quad (1.2)$$

$\chi^{(k)}$ is called the k -th order susceptibility. The most common effects we will discuss in nonlinear optics are based on the second-order nonlinear polarization. In general, when

the field has a vectorial character, the $\chi^{(k)}$ will be tensors.

Equations 1.1 and 1.1 also assumed that the polarization at time t depends instantaneously on the field at time t . This can only be true if there is no loss and no dispersion in the material[1]. In general, there will be loss and dispersion, of course.

The polarization of the material can again be the source of an electromagnetic field (light). The effects of sum- and difference-frequency generation I mentioned above are examples for that. We denote these effects as SFG and DFG, respectively. In the special case where two photons of the incoming beam lead to the creation of one outgoing photon, one calls the effect not SFG but second-harmonic generation (SHG) or frequency doubling, which we mentioned above.

As an example of a nonlinear process, let us consider SFG, where we have two incoming fields of frequencies ω_1 and ω_2 adding up:

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.} \quad (1.3)$$

Then the second-order contribution to the polarization in our nonlinear medium will be:

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} E(t)^2 = \epsilon_0 \chi^{(2)} [E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.}] + 2\epsilon_0 \chi^{(2)} [E_1 E_1^* + E_2 E_2^*]. \quad (1.4)$$

If we compare this with the decomposition of the polarization into its frequency components

$$P^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}, \quad (1.5)$$

we can associate different effects to the various summands:

$$\begin{aligned} P(2\omega_1) &= \epsilon_0 \chi^{(2)} E_1^2 (\text{SHG}) \\ P(2\omega_2) &= \epsilon_0 \chi^{(2)} E_2^2 (\text{SHG}) \\ P(\omega_1 + \omega_2) &= \epsilon_0 \chi^{(2)} E_1 E_2 (\text{SFG}) \\ P(\omega_1 - \omega_2) &= \epsilon_0 \chi^{(2)} E_1 E_2^* (\text{DFG}) \\ P(0) &= 2\epsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*) (\text{OR}). \end{aligned} \quad (1.6)$$

We will discuss the effect of optical rectification (OR) in a later lecture. Essentially, the mixture of positive and negative frequency parts of the same fields, one gets an effective DC polarization. Usually, this has no noticeable effect, but in the case of ultra-fast pulses passing through a $\chi^{(2)}$ -nonlinear medium, the rapid building up of a DC field, followed by its rapid decay will lead to the emission of electromagnetic waves. We will come across this effect again in the context of generating THz waves.

We should note two things: (1) the notation used for $E(t)$ and $P(t)$ often differ by a factor of 2 between different authors. (2) because $P(t)$ needs to be a real-valued function, the corresponding function in frequency space $P(\omega)$ needs to fulfill the relation

$$P(-\omega) = P^*(\omega).$$

1.1.2 Order of magnitude estimates

To get an impression of the strength or weakness of nonlinear optical effects, let us quickly consider some rough numerical estimates.

The linear susceptibility $\chi^{(1)}$ is defined such that it will be of order 1 in condensed matter[1]. To estimate $P^{(2)}$, let us assume that it should be of order 1 if the field amplitude E is on the order of the typical atomic electric field strength $E_{\text{at}} = e/(4\pi\epsilon_0 a_0^2)$. e is the electron charge, and $a = 4\pi\epsilon_0 \hbar^2/(me^2)$ is the Bohr radius. In particular, one gets $E_{\text{at}} = 6.14 \times 10^{11}$ V/m. Then one can expect that $\chi^{(2)}$ should be of order 1 if the applied field is on the order of $1/E_{\text{at}}$. Then one can estimate[1]:

$$\chi^{(2)} \approx 1.94 \times 10^{-12} \text{ m/V}. \quad (1.7)$$

Similarly, one can estimate that $\chi^{(3)}$ should be on the order of $1/E_{\text{at}}^2$ or:

$$\chi^{(3)} \approx 3.78 \times 10^{-24} \text{ m}^2/\text{V}^2. \quad (1.8)$$

1.1.3 The formal definition of the nonlinear susceptibility

The description above was strongly simplified because we only discussed the case of a scalar, real field with no losses and no dispersion. In general, the electric field will be a real function that we can write as a sum of complex frequency components:

$$\vec{E}(\vec{r}, t) = \sum_n \vec{E}_n(\vec{r}, t), \quad (1.9)$$

where this sum is only over positive frequencies parts.

Assuming that the field intensity varies on a time scale much slower than optical frequencies, we can write the field as a discrete sum of frequency components:

$$\vec{E}(\vec{r}, t) = \sum_n \vec{E}(\omega_n) e^{-i\omega_n t} = \sum_n \vec{A}(\omega_n) e^{i(\vec{k}_n \cdot \vec{r} - \omega_n t)}. \quad (1.10)$$

Here, the sum goes over positive and negative frequencies, and the last step assumes that \vec{A} is an amplitude that is varying spatially only very slowly[1].

One can choose a similar decomposition into different frequency components for the polarization vector. Then one can write:

$$\vec{P}(\vec{r}, t) = \sum_n \vec{P}(\omega_n) e^{-i\omega_n t}. \quad (1.11)$$

This latter notation is useful to see the frequency dependencies between the incident and

the outgoing light. In particular, consider the situation where two beams are incident on a $\chi^{(2)}$ non-linear medium and that all other orders of susceptibility can be assumed zero for now. Then the components of the polarization of the medium will take the following form[1]:

$$P_i(\omega_n + \omega_m) = \epsilon_0 \sum_{ij} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m). \quad (1.12)$$

Here, the sum over n and m runs over all indices **but** under the restriction that the sum $\omega_n + \omega_m$ be equal to the frequency of the outgoing field. To emphasize the distinction between the incoming and the outgoing field, the susceptibility $\chi^{(2)}(\omega_3, \omega_2, \omega_1)$ is sometimes written as $\chi^{(2)}(\omega_3; \omega_2, \omega_1)$, or one writes $\chi^{(2)}(\omega_3 = \omega_2 + \omega_1)$ for the case of SFG.

1.1.4 The nonlinear susceptibility of a classical anharmonic oscillator

The Lorentz model of an atom provides a very good description of the linear optical response of some materials like e.g. atomic vapours or non-metallic solids. In that model, the electrons in an atom move in a harmonic oscillator potential. Here, we will discuss what we can learn about non-linear optical effects by extending the Lorentz model by adding nonlinear terms to the potential of the electrons. An observation of particular interest will be influence of symmetry considerations on the nonlinear optical response of a medium. In particular, our analysis will depend on whether the medium is centrosymmetric or noncentrosymmetric. In both cases, we will consider the lowest-order nonlinear deviations from the harmonic potential.

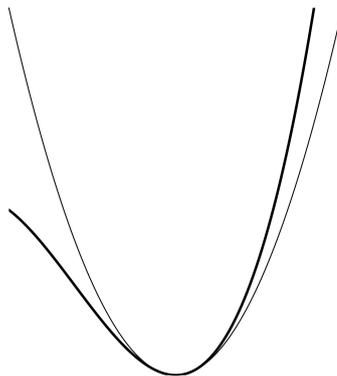


Figure 1: **A noncentrosymmetric potential.** The thin line shows a parabola, the thicker line shows a potential that also includes a third-order contribution. The latter is noncentrosymmetric.

Noncentrosymmetric media

We will see that a noncentrosymmetric potential will allow to describe nonlinear effects like SFG and DFG. Fig. 1 shows an example for a noncentrosymmetric potential. The motion of an electron in the lowest-order nonlinear noncentrosymmetric potential is given by:

$$\frac{d^2x(t)}{dt^2} + 2\gamma\frac{dx(t)}{dt} + \omega_0^2x(t) + ax^2(t) = -e\frac{E(t)}{m}, \quad (1.13)$$

where e is the elementary charge, and m is the mass of the electron.

For DFG/SFG, we want to overlap two fields in the crystal, which will add up to:

$$E(t) = E_1e^{-i\omega_1t} + E_2e^{-i\omega_2t} + \text{c.c.} \quad (1.14)$$

If one puts that into equation 1.13, there is no general solution known. However, one can solve the equation using a perturbation expansion if the applied field is sufficiently weak. In that case, we can write equation 1.13 as:

$$\frac{d^2x(t)}{dt^2} + 2\gamma\frac{dx(t)}{dt} + \omega_0^2x(t) + ax^2(t) = -\lambda e\frac{E(t)}{m}, \quad (1.15)$$

where λ is a parameter characterizing the strength of the perturbation. That means it should be a small positive number. We then seek a solution that we can express as a power series in λ . That means:

$$x(t) = \sum_{k=1}^{\infty} \lambda^k x^{(k)}(t). \quad (1.16)$$

One then inserts this ansatz of a solution on the left-hand side of equation 1.15. The equation must be fulfilled separately for each order of λ^k . From this, one arrives at a system of equations[1]:

$$\frac{d^2x^{(1)}(t)}{dt^2} + 2\gamma\frac{dx^{(1)}(t)}{dt} + \omega_0^2x^{(1)}(t) = -e\frac{E(t)}{m} \quad (1.17a)$$

$$\frac{d^2x^{(2)}(t)}{dt^2} + 2\gamma\frac{dx^{(2)}(t)}{dt} + \omega_0^2x^{(2)}(t) + a[x^{(1)}(t)]^2 = 0 \quad (1.17b)$$

$$\frac{d^2x^{(3)}(t)}{dt^2} + 2\gamma\frac{dx^{(3)}(t)}{dt} + \omega_0^2x^{(3)}(t) + 2ax^{(1)}(t)x^{(2)}(t) = 0, \text{ etc.} \quad (1.17c)$$

The lowest-order equation 1.17a corresponds to a harmonic oscillator equation as in the Lorentz model. Which makes sense - it describes the limiting case of the absence of non-linear effects. The steady-state solution (not including a decay of amplitudes due

to attenuation) is:

$$x^{(1)}(t) = x^{(1)}(\omega_1)e^{-i\omega_1 t} + x^{(1)}(\omega_2)e^{-i\omega_2 t} + \text{c.c.}, \quad (1.18)$$

where the amplitudes can be written as[1]:

$$x^{(1)}(\omega_j) = -\frac{e}{m} \frac{E_j}{D(\omega_j)} \quad (1.19)$$

if we introduce the “complex denominator function”[1]:

$$D(\omega_j) = \omega_0^2 - \omega_j^2 - 2i\omega_j\gamma. \quad (1.20)$$

One can get the lowest-order correction term $x^{(2)}$ by taking the solution for $x^{(1)}(t)$, putting it into equation 1.17b and then solving that equation. Since one needs to square $x^{(1)}(t)$ in that equation, one will get frequency contributions corresponding to $\pm 2\omega_1$, $\pm 2\omega_2$, $\pm(\omega_1 + \omega_2)$, $\pm(\omega_1 - \omega_2)$, and 0.

Following this approach, one gets the following solutions[1]

$$x^{(2)}(2\omega_1) = -\frac{a(e/m)^2 E_1^2}{D(2\omega_1)D^2(\omega_1)} \quad (1.21a)$$

$$x^{(2)}(2\omega_2) = -\frac{a(e/m)^2 E_2^2}{D(2\omega_2)D^2(\omega_2)} \quad (1.21b)$$

$$x^{(2)}(\omega_1 + \omega_2) = -\frac{2a(e/m)^2 E_1 E_2}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)} \quad (1.21c)$$

$$x^{(2)}(\omega_1 - \omega_2) = -\frac{2a(e/m)^2 E_1 E_2^*}{D(\omega_1 - \omega_2)D(\omega_1)D(-\omega_2)} \quad (1.21d)$$

$$x^{(2)}(0) = -\frac{2a(e/m)^2 E_1 E_1^*}{D(0)D(\omega_1)D(-\omega_1)} - \frac{2a(e/m)^2 E_2 E_2^*}{D(0)D(\omega_2)D(-\omega_2)}. \quad (1.21e)$$

During today’s exercises, we will express these solutions in terms of the first and second order susceptibilities by using the following relation:

$$P^{(k)}(\omega) = -Nex^{(k)}(\omega). \quad (1.22)$$

The above considerations show that the lowest-order non-linear effects in a noncentrosymmetric medium are of second order. We will also discuss higher-order nonlinear effects at a later point. Those will depend on higher-order susceptibilities of the medium.

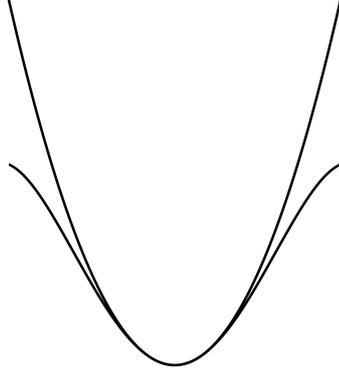


Figure 2: **A centrosymmetric potential.** The thin line shows a parabola, the thicker line shows a potential that also includes a fourth-order contribution. Both are centrosymmetric.

Centrosymmetric media

Similar to the case of the noncentrosymmetric media we just discussed, let us now do similar considerations for centrosymmetric media. In the case of a noncentrosymmetric medium, we added a second-order term to the force acting on the electron in equation 1.13. Fig. 2 shows an example for centrosymmetric potentials. To describe a nonlinear centrosymmetric medium, we will instead add a third-order term to the restoring force acting on our electron. The corresponding equation of motion will then be:

$$\frac{d^2x(t)}{dt^2} + 2\gamma\frac{dx(t)}{dt} + \omega_0^2x(t) - bx^3(t) = -e\frac{E(t)}{m}. \quad (1.23)$$

b quantifies the strength of the nonlinearity.

In three dimensions, that becomes[1]:

$$\frac{d^2\vec{r}(t)}{dt^2} + 2\gamma\frac{d\vec{r}(t)}{dt} + \omega^2\vec{r}(t) - b[\vec{r}(t) \cdot \vec{r}(t)]\vec{r}(t) = -\frac{e}{m}\vec{E}(t). \quad (1.24)$$

Now let us assume that we combine multiple electric fields inside our nonlinear medium. For simplicity, we will assume that all fields have the same polarization but that they can have different frequencies. We can write this field as:

$$\vec{E}(t) = \sum_n \vec{\omega}_n e^{-i\omega_n t}. \quad (1.25)$$

Again, there is no exact solution to equation 1.22, but one can make a similar perturbation approach as we did before. For that purpose, one replaces $\vec{E}(t)$ in equation 1.23 with $\lambda\vec{E}(t)$, where we again used λ as the perturbation parameter.

One then attempts to find a solution one can write as a power series in λ :

$$\vec{r}(t) = \sum_{k=1}^{\infty} \lambda^k \vec{r}^{(k)}. \quad (1.26)$$

If we insert this into equation 1.24, we can write individual equations for each individual λ^k and then simultaneously solve the differential equations for each power k . For $k = 1, 2, 3$, we get the following three equations, respectively:

$$\frac{d^2 \vec{r}^{(1)}(t)}{dt^2} + 2\gamma \frac{d\vec{r}^{(1)}(t)}{dt} + \omega_0^2 \vec{r}^{(1)}(t) = -\frac{e\vec{E}(t)}{m}, \quad (1.27a)$$

$$\frac{d^2 \vec{r}^{(2)}(t)}{dt^2} + 2\gamma \frac{d\vec{r}^{(2)}(t)}{dt} + \omega_0^2 \vec{r}^{(2)}(t) = 0, \quad (1.27b)$$

$$\frac{d^2 \vec{r}^{(3)}(t)}{dt^2} + 2\gamma \frac{d\vec{r}^{(3)}(t)}{dt} + \omega_0^2 \vec{r}^{(3)}(t) - b [\vec{r}^{(1)}(t) \cdot \vec{r}^{(1)}(t)] \vec{r}^{(1)}(t) = 0. \quad (1.27c)$$

The solution of the linear equation is again a superposition of the incoming waves:

$$\vec{r}^{(1)}(t) = \sum_{k=1}^{\infty} \vec{r}^{(1)}(\omega_k) e^{-i\omega_k t}, \quad (1.28)$$

where the frequency contributions can be written as:

$$\vec{r}^{(1)}(\omega_k) = -\frac{e\vec{E}(\omega_k)}{mD(\omega_k)}. \quad (1.29)$$

$D(\omega_k)$ is defined similar as before:

$$D(\omega_k) = \omega_0^2 - \omega_k^2 - 2i\omega_k\gamma. \quad (1.30)$$

The linear solution for the polarization then fulfills an equation that is essentially equivalent to the noncentrosymmetric case:

$$\vec{P}^{(1)}(\omega_k) = -Ne\vec{r}^{(1)}(\omega_k), \quad (1.31)$$

which we can again express in terms of the linear susceptibility:

$$P_i^{(1)}(\omega_k) = \epsilon_0 \sum_j \chi_{ij}^{(1)}(\omega_k) E_j(\omega_k), \quad (1.32)$$

where the linear susceptibility is diagonal: $\chi_{ij}^{(1)}(\omega_k) = \chi^{(1)}(\omega_k) \delta_{ij}$.

The more interesting case are the higher-order solutions for $\vec{r}(t)$. In particular, we can see from equation 1.27b that we have a local oscillator equation where there is damping

but no driving force. For that reason, the steady-state solution will be $\vec{r}^{(2)}(t) = 0$. That means that the lowest-order nonlinear effects we will see in a centrosymmetric medium are of at least third order. In that case, equation 1.27c shows that there is damping but there also is a driving force proportional to b .

1.1.5 Properties of the nonlinear susceptibility

The nonlinear susceptibility has symmetry properties we will discuss in this subsection. These symmetries help reducing the number of independent coefficients of the susceptibility. That this is prudent becomes apparent if one considers in detail how many independent coefficients the susceptibilities could in principle have. Let us consider the 2nd order susceptibility $\chi^{(2)}$ which corresponds to the interaction between three waves of frequencies ω_1 , ω_2 , and $\omega_3 = \omega_1 + \omega_2$. The susceptibility relates the Cartesian components of the polarization, which fulfill the following equation:

$$P_i(\omega_n + \omega_m) = \epsilon_0 \sum_{jk} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m). \quad (1.33)$$

The notation (nm) for the summation indices of the second sum again mean that we require $\omega_3 = \omega_1 + \omega_2$. In order to characterize all the possible interactions, we therefore need to determine the six tensors

$$\begin{aligned} \chi_{ijk}^{(2)}(\omega_1, \omega_3, -\omega_2), & \quad \chi_{ijk}^{(2)}(\omega_1, -\omega_2, \omega_3), & \quad \chi_{ijk}^{(2)}(\omega_2, \omega_3, -\omega_1), \\ \chi_{ijk}^{(2)}(\omega_2, -\omega_1, \omega_3), & \quad \chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2), \text{ and} & \quad \chi_{ijk}^{(2)}(\omega_3, \omega_2, \omega_1), \end{aligned}$$

and six more tensors where all the frequencies have the opposite sign. Each of these 12 tensors has 27 components, which means that we would in principle have to know 324 complex numbers. The situation becomes even more daunting for higher-order effects, but already for the 2nd order susceptibility it makes sense to take advantage of any symmetries we can find.

Reality of the fields

The polarization is a measurable quantity and must therefore be a real quantity. For that reason, the negative frequency components must be related to the positive ones in the following way:

$$P_i(-\omega_n - \omega_m) = P_i(\omega_n + \omega_m)^*. \quad (1.34)$$

Similarly, we know that we must have:

$$E_j(-\omega_n) = E_j(\omega_n)^*. \quad (1.35)$$

Given these relations and equation 1.33, we can conclude that the 2nd-order susceptibility fulfills the following symmetry:

$$\chi_{ijk}^{(2)}(-\omega_n - \omega_m, -\omega_n, -\omega_m) = \chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)^*. \quad (1.36)$$

Intrinsic permutation symmetry

If we have products of multiple fields like $E_j(\omega_m)E_k(\omega_n)$, it is pretty arbitrary, which of the fields we denote with $E_j(\omega_m)$ and which field we denote with $E_k(\omega_n)$. This is very similar to dummy indices in Einstein's summation convention, where it does not matter, which letter we use to make the summation.

As an example, let us consider the contribution of the product

$$\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)E_j(\omega_n)E_k(\omega_m)$$

to the nonlinear polarization $P_i(\omega_n + \omega_m)$ in equation 1.33. Because j, k, n , and m are dummy indices in the summation, we could have chosen a different order of these indices such that the factor would have read:

$$\chi_{ikj}^{(2)}(\omega_n + \omega_m, \omega_m, \omega_n)E_j(\omega_m)E_k(\omega_n).$$

These two terms will be equal if we require that the nonlinear susceptibility is symmetric under interchange of its last two frequency arguments and its last two Cartesian indices[1]

$$\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m) = \chi_{ikj}^{(2)}(\omega_n + \omega_m, \omega_m, \omega_n). \quad (1.37)$$

This *intrinsic permutation symmetry* simply means that it does not matter which field we denote as the first and which as the second in products like $E_j(\omega_n)E_k(\omega_m)$. Requiring intrinsic permutation symmetry as in equation 1.37 is just one way of dealing with this symmetry. Alternatively, we could have set one of the two components of $\chi^{(2)}$ equal to zero and doubled the value of the corresponding component with exchanged indices. The overall sum would still be the same[1].

Symmetries for lossless media

For lossless media, two additional symmetries occur. The first of these symmetries states that all of the components of $\chi_{ijk}^{(2)}(\omega_n + \omega_m, \omega_n, \omega_m)$ are real. That this is true for a lossless medium can be shown by doing the appropriate limit in the quantum-mechanical description or by considering the field energy density inside the medium[1]. We will not go to that level of detail here. In reality, of course no medium is lossless, but it will be a good approximation if the frequencies involved as well as their sums and differences are far from resonance.

The second symmetry for lossless media is *full* permutation symmetry. This means

that all the frequency arguments can be interchanged arbitrarily if one interchanges the indices of the susceptibility in the same order. Because the first frequency argument always needs to be the sum of the other two frequency arguments, one has to adapt the sign accordingly if one interchanges the frequency arguments. For example:

$$\chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{jki}^{(2)}(-\omega_1 = \omega_2 - \omega_3). \quad (1.38)$$

To simplify our notation, we did not write $\chi^{(2)}$ with three arguments but with only one.

Kleinman's symmetry

In many cases, the optical signals sent into the nonlinear medium have optical frequencies ω_i far from any relevant resonance frequency ω_0 of the material. If that is the case, the susceptibility will not depend on the frequency. That means there is no dispersion. Because we assume here that the optical frequencies are far from material resonances, the medium will be lossless, and full permutation symmetry will apply as in equation 1.38. In addition to that, if one assumes that the susceptibility does not depend on the frequency, we can interchange the indices of the susceptibility *without* interchanging the frequencies. The result is known as *Kleinman's symmetry*:

$$\begin{aligned} \chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2) &= \chi_{jki}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{kij}^{(2)}(\omega_3 = \omega_1 + \omega_2) \\ &= \chi_{ikj}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{jik}^{(2)}(\omega_3 = \omega_1 + \omega_2) \\ &= \chi_{kji}^{(2)}(\omega_3 = \omega_1 + \omega_2). \end{aligned} \quad (1.39)$$

1.1.6 Contracted notation

If Kleinman's symmetry holds, one often uses a notation for the components of the susceptibility that takes this symmetry into account. To this end, let us first introduce the following tensor:

$$d_{ijk} = \frac{1}{2}\chi_{ijk}^{(2)}, \quad (1.40)$$

and let us suppress the frequency arguments because there is no dispersion when Kleinman's symmetry holds true. The factor 1/2 is a historic convention[1].

Then we can write the nonlinear polarization as:

$$P_i(\omega_n + \omega_m) = \epsilon_0 \sum_{jk} \sum_{(nm)} 2d_{ijk} E_j(\omega_n) E_k(\omega_m). \quad (1.41)$$

If Kleinman's symmetry holds, then one can always assume that d_{ijk} is symmetric in its last two indices. Moreover, this is always true in the case of second-harmonic generation because in this case we have $\omega_n = \omega_m$.

The contracted notation then is to write d_{il} with only two indices instead of three, where the indices jk are combined into the index l using the following prescription:

$$\begin{array}{rcccccc}jk : & 11 & 22 & 33 & 23, 32 & 31, 13 & 12, 21 \\l : & 1 & 2 & 3 & 4 & 5 & 6\end{array}\quad (1.42)$$

Following Kleinman's symmetry, the indices of d_{ijk} can be freely permuted. Because of that, not all components of d_{il} are independent. For example, one has:

$$\begin{aligned}d_{12} &\equiv d_{122} = d_{212} \equiv d_{26} \text{ and} \\d_{14} &\equiv d_{123} = d_{213} \equiv d_{25}.\end{aligned}\quad (1.43)$$

By applying these arguments for all components, one can show that:

$$d_{il} = \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{bmatrix}.\quad (1.44)$$

1.1.7 Effective value of d : d_{eff}

If the directions of propagation and the polarizations of the fields are fixed, one can simplify the expressions for the nonlinear polarization in the context of sum-frequency generation and use the following scalar relationship:

$$P(\omega_3) = 4\epsilon_0 d_{\text{eff}} E(\omega_1) E(\omega_2).\quad (1.45)$$

For second-harmonic generation, this simplifies further to:

$$P(2\omega) = 2\epsilon_0 d_{\text{eff}} E(\omega)^2,\quad (1.46)$$

where $E(\omega) = |\vec{E}(\omega)|$, and $P(\omega) = |\vec{P}(\omega)|$.

One can calculate d_{eff} for each crystal class. For example, in the case that the two lower-frequency waves have the same polarization (type-I phase matching) and assuming that the medium is a negative uniaxial crystal of crystal class $3m$, one gets[1]:

$$d_{\text{eff}} = d_{31} \sin \theta - d_{22} \cos \theta \sin 3\phi,\quad (1.47)$$

where θ is the angle between the direction of propagation and the crystal's z axis, and ϕ is the azimuthal angle between the direction of propagation and the xz plane of the crystal.

When the two incoming waves in the same type of crystal have orthogonal polarization (type-II phase matching), one gets:

$$d_{\text{eff}} = d_{22} \cos^2 \theta \cos 3\phi.\quad (1.48)$$

Triclinic	$\begin{bmatrix} xx & xy & xz \\ yx & yy & yz \\ zx & zy & zz \end{bmatrix}$
Monoclinic	$\begin{bmatrix} xx & 0 & xz \\ 0 & yy & 0 \\ zx & 0 & zz \end{bmatrix}$
Orthorhombic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$
Tetragonal	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$
Trigonal	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$
Hexagonal	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$
Cubic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & xx \end{bmatrix}$
Isotropic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & xx \end{bmatrix}$

Table 1.1: The form of the linear susceptibility $\chi^{(1)}$ for all seven crystal classes as well as for isotropic media. Each non-vanishing component is denoted by its cartesian indices. The table structure and content reproduce a table in Ref. [1].

1.1.8 Spatial symmetry of a nonlinear medium

Apart from the general symmetry considerations we discussed earlier, additional symmetries in the susceptibility tensor result from symmetry properties of the optical medium. As an example[1], consider a crystal, where the x and y directions are equivalent while the z direction is different. That means that a rotation by 90 degrees around the z axis will not lead to a noticeable change of the physical system. In such a crystal, we expect there to be no difference if an optical field is polarized along the x direction or along the y direction. For that reason, we should have $\chi_{zxx}^{(2)} = \chi_{zyy}^{(2)}$.

All crystals can be categorized into 32 crystal classes, and the symmetry properties of each of these classes then result in corresponding symmetry properties for the susceptibility tensor. While we will not go into the detailed meaning of how these crystal classes are defined, suffice it to say that the crystal classes can be derived from group theoretical considerations. In particular, the 32 crystal classes are related to the point group symmetry of crystals.

1.1.9 Influence of spatial symmetry on the linear optical properties

We will first discuss the influence of spatial symmetry on the first-order susceptibility. The symmetry properties of the crystal result in symmetries of the $\chi^{(1)}$ susceptibility tensor. Table 1.1 illustrates these symmetries for all seven crystal classes as well as for

Crystal system	Crystal class	Nonvanishing tensor elements	
Triclinic	$\bar{1} = C_1$	All elements are independent and nonzero	
Monoclinic	$\bar{1} = S_2$	Each element vanishes	
	$2 = C_2$	$xyz, xzy, xxy, xyx, yxx, yyy, yzz, yzx, yxz, zyz,$ zzy, xxy, zyx (twofold axis parallel to \hat{y})	
Orthorhombic	$m = C_{1h}$	$xxx, xyy, xzz, xzx, xxz, yyz, yzy, yxy, yyx, zxx,$ zyy, zzz, zxz (mirror plane perpendicular to \hat{y})	
	$2/m = C_{2h}$	Each element vanishes	
	$222 = D_2$	xyz, xzy, yzx, zxy, zyx	
	$mm2 = C_{2v}$	$xzx, xxz, yyz, yzy, zxx, zyy, zzz$	
Tetragonal	$mmm = D_{2h}$	Each element vanishes	
	$4 = C_4$	$yyz = -yxz, xzy = -yzx, xzx = yzy, xxz = yyz,$ $zxx = zyy, zzz, zxy = -zyx$	
	$\bar{4} = S_4$	$xyz = -yxz, xzy = yzx, xzx = -yzy, xxz = -yyz,$ $zxx = -zyy, zxy = zyx$	
	$422 = D_4$	$xyz = -yxz, xzy = -yzx, zxy = -zyx$	
	$4mm = C_{4v}$	$xzx = yzy, xxz = yyz, zxx = zyy, zzz$	
	$\bar{4}2m = D_{2d}$	$xyz = yxz, xzy = yzx, zxy = zyx$	
	$4/m = C_{4h}$	Each element vanishes	
	$4/mmm = D_{4h}$	Each element vanishes	
	Cubic	$432 = O$	$xyz = -xzy = yzx = -yxx = zxy = -zyx$
		$\bar{4}3m = T_d$	$xyz = xzy = yzx = yxz = zxy = zyx$
$23 = T$		$xyz = yzx = zxy = xzy, xzy = yxz = zyx$	
$m\bar{3} = T_h, m\bar{3}m = O_h$		Each element vanishes	
Trigonal	$3 = C_3$	$zxx = xyy = -yyz = -yxy, xyz = -yxz, xzy = -yzx,$ $xzx = yzy, xxz = yyz, yyy = -yxx = -xxy = -xyx,$ $zxx = zyy, zzz, zxy = -zyx$	
	$32 = D_3$	$zxx = -xyy = -yyx = -yxy, xyz = -yxz,$ $xzy = -yzx, zxy = -zyx$	
	$3m = C_{3v}$	$xzx = yzy, xxz = yyz, zxx = zyy, zzz, yyy = -yxx =$ $-xxy = -xyx$ (mirror plane perpendicular to \hat{x})	
Hexagonal	$\bar{3} = S_6, \bar{3}m = D_{3d}$	Each element vanishes	
	$6 = C_6$	$xyz = -yxz, xzy = -yzx, xzx = yzy, xxz = yyz,$ $zxx = zyy, zzz, zxy = -zyx$	
	$\bar{6} = C_{3h}$	$xxx = -xyy = -yxy = -yyx,$ $yyy = -yxx = -xyx = -xxy$	
	$622 = D_6$	$xyz = -yxz, xzy = -yzx, zxy = -zyx$	
	$6mm = C_{6v}$	$xzx = yzy, xxz = yyz, zxx = zyy, zzz$	
	$\bar{6}m2 = D_{3h}$	$yyy = -yxx = -xxy = -xyx$	
	$6/m = C_{6h}$	Each element vanishes	
	$6/mmm = D_{6h}$	Each element vanishes	

Table 1.2: Non-vanishing components for each of the 32 crystal classes. Table structure and content from Ref. [1].

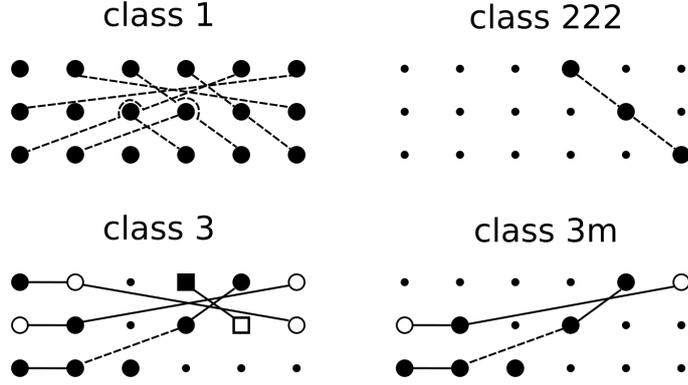


Table 1.3: Form of the d_{ij} matrix for some of the crystal classes lacking inversion symmetry. The upper two classes correspond to biaxial crystals, the lower two classes are uniaxial crystals. Small dot: zero coefficient; large dot: nonzero coefficient; square: coefficient that is zero when Kleinman’s symmetry is fulfilled; connected symbols: numerically equivalent coefficients, but if the symbol is empty, the sign is opposite. Dashed connections are only valid if Kleinmann’s symmetry is valid. Figures based on Ref.[1].

isotropic media.

1.1.10 Influence of inversion symmetry on 2nd-order nonlinearity

We discussed this case already briefly in section 1.1.4. There, we concluded from a nonlinear modification of the Lorentz model that the lowest-order non-linear effects in an inversion symmetric medium would have to be of 3rd order. Here, we will arrive at the same conclusion in a more general fashion. This is a very important result because 11 out of the 32 existing crystal classes are centrosymmetric. No crystal from these 11 classes shows 2nd-order nonlinear optical effects. While this result can be shown to be true in general, for the sake of simplicity, we will demonstrate it here only for second harmonic generation (SHG) following the treatment in Ref. [1]. In particular, we will assume a medium that responds instantaneously to the applied optical fields.

Given the geometry is fixed, we can express the polarization using scalar quantities as we did earlier:

$$P(t) = \epsilon_0 \chi^{(2)} E^2(t), \tag{1.49}$$

where we assume $E(t) = E_0 \cos \omega t$ to be the applied field.

If we assume inversion symmetry, a change of the sign of $E(t)$ must also result in a

change of the sign of $P(t)$. Using equation 1.49, we must therefore have:

$$-P(t) = \epsilon_0 \chi^{(2)} [-E(t)]^2 = \epsilon_0 \chi^{(2)} E^2(t) = P(t). \quad (1.50)$$

This equation can only be correct if $P(t) = 0$, and because this is true for any $E(t)$ fulfilling our initial assumptions, we must have:

$$\chi^{(2)} = 0. \quad (1.51)$$

1.1.11 Influence of spatial symmetry on 2nd-order nonlinearity

Similar to the consequences of inversion symmetry, which we just discussed, other symmetries of the various crystal classes will also result in symmetries of the nonlinear susceptibility. These properties are summarized in Table 1.2.

One can also display these symmetry properties graphically. Table 1.3 shows a few examples of such graphical representations.

1.2 The electro-optic effect

In some materials, the refractive index can be changed by applying an electric field. This is called the linear electro-optic effect or the Pockels effect. It is a convenient way to quickly modulate the amplitude of polarized light or to generate frequency side-bands, but we will discuss that later. We can describe the linear electro-optic effect using the 2nd-order susceptibility. If the applied field is DC, then the polarization of the medium will be:

$$P_i(\omega) = 2\epsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(\omega = \omega + 0) E_j(\omega) E_k(0). \quad (1.52)$$

As we discussed in section 1.1.10, 2nd-order non-linear effects can only occur in noncentrosymmetric media. While it is possible to describe the linear electro-optic effect using this description, due to historic convention we will use a different formalism, which we will introduce in subsection.

In centrosymmetric media, one can still have higher-order electro-optic effects. The lowest order electro-optic effect in such materials is an effect where the refractive index depends quadratically on the amplitude of the applied low-frequency electric field. This phenomenon is known as the Kerr electro-optic effect or as the quadratic electro-optic effect. It is often referred to as Kerr effect, but this would make it easy to confuse with the Kerr magneto-optic effect.

1.2.1 The linear electro-optic effect

In this subsection, we will introduce the usual mathematical description of the linear electro-optic effect. In an anisotropic medium, the relation between the electric field \vec{E} and the electric displacement field \vec{D} is mediated by the dielectric permeability tensor ϵ_{ij} :

$$D_i = \epsilon_0 \sum_j \epsilon_{ij} E_j. \quad (1.53)$$

If the medium is lossless and non-optically active, then ϵ_{ij} is symmetric and real. That means it will have 6 independent elements: ϵ_{xx} , ϵ_{yy} , ϵ_{zz} , $\epsilon_{xy} = \epsilon_{yx}$, $\epsilon_{xz} = \epsilon_{zx}$, and $\epsilon_{yz} = \epsilon_{zy}$. Symmetric, real matrices can in general be brought into diagonal form by using an orthogonal transformation[1] - that means by going into an appropriate coordinate system with coordinates (X, Y, Z) . In that system, equation 1.53 becomes:

$$\begin{bmatrix} D_X \\ D_Y \\ D_Z \end{bmatrix} = \epsilon_0 \begin{bmatrix} \epsilon_{XX} & 0 & 0 \\ 0 & \epsilon_{YY} & 0 \\ 0 & 0 & \epsilon_{ZZ} \end{bmatrix} \begin{bmatrix} E_X \\ E_Y \\ E_Z \end{bmatrix}. \quad (1.54)$$

This coordinate system is called the principal-axis system.

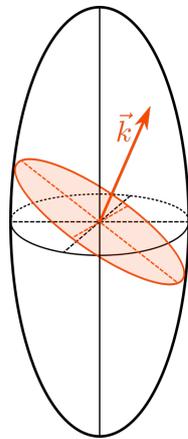


Figure 3: **The optical indicatrix.** Equation defines an ellipsoid. If one cuts the ellipsoid with a plane that is perpendicular to the direction of propagation (\vec{k}) and passes through the origin, one gets an ellipse (indicated in red). The semimajor and semiminor axes of the ellipse are drawn as red, dashed lines. These axes give the two allowed values of the refractive index for the given propagation direction.

It will be useful to consider the energy density per unit volume inside the medium:

$$U = \frac{1}{2} \vec{D} \cdot \vec{E} = \frac{1}{2} \epsilon_0 \sum_{ij} \epsilon_{ij} E_i E_j = \frac{1}{2\epsilon_0} \left[\frac{D_X^2}{\epsilon_{XX}} + \frac{D_Y^2}{\epsilon_{YY}} + \frac{D_Z^2}{\epsilon_{ZZ}} \right]. \quad (1.55)$$

From this equation, we can conclude that surfaces of constant energy density in \vec{D} -space are ellipsoids. If we define the quantities:

$$X = \left(\frac{1}{2\epsilon_0 U} \right)^{1/2} D_X, Y = \left(\frac{1}{2\epsilon_0 U} \right)^{1/2} D_Y, \quad Z = \left(\frac{1}{2\epsilon_0 U} \right)^{1/2} D_Z, \quad (1.56)$$

then equation 1.55 becomes:

$$\frac{X^2}{\epsilon_{XX}} + \frac{Y^2}{\epsilon_{YY}} + \frac{Z^2}{\epsilon_{ZZ}} = 1. \quad (1.57)$$

The ellipsoid defined by this equation is known as the *optical indicatrix* or the *index ellipsoid*. The form of the equation describing the indicatrix is simplest in the principal-axis coordinate system. In a different coordinate system, we can write this equation as:

$$\begin{aligned} \left(\frac{1}{n^2} \right)_1 x^2 + \left(\frac{1}{n^2} \right)_2 y^2 + \left(\frac{1}{n^2} \right)_3 z^2 + \left(\frac{1}{n^2} \right)_4 yz + \\ \left(\frac{1}{n^2} \right)_5 xz + \left(\frac{1}{n^2} \right)_6 xy = 1, \end{aligned} \quad (1.58)$$

where the coefficients $(1/n^2)_i$ are optical constants describing the indicatrix in this new coordinate system. One can describe them in terms of ϵ_{XX} , ϵ_{YY} , and ϵ_{ZZ} by using standard coordinate transformation rules.

Fig. 3 illustrates the indicatrix. If we cut the plane that is perpendicular to the direction of propagation (along \vec{k}) and passes through the coordinate origin with the indicatrix, we will get an ellipse. Depending on the orientation of the polarization of the field, this allows us to calculate the refractive index the field will experience when passing through the material.

If we apply an electric field to our material, the shape of the indicatrix will change due to the linear electro-optic effect. One can conveniently describe this change by using the *impermeability tensor* η_{ij} , which we define via the following relation:

$$E_i = \frac{1}{\epsilon_0} \sum_j \eta_{ij} D_j. \quad (1.59)$$

This relation is the inverse of equation 1.53. That means, we have $\eta_{ij} = (\epsilon^{-1})_{ij}$.

Let us express our indicatrix in terms of the impermeability tensor. First, we note that the energy density can be written as $U = (2\epsilon_0) \sum_{ij} \eta_{ij} D_i D_j$. If we then transform to a new coordinate system x, y, z fulfilling $x = D_x / (2\epsilon_0 U)^{1/2}$ etc, the expression for U becomes:

$$1 = \eta_{11}x^2 + \eta_{22}y^2 + \eta_{33}z^2 + 2\eta_{12}xy + 2\eta_{23}yz + 2\eta_{13}xz. \quad (1.60)$$

If we compare this with our equation 1.58 for the indicatrix in a general coordinate system, we get:

$$\begin{aligned} \left(\frac{1}{n^2}\right)_1 &= \eta_{11}, & \left(\frac{1}{n^2}\right)_2 &= \eta_{22}, & \left(\frac{1}{n^2}\right)_3 &= \eta_{33}, \\ \left(\frac{1}{n^2}\right)_4 &= \eta_{23} = \eta_{32}, & \left(\frac{1}{n^2}\right)_5 &= \eta_{13} = \eta_{31}, & \left(\frac{1}{n^2}\right)_6 &= \eta_{12} = \eta_{21}. \end{aligned} \quad (1.61)$$

The next step is to express the impermeability tensor as a power series in the strength of the components E_k of the electric field:

$$\eta_{ij} = \eta_{ij}^{(0)} + \sum_k r_{ijk} E_k + \sum_{kl} s_{ijkl} E_k E_l + \dots \quad (1.62)$$

Here, we introduced the tensor r_{ijk} , which describes the linear electro-optic effect, and the tensor s_{ijkl} , which describes the quadratic electro-optic effect. We will only consider effects up to 2nd order.

Because ϵ_{ij} is real and symmetric, the same will be true for its inverse η_{ij} . For the same reason, the tensor r_{ijk} must be symmetric in its first two indices. The same holds true for the tensor s_{ijkl} . The latter is also symmetric in its last two indices due to the same argument we had earlier regarding inherent permutation symmetry. In particular, because we sum over the last two indices and multiply with a product of $E_k E_l$, we can always switch the order of the two fields and should get the same result.

Due to these symmetry considerations, it is useful once again to introduce a contracted notation, where we replace two indices ij with one index l according to the prescription we gave in equation 1.42. For convenience, we will repeat this equation here:

$$\begin{array}{l} jk : \quad 11 \quad 22 \quad 33 \quad 23, 32 \quad 31, 13 \quad 12, 21 \\ l : \quad 1 \quad 2 \quad 3 \quad 4 \quad 5 \quad 6 \end{array} \quad (1.63)$$

Using this contracted notation, we can replace r_{ijk} with r_{lk} , and we can express deviations of the optical constants $(1/n^2)_l$ from their zero-order form as:

$$\Delta \left(\frac{1}{n^2}\right)_l = \sum_j r_{lj} E_j. \quad (1.64)$$

To see that this is correct, let us first rewrite equation 1.61 as:

$$\left(\frac{1}{n^2}\right)_l = \eta_{ij}^{(0)}, \quad (1.65)$$

where the cartesian indices ij correspond to the contracted index l .

If we neglect contributions of higher than linear order in equation 1.62, then the deviations of η_{ij} from its zero-order expression $\eta_{ij}^{(0)}$ are given by equation 1.64. The quantities r_{lj} are called electro-optic coefficients and describe the rate with which the coefficients $(1/n^2)_l$ change as we apply an electric field to our material.

We noted earlier that there are no quadratic nonlinear optical effects in centrosymmetric materials. That also holds true for the electro-optic effect. In addition to that, any rotational symmetries of the material will have a direct impact on which elements of r_{lj} are non-zero. As an example, let us consider the form of the electro-optic coefficients for Lithium Niobate (LiNbO_3). This is a crystal of class $3m$ often used in optical experiments. Due to the symmetry properties of this crystal class, several coefficients vanish and others have to be equal:

$$r_{lj} = \begin{bmatrix} 0 & -r_{22} & r_{13} \\ 0 & r_{22} & r_{13} \\ 0 & 0 & r_{33} \\ 0 & r_{42} & 0 \\ r_{42} & 0 & 0 \\ r_{22} & 0 & 0 \end{bmatrix} \quad (\text{for class } 3m) \quad (1.66)$$

1.2.2 Electro-optic modulators

To illustrate the principles of the electro-optic effect, let us discuss an example. In particular, consider we use a Potassium dihydrogen phosphate (KH_2PO_4 or KDP) crystal. It is a uniaxial crystal that belongs to the crystal class $\bar{4}2m$, which means that r_{lj} takes the following form:

$$r_{lj} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \quad (\text{for class } \bar{4}2m). \quad (1.67)$$

If we do not apply an electric field to the crystal, the indicatrix is given by the following

equation in the principle-axis coordinate system:

$$\frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2} = 1, \quad (1.68)$$

where n_o and n_e are the ordinary and extraordinary refractive indices of the crystal, respectively.

If we apply an electric field, then the shape of the indicatrix changes according to equation 1.60, which becomes:

$$\frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2} + 2r_{41}E_XYZ + 2r_{41}E_YXZ + 2r_{63}E_ZXY = 1. \quad (1.69)$$

For KDP, we have $r_{41} = 8.77 \text{ pm/V}$, $r_{63} = 10.5 \text{ pm/V}$, $n_o = 1.514$, and $n_e = 1.472$. The refractive indices here are given for a wavelength of 546.1 nm. The values provided here are reproduced from Ref.[1].

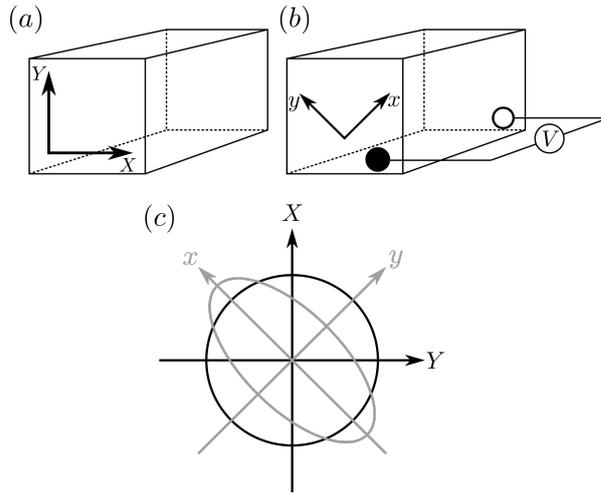


Figure 4: **Linear electro-optic effect in KDP.** (a) shows the orientation of the principal axes in the crystal. (b) When we apply an electric field, we can diagonalize the indicatrix in a rotated coordinate system, indicated by x, y . (c) This figure shows a cut through the indicatrix perpendicular to the Z (or z) axis. Without a field, the refractive index is independent of the direction as indicated by the black circle. With an applied field, the cut becomes an ellipse with refractive indices n_x and n_y along the semimajor and semiminor axis, respectively.

You can see that equation 1.70 contains off-diagonal terms. That means when we apply an electric voltage, the field does not simply change n_o and n_e , but the orientation of the indicatrix depending on the orientation of the electric field applied. A special case that is often encountered in experiments is that the field is applied along one of the principal

axes of the crystal. For example, consider the case where the E-field is applied along the Z axis. Then the equation for the indicatrix simplifies to:

$$\frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2} + 2r_{63}E_ZXY = 1. \quad (1.70)$$

In the exercises, we will find a new principal-axis system with coordinates x, y, z such that equation 1.70 becomes:

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_e^2} = 1, \quad (1.71)$$

where the refractive indices along the three coordinate axes are given by $n_z = n_e$ and:

$$\begin{aligned} n_x &= n_o - \frac{1}{2}n_o^3r_{63}E_z \\ n_y &= n_o + \frac{1}{2}n_o^3r_{63}E_z. \end{aligned} \quad (1.72)$$

Fig. 4 illustrates the action of an applied field $E/V/L$ on the orientation of the principal axes, where V is the applied voltage, and L is the length of the crystal. The shape of the electrodes in Fig. 4(b) should not be taken literally. For example, the electrodes could be gold coating covering the two faces of the crystal. For the figure, it was assumed that $r_{63}E_z$ is negative. In that case, $n_y < n_x$.

Let us now consider an optical beam travelling through the crystal along its z axis. For this to be possible, the electrodes on the crystal surfaces must have apertures to allow a beam to pass, of course. If the light has a polarization that is not along the x or y axis, the components of the E-field along these two axes will acquire a phase difference Γ because of the difference in the refractive indices:

$$\Gamma = (n_y - n_x) \frac{\omega L}{c}. \quad (1.73)$$

This phase difference is called retardation, and it introduces a rotation in the polarization of the light sent through. If we insert the expressions from equation 1.72, we find:

$$\Gamma = \frac{n_o^3r_{63}E_z\omega L}{c} = \frac{n_o^3r_{63}V\omega}{c}. \quad (1.74)$$

That means, the retardation in this configuration will not depend on the crystal length but only on the voltage applied.

A device of this type is called an electro-optic modulator (EOM). Sometimes, they are also called Pockels cells. If we apply the correct voltage to our crystal, we can achieve a π phase shift between the polarization components along x and y . This voltage is called

the half-wave voltage:

$$V_{\lambda/2} = \frac{\pi c}{\omega n_o^3 r_{63}}. \quad (1.75)$$

Depending on the crystal, the half-wave voltage for configurations like the one we discussed here are typically in the range of several kV or around 10 kV for visible or near-infrared light.

Depending on the voltage applied and the resulting retardation, we can use an EOM to switch between different polarization states or to modulate the intensity of a light beam.

Intensity modulation using an EOM

In today's exercises, we will discuss one common application of EOMs. By placing an EOM between crossed polarizers, one can modulate the intensity of the light passing through by changing the voltage applied to the EOM.

Phase and frequency modulation with an EOM

Instead of using an EOM to change the polarization of the light passing the crystal, one can set the polarization to be parallel to the principal axis of the crystal. In that case, the EOM will not modulate the amplitude or the intensity of the light, but it can modulate the phase and/or frequency. Let us consider how an EOM in this particular orientation of the polarization along the principal axis of the crystal will influence the phase and the frequency of the light.

In this configuration, the crystal will still introduce a relative phase compared to if we would not apply any voltage. If our light is polarized along the x axis, that phase will be:

$$\phi = (n_x - n_o) \frac{\omega L}{c} = -\frac{\omega r_{63} n_o^3 V}{2c}. \quad (1.76)$$

Let us now assume that we do not simply apply a constant field to the EOM but an oscillating field with an angular frequency Ω :

$$E(t) = E_0 \sin(\Omega t) = \frac{V(t)}{L} = \frac{V_0}{L} \sin(\Omega t). \quad (1.77)$$

If we apply this varying electric field to our crystal, we will get the following:

$$\begin{aligned} \phi(t) &= \omega t + m \sin(\Omega t), \\ E(t) &= \text{Re} \{ E_0 \exp(-i\omega t) \exp[-im \sin(\Omega t)] \}. \end{aligned} \quad (1.78)$$

Here, m is the modulation index, which reflects the amplitude of the modulation. It depends on the material parameters and the amplitude of the applied voltage via the

relation:

$$m = \frac{\omega r_{63} n_o^3 V_0}{2c}. \quad (1.79)$$

In addition to this phase modulation, there will also be a modulation of the light's instantaneous frequency:

$$\omega(t) = \frac{d\phi(t)}{dt} = \omega + m\Omega \cos(\Omega t). \quad (1.80)$$

While it is not strictly possible to separate phase modulation (PM) from frequency modulation (FM) in this situation, one can roughly categorize the effect one will see in the following way[2]:

- for $m < 1$: PM
- for $m > 1$: FM

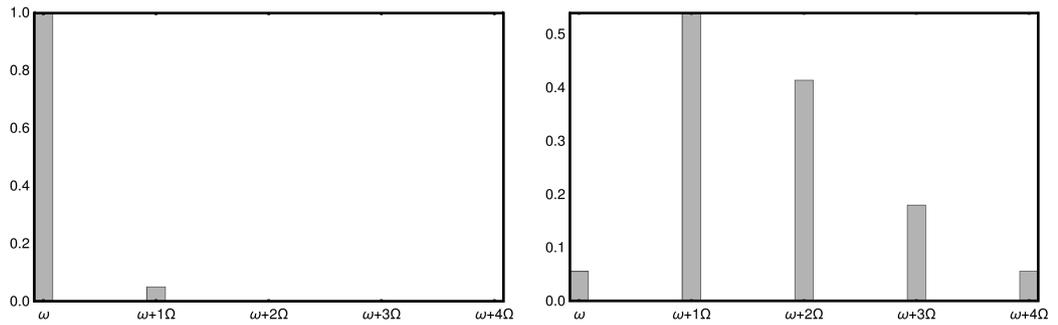


Figure 5: **Sideband amplitudes for $\omega + m\Omega$.** The figure is intended to illustrate the regime where we mostly have phase modulation vs the regime where we have significant frequency modulation. The left and right figures are for $m = 0.1$ and for $m = 2.3$, respectively. (left) For small m , most light will remain at the fundamental frequency. (right) for larger m , a significant amount of light will be in the sidebands. For simplicity, the figure does not show the negative sidebands.

Let us look at this more carefully. In particular, let us decompose our electromagnetic wave into its frequency components. The field we apply to the crystal is proportional to[2]:

$$e^{-im \sin(\Omega t)} = J_0 + 2[J_2(m) \cos(2\Omega t) + J_4(m) \cos(4\Omega t) + \dots] - 2i[J_1(m) \sin(\Omega t) + J_3(m) \sin(3\Omega t) + \dots],$$

where the J_n are Bessel functions of the 1st kind. We can then write $E(t)$ as:

$$E(t) = E_0 e^{-i\omega t} [J_0(m) + J_1(m)(e^{-i\Omega t} - e^{i\Omega t}) + \quad (1.81)$$

$$J_2(m)(e^{-2i\Omega t} + e^{2i\Omega t}) + J_3(m)(e^{-3i\Omega t} - e^{3i\Omega t}) + \dots]. \quad (1.82)$$

This decomposition shows that the spectrum of the field will have a fundamental peak at ω but the spectrum will also have peaks at $\omega \pm k\Omega$ for $k \geq 1$. The signals leading to these peaks are called *sidebands*. Fig.5 shows how the light becomes distributed into various frequency sidebands depending on the size of the modulation index m . For low m , most of the light remains at the fundamental (or “carrier”) frequency. For higher m , the carrier becomes more and more suppressed. For $m = 2.4$, the carrier even vanishes and all the light intensity will be in the sidebands.

1.3 The magneto-optic effect

In the last section, we saw that we can use electric fields that are stable or vary at low frequencies in order to modulate the optical properties of certain nonlinear crystals. For some materials, it is also possible to affect the light passing through a crystal by applying a magnetic field to the crystal. We can distinguish two magneto-optic effects. The first is the Faraday effect, where the polarization of light is affected when it passes through material to which a magnetic field is applied. The second effect is the magneto-optic Kerr effect, which affects light reflected from the surface of a material to which a magnetic field is applied. Here, we will give a short introduction to the Faraday effect. We will not go into details about the Kerr magneto-optic effect.

1.3.1 The Faraday effect

Consider light passing through a medium along the same axis along which a magnetic field is applied (see Fig. 6). If we assume that the incident field is polarized linearly along \vec{E} , then the polarization \vec{E}' after the medium will be given by:

$$\vec{E}' = \begin{pmatrix} \cos \alpha & -\sin \alpha \\ \sin \alpha & \cos \alpha \end{pmatrix} \vec{E}. \quad (1.83)$$

The rotation angle α depends on the magnetic field strength and the length L of the medium:

$$\alpha = V|\vec{B}|L, \quad (1.84)$$

where V is the Verdet constant, which depends on the material chosen. Example values for light of 589 nm are $209^\circ\text{m}^{-1}\text{T}^{-1}$ for quartz, $528^\circ\text{m}^{-1}\text{T}^{-1}$ for heavy flint, and $-145^\circ\text{m}^{-1}\text{T}^{-1}$ for terbium-gallium garnet[2].

The material results in different phase shifts for left-hand and right-hand circularly

polarized light. Because these two polarizations form a basis for all polarizations, this leads to an effective rotation of polarization when light passes through such a medium in the presence of a magnetic field. A very interesting fact is that this effect is not directionally reversible. That means, if light is back-reflected after passing through such a material, the light does experience twice as much rotation. For that reason, the Faraday effect is often applied as a means to build *optical isolators*, which prevent back reflection of light.

To understand the working principle of an optical isolator, consider the following situation: (1) incident light passes through a horizontal polarizer. (2) it then is rotated by 45° in a Faraday rotator. (3) at the output, the light passes through a 45° polarizer. Light that is backreflected, will first have to pass through the 45° polarizer. Then it is rotated by 45° in the Faraday rotator and becomes vertically polarized. It then will be blocked by the horizontal polarizer.

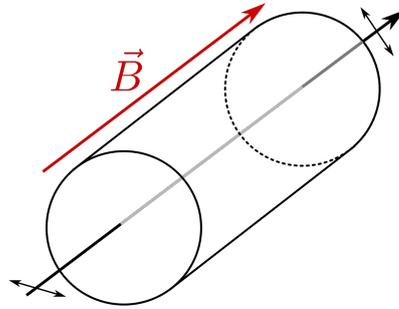


Figure 6: **Faraday effect.** A beam of light passes through a medium along the arrow indicated. A magnetic field \vec{B} pointing in the same direction is applied to the medium. Due to the Faraday effect, the polarization of the beam will rotate as it rotates through the medium. The amount of rotation will depend on the magnetic field applied.

1.4 Acousto-optics

In contrast to the electro-optic effect and the magneto-optic effect, this is not a non-linear optical effect. Here, a sound-wave is applied to an optical medium - for example, via the means of an external piezo-electric transducer. Because sound waves inside a solid go along with changes to material density. In turn, that can influence the optical properties of the material.

One can classify acousto-optic devices into two main categories, depending on the typical length scales involved:

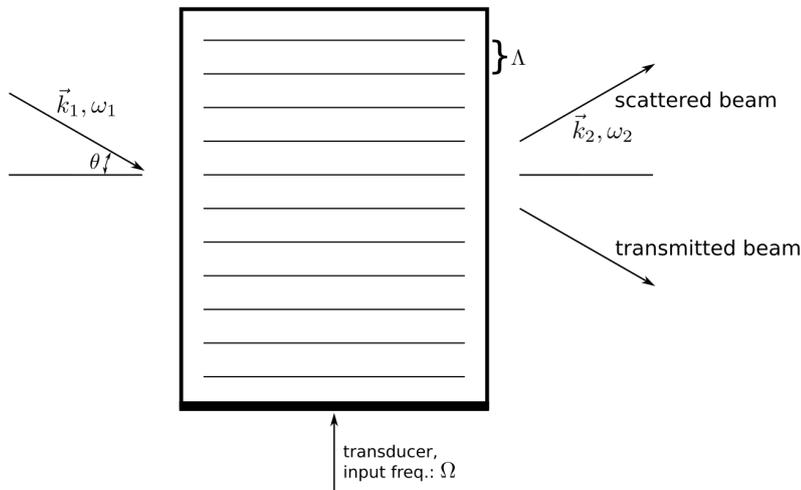


Figure 7: **Bragg acousto-optic modulation.** An rf signal with angular frequency Ω applied to a transducer generates an acoustic wave with wavelength Λ . An incoming beam with optical frequency ω_1 and wavevector \vec{k}_1 is diffracted. The scattered beam has ω_2 and \vec{k}_2 , part of the beam is transmitted.

- Bragg scattering: We operate in this regime in case the interaction length is long, such that phase-matching conditions become important (we will discuss that in more detail later). In this case, one has a single diffracted beam.
- Raman-Nath scattering: We operate in this regime if the interaction length is short and phase-matching conditions are not important.

Mostly we will concentrate on the Bragg scattering regime, but before we do that, let us first introduce some central concepts.

In particular, we will discuss the basic concept we need to describe the following situation: assume that an acoustic sound wave passes through a medium, and that an input beam is scattered by the modulation of the refractive index induced in the medium. We assume that the sound wave is a plane wave with a frequency Ω and a wavelength $\Lambda = 2\pi v/\Omega$ inside the medium. v is the velocity of sound in the medium. This situation is illustrated in Fig. 7.

Spontaneous Brillouin scattering

In particular, the acoustic wave will lead to a modulation $\Delta\chi$ of the susceptibility in the medium. That will lead to an additional polarization in a volume V of our medium:

$$\vec{P} = \Delta\chi\vec{E}_0 = \Delta\epsilon\vec{E}_0. \quad (1.85)$$

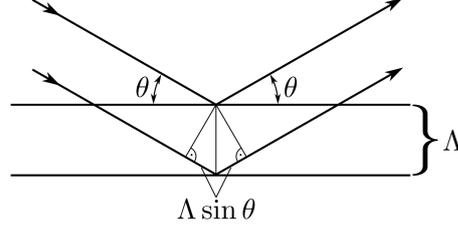


Figure 8: **Bragg condition.** The figure illustrates the condition under which constructive interference occurs between waves reflected at planes with a distance Λ .

For simplicity, we assumed that we can represent the susceptibility as a scalar, and we used the relation $\epsilon = 1 + \chi$. That polarization then leads to an additional dipole moment that will in turn lead to the emission of radiation: scattered light.

Changes to the dielectric constant can be written in dependence of the density ρ and the temperature T of the material:

$$\Delta\epsilon = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T \Delta\rho + \left(\frac{\partial\epsilon}{\partial T}\right)_\rho \Delta T. \quad (1.86)$$

Typically, the dielectric constant depends significantly more on variation of the pressure than on the temperature. For that reason, one can neglect the second term and assume:

$$\Delta\epsilon \approx \left(\frac{\partial\epsilon}{\partial\rho}\right)_T \Delta\rho. \quad (1.87)$$

Instead of pressure and temperature, let us now choose the pressure p and the entropy s as the independent thermodynamic variables. Then we can write changes to the pressure density $\Delta\rho$ as:

$$\Delta\rho = \left(\frac{\partial\rho}{\partial p}\right)_s \Delta p + \left(\frac{\partial\rho}{\partial s}\right)_p \Delta s. \quad (1.88)$$

The first term describes (adiabatic) density fluctuations that represent acoustic waves[1].

The equation of motion for a pressure wave inside the medium is given by[1]:

$$\frac{\partial^2 \Delta p}{\partial t^2} - \Gamma \nabla^2 \frac{\partial \Delta p}{\partial t} - v^2 \nabla^2 \Delta p = 0, \quad (1.89)$$

where Γ is a damping parameter, and v is the speed of sound in the medium. It can be expressed as:

$$v^2 = \frac{K_s}{\rho} = \frac{1}{C_s \rho} = \left(\frac{\gamma k_B T}{\mu}\right)^{1/2}, \quad (1.90)$$

where C_s and K_s are the bulk modulus and the compressibility measured at constant entropy, respectively. μ denotes the molecular mass, k_B is Boltzmann's constant, and γ is the adiabatic index[1]:

$$\gamma = \frac{C_T}{C_s} = \frac{c_p}{c_V}. \quad (1.91)$$

Here C_T is the compressibility measured at constant temperature, and c_p and c_V are the specific heat capacities measured at constant pressure and at constant volume, respectively. The relation between the compressibility, the bulk modulus, the pressure and the material density is given by:

$$C = \frac{1}{K} - \frac{1}{V} \frac{\partial V}{\partial p} = \frac{1}{\rho} \frac{\partial \rho}{\partial p}. \quad (1.92)$$

Let us investigate the behaviour of a plane acoustic wave if we apply equation 1.89. We assume the plane wave to be of the form:

$$\Delta p(\vec{r}, t) = \Delta p e^{i(\vec{q} \cdot \vec{r} - \Omega t)} + \text{c.c.} \quad (1.93)$$

If we put this form into equation 1.89, we can see that the following dispersion relation must be fulfilled:

$$\Omega^2 = q^2(v^2 - i\Omega\Gamma). \quad (1.94)$$

At the same time, incoming light is scattered by the acoustic wave. Let us assume that the incoming field can be written as:

$$\vec{E}_0(\vec{r}, t) = \vec{E}_0 e^{i(\vec{k} \cdot \vec{r} - \omega t)} + \text{c.c.} \quad (1.95)$$

The field inside the medium has to fulfill the wave equation:

$$\nabla^2 \vec{E} - \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}. \quad (1.96)$$

To determine the term on the right-hand side, let us assume that \vec{P} is given by equation 1.85, and that $\Delta\epsilon$ is given by equation 1.87. Moreover, we concentrate only on the first term of equation 1.88. That means, we assume that:

$$\Delta\rho \approx \left(\frac{\partial\rho}{\partial p} \right)_s \Delta p. \quad (1.97)$$

Then we get:

$$\begin{aligned}\vec{P}(\vec{r}, t) &= \epsilon_0 \left(\frac{\partial \epsilon}{\partial \rho} \right) \left(\frac{\partial \rho}{\partial p} \right)_s \Delta p(\vec{r}, t) \vec{E}_0(\vec{r}, t) \\ &= \epsilon_0 \gamma_e C_s \Delta p(\vec{r}, t) \vec{E}_0(\vec{r}, t),\end{aligned}\quad (1.98)$$

where γ_e is the electrostrictive constant given by:

$$\gamma_e = \left(\rho \frac{\partial \epsilon}{\partial \rho} \right)_{\rho=\rho_0}, \quad (1.99)$$

and ρ_0 is the undisturbed material density.

If we put this into our wave equation for the electric field, we finally get the following:

$$\begin{aligned}\nabla^2 \vec{E} - \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} &= - \frac{\gamma_e C_s}{c^2} \left[(\omega - \Omega)^2 E_0 \Delta p^* e^{i(\vec{k}-\vec{q}) \cdot \vec{r} - i(\omega-\Omega)t} + \right. \\ &\quad \left. (\omega + \Omega)^2 E_0 \Delta p e^{i(\vec{k}+\vec{q}) \cdot \vec{r} - i(\omega+\Omega)t} \right],\end{aligned}\quad (1.100)$$

where our interest will be on the right-hand side, which we can split into two separate parts. We will describe them in the following subsection.

Stokes and Anti-Stokes scattering

The first and the second summand comprising the right-hand side of equation 1.100 correspond to *Stokes Scattering* and *Anti-Stokes Scattering*, respectively.

Let us first take a look at the term for Stokes scattering. In that case, the wavevector \vec{k}' and the angular frequency ω' of the scattered field will fulfill:

$$\vec{k}' = \vec{k} - \vec{q}, \quad (1.101)$$

and

$$\omega' = \omega - \Omega. \quad (1.102)$$

It is important to note that satisfying the above equations is not sufficient to warrant efficient scattering of the incoming wave into the outgoing wave described by the equations above. In addition, the wave vector and the frequency of the scattered light needs to fulfill the general optical dispersion relation:

$$\omega' = |\vec{k}'|c/n, \quad (1.103)$$

where n is the refractive index of our medium.

For Anti-Stokes scattering the wavevector and the angular frequency fulfill:

$$\vec{k}' = \vec{k} + \vec{q}, \quad (1.104)$$

and

$$\omega' = \omega + \Omega. \quad (1.105)$$

Again, the general optical dispersion relation in equation 1.103 has to be fulfilled for efficient scattering.

1.4.1 Bragg-scattering of light by acoustic waves

Now let us get back to the situation described in Fig. 7. The incoming beam can be partially reflected at the wavefronts of the acoustic wave inside the medium. For certain angles of incidence, the waves reflected from the various wavefronts can decohere constructively if

$$\lambda = 2\Lambda \sin \theta. \quad (1.106)$$

One can arrive at this condition, which is known as the *Bragg condition*, by considering Fig. 8. In particular, it provides the angle of an incoming wave with respect to crystal planes such that waves reflected at different planes will have a relative delay that is multiples of 2π .

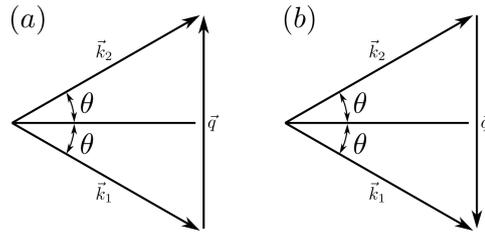


Figure 9: **Bragg condition & phase matching.** We can express the Bragg condition in terms of phase matching. Figures (a) and (b) illustrate that the sign of the wave vector \vec{q} of the acoustic wave must be adapted to the direction the sound wave is traveling.

The Bragg condition can also be understood as a **phase-matching condition**. Similar to equations like 1.101, and 1.102, which we encountered in the context of Brioullin scattering. In particular, let us assume that \vec{k}_1 and \vec{k}_2 represent the wave vectors of the incoming and the scattered wave, respectively, as in Fig. 7, and let us also assume that \vec{q} is the wave vector of the acoustic wave. Then we can write the Bragg condition as:

$$\vec{k}_2 = \vec{k}_1 + \vec{q}. \quad (1.107)$$

Similar to equation 1.105, we will get:

$$\omega_2 = \omega_1 + \Omega. \quad (1.108)$$

Because $\Omega \ll \omega$, the two wave-vectors will have approximately the same length. That means: $|\vec{k}_2| \approx |\vec{k}_1|$.

If the sound waves travel in the opposite directions, we will instead get:

$$\begin{aligned}\vec{k}_2 &= \vec{k}_1 - \vec{q}, \text{ and} \\ \omega_2 &= \omega_1 - \Omega.\end{aligned}\tag{1.109}$$

To accurately describe the theory of Bragg scattering of light by sound, we need to investigate changes to the dielectric constant due to the acoustic waves as we did in the context of Bragg scattering. Strictly speaking, the description there only applies to isotropic media. In the case of anisotropic media, the dielectric constant is a tensor related to the strain tensor S_{ij} and the strain-optic tensor p_{ijkl} as follows:

$$[\Delta(\epsilon^{-1})_{ij}] = \sum_{kl} p_{ijkl} S_{kl}.\tag{1.110}$$

The strain tensor is given by:

$$S_{kl} = \frac{1}{2} \left(\frac{\partial d_k}{\partial x_l} + \frac{\partial d_l}{\partial x_k} \right)\tag{1.111}$$

From equation 1.110 for deviations to the inverse strain tensor, one can calculate the deviations to the strain tensor as follows:

$$(\Delta\epsilon)_{il} = - \sum_{jk} \epsilon_{ij} [\Delta(\epsilon^{-1})]_{jk} \epsilon_{kl}.\tag{1.112}$$

Let us assume a layout as in Fig. 7 with \vec{q} pointing in the z -direction, and x perpendicular to that and along the wavefronts shown in the figure. Once this geometry is fixed, we can describe the incident and diffracted waves, respectively, as follows (pointing in the y direction)[1]:

$$\begin{aligned}E_1 &= A_1 e^{i(\vec{k}_1 \cdot \vec{r} - \omega_1 t)} + \text{c.c.}, \\ E_2 &= A_2 e^{i(\vec{k}_2 \cdot \vec{r} - \omega_2 t)} + \text{c.c.}\end{aligned}\tag{1.113}$$

$A_{1,2}$ are the amplitudes of these fields that vary only slowly in space and time. They only depend on x and z , but *not* on y .

If the interaction volume is sufficiently large, the diffracted beam will (approximately) fulfill Bragg phase matching:

$$\vec{k}_2 \approx \vec{k}_1 + \vec{q}.\tag{1.114}$$

The total electric field will be the superposition of the incoming and the diffracted field ($E = E_1 + E_2$), and that total field needs to fulfill the wave equation in our dielectric

medium:

$$\nabla^2 E - \frac{n^2 + \Delta\epsilon}{c^2} \frac{\partial^2 E}{\partial t^2} = 0. \quad (1.115)$$

Let us solve this equation now in order to gain some basic insights into the process of acoustic modulation. It will facilitate the solution if we separate $\Delta\epsilon$ into a product of rapidly and slowly varying factors. In particular, let us write:

$$\Delta\epsilon = \Delta\tilde{\epsilon} e^{i\vec{q}\cdot\vec{r} - \Omega t} + \text{c.c.} \quad (1.116)$$

The exponential now contains the rapid spatial and time dependence while $\Delta\epsilon$ is assumed to be only slowly varying in space and time.

We can then insert the field amplitudes from equation 1.113 into equation 1.115 and first collect *only* terms that vary periodically with ω_1 . Keeping in mind that $\Omega = \omega_2 - \omega_1$, this leads to the following differential equation (see today's exercises):

$$\begin{aligned} \frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial z^2} + 2ik_{1x} \frac{\partial A_1}{\partial x} + 2ik_{1z} \frac{\partial A_1}{\partial z} - (k_{1x}^2 + k_{1z}^2)A_1 + \\ \frac{n^2\omega_1^2}{c^2} A_1 + \frac{\omega_2^2}{c^2} A_2 \Delta\tilde{\epsilon}^* e^{i(\vec{k}_2 - \vec{k}_1 - \vec{q})\cdot\vec{r}} = 0. \end{aligned} \quad (1.117)$$

Assuming that A_1 varies only slowly in x , we can neglect the 2nd-order derivatives. If we also assume that A_1 does not depend on z , and that the optical dispersion relation holds for \vec{k}_1 and ω_1 , equation 1.117 simplifies to:

$$2ik_{1x} \frac{dA_1}{dx} = -\frac{\omega_2^2}{c^2} A_2 \Delta\tilde{\epsilon}^* e^{i(\vec{k}_2 - \vec{k}_1 - \vec{q})\cdot\vec{r}}. \quad (1.118)$$

A further simplification can be made if one assumes that the extent of our medium is much larger in the z direction than in the x direction. In this case, the phase matching of the z components must be significantly better than for the x components. We can therefore neglect any phase mismatch in the z direction, and we can therefore write:

$$(\vec{k}_2 - \vec{k}_1 - \vec{q}) \cdot \vec{r} = -\Delta k x, \quad (1.119)$$

where we introduced the $\Delta k = |\Delta\vec{k}|$ with $\Delta\vec{k} = \vec{k}_2 - \vec{k}_1 - \vec{q}$.

This will simplify our differential equation 1.117 still further, and if we apply similar arguments as above to solve equation 1.115 for terms oscillating with ω_2 , we get the following two coupled equations for $A_{1,2}$:

$$\begin{aligned} \frac{dA_1}{dx} &= \frac{i\omega_2^2 \Delta\tilde{\epsilon}^*}{2k_{1x} c^2} A_2 e^{-i\Delta x} \\ \frac{dA_2}{dx} &= \frac{i\omega_1^2 \Delta\tilde{\epsilon}}{2k_{2x} c^2} A_1 e^{i\Delta x}. \end{aligned} \quad (1.120)$$

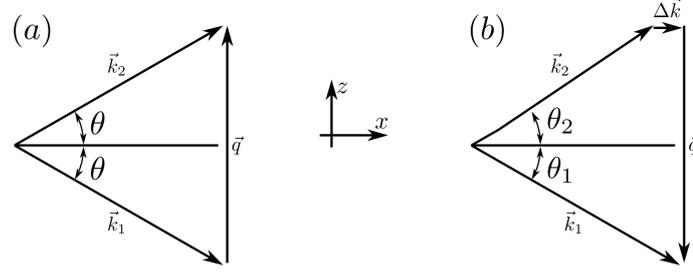


Figure 10: **Phase matching.** (a) shows perfect phase matching ($\Delta k = 0$). (b) shows imperfect phase matching, where there is a non-vanishing $\Delta \vec{k}$ necessary to add up the wave vectors to equal \vec{q} .

These equations can be simplified even further when assuming $\omega_1 \approx \omega_2 \equiv \omega$ and $k_{1x} \approx k_{2x} \equiv k_x$:

$$\begin{aligned} \frac{dA_1}{dx} &= i\kappa A_2 e^{-i\Delta k x} \\ \frac{dA_2}{dx} &= i\kappa^* A_1 e^{i\Delta k x}. \end{aligned} \quad (1.121)$$

Here, we introduced the coupling constant

$$\kappa = \frac{\omega^2 \Delta \tilde{\epsilon}^*}{2k_x c^2}. \quad (1.122)$$

If the incoming beam is incident at the Bragg angle, the phase mismatch will vanish ($\Delta k = 0$), and the solutions of equation 1.121 become particularly simple. For our example here, the solutions become:

$$\begin{aligned} A_1(x) &= A_1(0) \cos(|\kappa|x), \\ A_2(x) &= \frac{i\kappa^*}{|\kappa|} A_1(0) \sin(|\kappa|x), \end{aligned}$$

where the solutions fulfill $|A_1(x)|^2 + |A_2(x)|^2 = |A_1(0)|^2$.

In order to estimate the efficiency of the process, we can calculate the ratio between the input intensity and the intensity of the scattered light:

$$\eta \equiv \frac{|A_2(L)|^2}{|A_1(0)|^2} = \sin^2(|\kappa|L). \quad (1.123)$$

To make numerical estimations given experimental parameters it is useful to express κ in

terms of the intensity of the acoustic wave. That intensity is given by [1]:

$$I = Kv \frac{\langle \Delta \rho^2 \rangle}{\rho_0^2} = 2Kv |\Delta \tilde{\epsilon}|^2 / \gamma_e^2. \quad (1.124)$$

Then the coupling constant becomes:

$$|\kappa| = \frac{\omega \gamma_e}{2nc \cos \theta} \left(\frac{I}{2Kv} \right)^{1/2}. \quad (1.125)$$

In the simplified calculations above, we assumed that the incoming beam is incident at the Bragg angle such that Δk vanishes. This is not the case in general as illustrated in Fig. 10. The vectors in panel (b) of that figure satisfy the following equations:

$$k \cos \theta_1 - k \cos \theta_2 = \Delta k \quad (1.126)$$

$$k \sin \theta_1 + k \sin \theta_2 = q, \quad (1.127)$$

where it is assumed that $k_1 \approx k_2 = k$.

If the incoming light is not incident at the Bragg angle θ_B , we will have $\theta_1 = \theta_B + \Delta\theta$. Typically, we will assume that $\Delta\theta \ll 1$. Equation 1.127 will be satisfied if we have $\theta_2 = \theta_B - \Delta\theta$. If we insert these expressions for $\theta_{1,2}$ into equation 1.126, we get:

$$\cos(\theta_B \pm \Delta\theta) = \cos \theta_B \mp (\sin \theta_B) \Delta\theta, \text{ and} \quad (1.128)$$

$$(2k \sin \theta_B) \Delta\theta = \Delta k, \quad (1.129)$$

resulting in:

$$\Delta k = -\Delta\theta q. \quad (1.130)$$

If we solve the differential equations 1.121 for $\Delta k \neq 0$ for the case in which the external field applied does not include contributions with ω_2 , then we get:

$$\begin{aligned} A_1(x) &= e^{-i\Delta k x/2} A_1(0) \left(\cos sx + i \frac{\Delta k}{2s} \sin sx \right) \\ A_2(x) &= i e^{i\Delta k x/2} A_1(0) \frac{\kappa^*}{s} \sin sx. \end{aligned} \quad (1.131)$$

The parameter s is defined via the relation:

$$s^2 = |\kappa|^2 + (\Delta k/2)^2. \quad (1.132)$$

Using these relations, we can derive a more general relation for the diffraction efficiency:

$$\eta(\Delta k) \equiv \frac{|A_2(L)|^2}{|A_1(0)|^2} = \frac{|\kappa|^2}{|\kappa|^2 + (\Delta k/2)^2} \sin^2 \left\{ \left[|\kappa|^2 + (\Delta k/2)^2 \right]^{1/2} L \right\}. \quad (1.133)$$

1.5 Liquid crystal modulators

As an example, we will discuss a liquid crystal (LC) modulator using molecules in the nematic phase. There also exist other phases like the smectic and the cholesteric phase[2]. In the nematic phase, all the molecular slabs inside a liquid point in one direction. If they are exposed to a surface that imposes a preferred direction (e.g., grooves in the surface), then the molecular slabs will orient themselves along that direction. If such a liquid is placed between two such surfaces where the grooves on one surface are perpendicular to the grooves on the other surface, then the molecular slabs will rotate their orientation along the way from one surface to the other. This is illustrated in Fig. 11. If a voltage is applied between the two surfaces, the molecular slabs orient themselves along the field lines. In the former case, the polarization of a light beam passing through the liquid will be rotated along with the molecular slabs. When the voltage is applied, this is not the case any longer, and the polarization of the light will be unchanged.

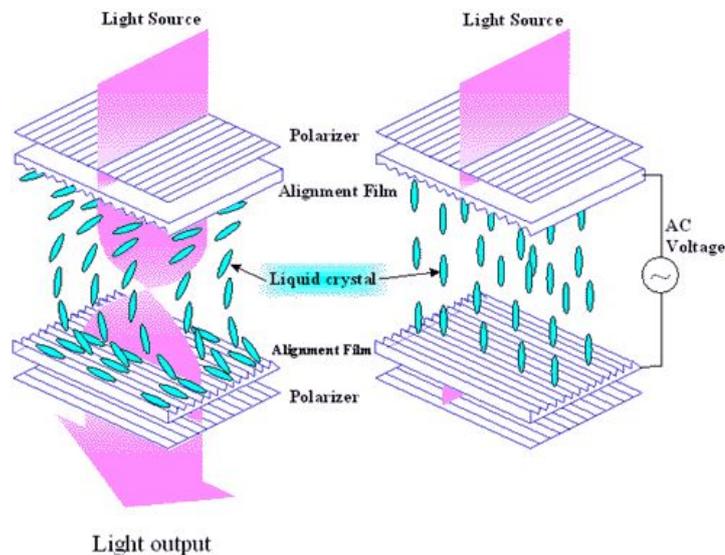


Figure 11: **Liquid crystal modulator.** (left) The molecular slabs orient themselves along the grooves on the surfaces. Between the two surfaces, the slabs reorient themselves. (right) if a voltage is applied, the molecular slabs orient themselves along the field lines. The figure is from Ref. [3].

Liquid crystal displays (LCDs) work based on the same principles, but they typically work in reflection.

2 Wave-equation description of nonlinear effects

In section 1.1, we introduced and briefly discussed a number of 2nd-order nonlinear optical effects. In this chapter, we will discuss the theory describing these effects in more detail, and we will review a few examples of how these effects are put to use in experiments. In the same section, we discussed the Lorentz dipole model in order to get some (classical) insight into key aspects of the nonlinear susceptibility. It is worth repeating the central idea here in order to get a better intuitive understanding of the nonlinear processes we are going to discuss. In the Lorentz model, it is assumed that there is a dipole associated with each atom in a material. Incident electromagnetic waves drive the oscillation of these dipoles, and they will then again emit radiation at the same frequency as the incoming light. In the nonlinear extension of this model, the radiation emitted by the dipoles will not necessarily be at the same frequency as the incident light. This is the origin of the nonlinear optical effects we will discuss here.

This intuitive model also provides some additional relevant insight: in order for there to be a significant amount of radiation exiting the nonlinear material, the dipoles contributing to the process have to emit radiation that is in phase. Only then will the radiation emitted from different dipoles interfere constructively. If all dipoles emitted with random relative phases, the resulting overall field would average out. This is the intuition behind “phase matching” conditions for the various nonlinear processes we will discuss.

2.1 The wave equation for nonlinear optical media

We will mostly be interested in media that are free of charges, free of currents, and that are nonmagnetic. The Maxwell equations for the electromagnetic field in such a medium will then be:

$$\begin{aligned}\vec{\nabla} \cdot \vec{D} &= 0, & \vec{\nabla} \cdot \vec{B} &= 0, \\ \vec{\nabla} \times \vec{E} &= -\frac{\partial \vec{B}}{\partial t}, & \vec{\nabla} \times \vec{H} &= \frac{\partial \vec{D}}{\partial t},\end{aligned}\tag{2.1}$$

where we used:

$$\vec{B} = \mu_0 \vec{H}, \quad \vec{D} = \epsilon_0 \vec{E} + \vec{P}, \quad (2.2)$$

where the polarization vector \vec{P} depends nonlinearly on the field \vec{E} .

Comining these equations results in the most general form of the wave equation for the electromagnetic fields in nonlinear optical media:

$$\vec{\nabla}(\vec{\nabla} \cdot \vec{E}) - \vec{\nabla}^2 \vec{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \vec{E} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}. \quad (2.3)$$

In many cases, e.g., when the electric field is a plane wave, the first term on the left-hand side will vanish, further simplifying the equation. If we assume that, we can write the wave equation as:

$$\vec{\nabla}^2 \vec{E} - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{D}}{\partial t^2} = 0, \quad (2.4)$$

where $\vec{D} = \epsilon_0 \vec{E} + \vec{P}$.

It is often convenient to split \vec{D} and \vec{P} into linear and nonlinear parts as follows:

$$\begin{aligned} \vec{P} &= \vec{P}^{(1)} + \vec{P}^{\text{NL}} \\ \vec{D} &= \vec{D}^{(1)} + \vec{D}^{\text{NL}} = \vec{D}^{(1)} + \vec{P}^{\text{NL}}, \end{aligned}$$

where

$$\vec{D}^{(1)} = \epsilon_0 \vec{E} + \vec{P}^{(1)}.$$

Then the wave equation becomes:

$$\vec{\nabla}^2 \vec{E} - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{D}^{(1)}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}^{\text{NL}}}{\partial t^2}. \quad (2.5)$$

In the context of nonlinear optics, we often deal with dispersive media and with the case where the electromagnetic field as well as the polarization consist of distinct frequency components. To describe that situation, let us split the field, the displacement vector and the polarization into these different frequency components, and for each frequency component, let us assume that the time dependence is periodic with the

respective angular frequency ω_n :

$$\begin{aligned}\vec{E}(\vec{r}, t) &= \sum'_n \vec{E}_n(\vec{r}, t) = \sum'_n \left(\vec{E}_n(\vec{r}) e^{-i\omega t} + \text{c.c.} \right), \\ \vec{D}(\vec{r}, t) &= \sum'_n \vec{D}_n(\vec{r}, t) = \sum'_n \left(\vec{D}_n(\vec{r}) e^{-i\omega t} + \text{c.c.} \right), \\ \vec{P}^{\text{NL}}(\vec{r}, t) &= \sum'_n \vec{P}_n^{\text{NL}}(\vec{r}, t) = \sum'_n \left(\vec{P}_n^{\text{NL}}(\vec{r}) e^{-i\omega t} + \text{c.c.} \right).\end{aligned}\quad (2.6)$$

where the prime indicates that we sum only over the positive frequency parts.

If we introduce the following short-hand notation for the product between the dielectric tensor and the e-field:

$$\left(\vec{\epsilon}^{(1)} \cdot \vec{E} \right)_i = \sum_j \epsilon_{ij}^{(1)} E_j, \quad (2.7)$$

we can write the relation between the linear component of the displacement vector and the electric field as:

$$\vec{D}_n^{(1)}(\vec{r}) = \epsilon_0 \vec{\epsilon}^{(1)}(\omega_n) \cdot \vec{E}. \quad (2.8)$$

All frequency components of the field separately have to fulfill the wave equation, which we can then write as:

$$\vec{\nabla}^2 \vec{E}_n(\vec{r}) + \frac{\omega_n^2}{c^2} \vec{\epsilon}^{(1)}(\omega_n) \cdot \vec{E}_n(\vec{r}) = -\frac{\omega_n^2}{\epsilon_0 c^2} \vec{P}_n^{\text{NL}}(\vec{r}). \quad (2.9)$$

2.2 The coupled wave equation for sum-frequency generation

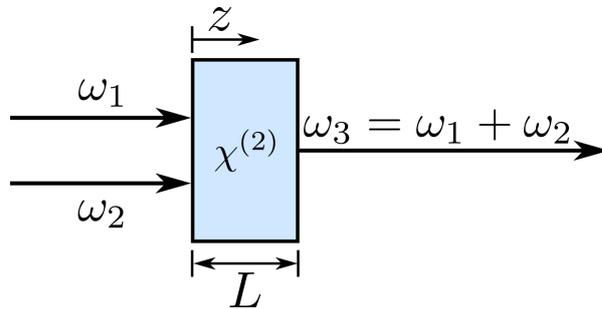


Figure 12: **Sum frequency generation (SFG)**. Two incident fields with frequencies ω_1 and ω_2 generate a field with frequency $\omega_3 = \omega_1 + \omega_2$.

We will now aim to use the wave equations above to describe non-linear optical processes in increasing detail. We will begin very simple: two fields with frequencies $\omega_{1,2}$ are incident on a $\chi^{(2)}$ nonlinear crystal, and they generate a field at the sum frequency $\omega_3 = \omega_1 + \omega_2$. We illustrate the situation in Fig. 12.

The wave equation 2.9 must hold for all frequencies involved. In the absence of any nonlinear interaction, the nonlinear source-term on the right-hand side of the wave equation will be zero, and the solution for ω_3 will simply be:

$$E_3(z, t) = A_3 e^{i(k_3 z - \omega_3 t)} + \text{c.c.} \quad (2.10)$$

We only wrote down the equation for a scalar field because we assume for now that the polarizations of the involved fields are well defined. The dispersion relation is:

$$k_3 = \frac{n_3 \omega_3}{c}, \quad n_3^2 = \epsilon^{(1)}(\omega_3). \quad (2.11)$$

As a next step, we will assume that the amplitude A_3 in equation 2.10 depends (slowly) on the position z along the crystal, and that we can write the polarization at ω_3 as:

$$\begin{aligned} P_3(z, t) &= P_3(z) e^{-i\omega_3 t} + \text{c.c.} \\ P_3(z) &= 4\epsilon_0 d_{\text{eff}} E_1(z) E_2(z). \end{aligned} \quad (2.12)$$

For the E fields we also separate the quickly oscillating parts from the slowly varying parts in space and time:

$$\begin{aligned} E_i(z, t) &= E_i(z) e^{-i\omega_i t} + \text{c.c.}, \\ E_i(z) &= A_i(z) e^{ik_i z} + \text{c.c.} \end{aligned} \quad (2.13)$$

In the above equations, we assumed that our medium is lossless and fulfills full permutation symmetry as discussed in chapter 1, and that we can

If we insert all that into wave equation 2.9, and if we take into account the dispersion relation $k_3^2 = \epsilon^{(1)}(\omega_3)\omega_3^2/c^2$, we get the following (see exercises):

$$\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} = -4 \frac{d_{\text{eff}} \omega_3^2}{c^2} A_1 A_2 e^{i(k_1 + k_2 - k_3)z}, \quad (2.14)$$

where the A_i are functions of z . If A_3 is varying slowly with z , we can neglect the first term in this equation, and the above equation becomes:

$$\frac{dA_3}{dz} = \frac{2id_{\text{eff}}\omega_3}{c} A_1 A_2 e^{i\Delta k z}, \quad (2.15)$$

with the wavevector mismatch $\Delta k = k_1 + k_2 - k_3$. This is called a coupled-amplitude

equation. We can derive similar equations for the other two amplitudes:

$$\begin{aligned}\frac{dA_1}{dz} &= \frac{2id_{\text{eff}}\omega_1}{c}A_3A_2^*e^{-i\Delta kz}, \\ \frac{dA_2}{dz} &= \frac{2id_{\text{eff}}\omega_2}{c}A_3A_1^*e^{-i\Delta kz}.\end{aligned}\quad (2.16)$$

2.2.1 General considerations about phase-matching

In equations 2.15 and 2.16 we introduced the wavevector mismatch Δk . Trying to achieve $\Delta k = 0$ is called *phase matching*. It essentially means that one makes sure that the contributions from all atomic dipoles in the material are in phase with each other and interfere constructively. If we set $\Delta k = 0$ in equation 2.15, and if we assume for now that A_1 and A_2 are constant, then we get a linear increase in A_3 . That $A_{1,2}$ are constant is of course impossible because it would violate energy conservation, but for small A_3 the depletion of power from $A_{1,2}$ can often be neglected.

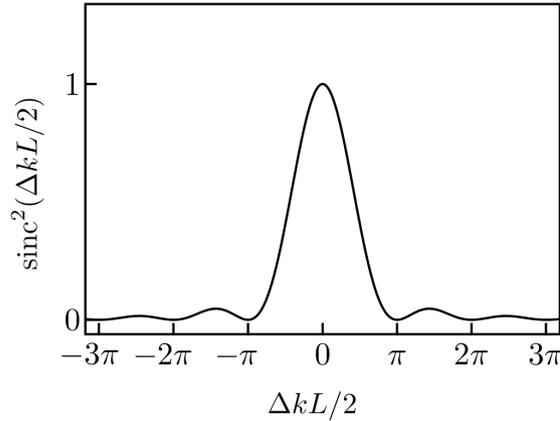


Figure 13: **Phase mismatch factor.** .

If $\Delta k \neq 0$ but we still assume no depletion, we can integrate equation 2.15, and we will get the following expression for A_3 at the exit face of the crystal:

$$A_3(L) = \frac{2id_{\text{eff}}\omega_3A_1A_2}{c} \int_0^L dz e^{i\Delta kz} \frac{2id_{\text{eff}}\omega_3A_1A_2}{c} \left(\frac{e^{i\Delta kL} - 1}{i\Delta k} \right), \quad (2.17)$$

where L is the length of the crystal.

The intensity of the SFG light will then be proportional to:

$$\left| \frac{e^{i\Delta k L} - 1}{\Delta k} \right|^2 = L^2 \text{sinc}^2(\Delta k L/2), \quad (2.18)$$

where $\text{sinc}(x) \equiv \sin(x)/x$. This is called the phase mismatch factor, and it contains the full dependence on Δk . Fig. 13 shows a plot of the phase mismatch factor.

The full expression for the intensity of the SFG field is:

$$I_3 = \frac{8d_{\text{eff}}^2 \omega_3^2 I_1 I_2}{n_1 n_2 n_3 \epsilon_0 c^2} L^2 \text{sinc}^2\left(\frac{\Delta k L}{2}\right), \quad (2.19)$$

where n_i are the refractive indices for the three fields, and we have defined the intensities of the three fields as:

$$I_i = 2n_i \epsilon_0 c |A_i|^2, \quad i = 1, 2, 3. \quad (2.20)$$

Fig. 13 shows that the phase mismatch factor is 1 for perfect phase matching and then vanishes it rapidly for increasing $|\Delta k L|$. However, there are some oscillations. These occur because if L is roughly greater than $1/\Delta k$, the output wave E_3 will get out of phase with the driving polarization depending on the two input fields. Then power from the E_3 field can flow back into the other frequency modes.

One can therefore define the length $L_{\text{coh}} = 2/\Delta k$ as the length of coherent buildup of the interaction. Then the phase mismatch factor can be written as:

$$\text{sinc}^2(L/L_{\text{coh}}). \quad (2.21)$$

2.3 Phase matching

We have seen in subsection 2.2.1 that the efficiency of nonlinear processes like SFG will drop drastically if the phasematching condition between the wavevectors of the participating fields is not fulfilled. For the case of SFG with $\omega_3 = \omega_1 + \omega_2$ that means if Δk is too different from zero with:

$$\Delta k = k_1 + k_2 - k_3. \quad (2.22)$$

In our treatment so far we did not take into account the depletion of the pump beams. That means, the case when so much power of the driving field is converted into the output field that the efficiency of the process will drop due to the lack of driving fields. But even in that case, an efficient conversion can only be realized if the phase matching condition is fulfilled. That means, if $\Delta k = 0$.

It is not straight forward to achieve perfect phase matching. For that reason, let us take a closer look at what phase matching entails. In the case of SFG, we can write

equation 2.22 as:

$$\frac{n_1\omega_1}{c} + \frac{n_2\omega_2}{c} = \frac{n_3\omega_3}{c}. \quad (2.23)$$

If the polarization of all three fields were the same, the refractive indices would only depend on the frequencies of the light, and we could write the phase matching condition as:

$$\frac{n(\omega_1)\omega_1}{c} + \frac{n(\omega_2)\omega_2}{c} = \frac{n(\omega_3)\omega_3}{c}. \quad (2.24)$$

The trouble is that in materials with normal dispersion, $n(\omega)$ increases monotonically with ω . To see that this is an issue, let us rewrite equation 2.22 as:

$$n_3 = \frac{n_1\omega_1 + n_2\omega_2}{\omega_3}. \quad (2.25)$$

Then we can write:

$$n_3 - n_2 = (n_1 - n_2) \frac{\omega_1}{\omega_3}. \quad (2.26)$$

Without loss of generality, we can assume $\omega_3 > \omega_2 \geq \omega_1$. In the case of normal dispersion, we would then have $n_3 > n_2$, but we also would have $n_2 > n_1$. That means while the left-hand side of equation 2.26 will be positive, the right-hand side will be negative. This shows that phase matching is not possible in a material with normal dispersion if all fields have the same polarization. This situation where all three fields have the same polarization is sometimes denoted as *type-0* phase matching.

To overcome this limitation, one can use birefringent crystals. In this case, one can distinguish between two cases: *type-I phase matching*, where the lower-frequency fields have the same polarization, and *type-II phase matching* where the two lower-frequency fields have polarizations that are orthogonal to each other. In order to achieve phase matching, we can use uniaxial or biaxial crystals. For simplicity, let us concentrate on uniaxial crystals. The crystal z axis and the wavevector \vec{k} define a plane. This axis is also often referred to as the crystal's c axis. Light that is polarized parallel to that plane is said to have *ordinary* polarization, and light that is polarized perpendicular to that plane is said to have *extraordinary* polarization. The refractive index for ordinary polarization is denoted as n_o . For extraordinary polarization, the refractive index $n_e(\theta)$ depends on the angle θ of \vec{k} relative to the z axis. It is defined by the relation:

$$\frac{1}{n_e(\theta)} = \frac{\sin^2 \theta}{\bar{n}_e^2} + \frac{\cos^2 \theta}{n_o^2}, \quad (2.27)$$

where $\bar{n}_e = n_e(\pi/2)$ is called the *principal value* of the extraordinary refractive index.

By adjusting the angle θ , one can now tune the extraordinary refractive index to try and achieve phase matching. One can illustrate the solution of the phasematching equation graphically as we illustrate in Fig. 14. Let us consider a simple example for second harmonic generation (SHG). In that case, we have $\omega_1 = \omega_2 = \omega$, and $\omega_3 = 2\omega$.

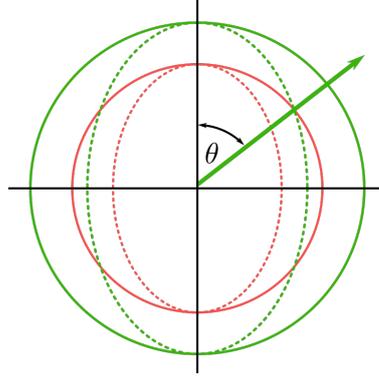


Figure 14: **Phase matching.** SHG in a negative uniaxial crystal. The ordinary refractive indices for the fundamental wavelength and for the SHG wavelength are indicated as solid circles in red and green, respectively. The extraordinary refractive indices for the two wavelengths are plotted as dashed ellipses. The phase-matching angle θ is given by the point where the extraordinary refractive index for the SHG light matches the ordinary refractive index of the fundamental wavelength. That is indicated by the green arrow.

One also distinguishes between positive uniaxial crystals for $n_e > n_o$ and negative uniaxial crystals for $n_o > n_e$. For our example, we choose a negative uniaxial crystal. In that case, the input field has to have ordinary polarization, and phase matching is fulfilled if:

$$n_e(2\omega, \theta) = n_o(\omega). \quad (2.28)$$

Using this and equation 2.27, we get:

$$\frac{\sin^2 \theta}{\bar{n}_e(2\omega)^2} + \frac{\cos^2 \theta}{n_o(2\omega)^2} = \frac{1}{n_o(\omega)^2}. \quad (2.29)$$

This equation does not necessarily have a solution. One will have to choose a crystal with n_o and n_e such that one can solve this equation to achieve phase matching.

There is a significant drawback of using a birefringent crystal and angle tuning to achieve phase matching. If the angle θ is not 0 or $\pi/2$, the Poynting vector \vec{S} and the wavevector \vec{k} will not point in the same direction. That means the input beams and the light generated will not travel in the same direction even though their wavevectors may point in the same direction. This leads to a *transverse walk-off* between these beams. This limits the interaction volume for the non-linear effect. In other words, one cannot use arbitrarily long crystals because at some point the walk-off becomes larger than the beams. This phenomenon is illustrated in Fig. 15. If one uses a pulsed laser for non-linear processes, there can in addition also be a *longitudinal walk-off* because the group

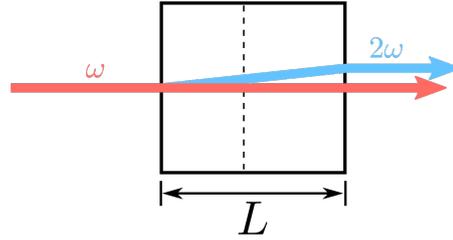


Figure 15: **Transverse walk-off in SHG.** If the incoming beam has ordinary polarization and the SHG beam has extraordinary polarization, the SHG beam will experience a transverse walk-off. After some distance indicated by the dashed line, there will essentially be no overlap between the two beams, and a longer crystal will not lead to increased SHG output.

velocities for the beams involved may differ.

2.4 Quasi phasematching

We already showed that it is not possible to achieve phasematching in a crystal with normal dispersion if all waves have the same polarization. Using angle tuning to achieve phase matching is only possible in birefringent materials, and even then the crystals have to be cut properly, and one has to make use of certain nonlinear coefficients, depending on the polarizations chosen. If one wants to achieve very high conversion efficiency, for example, it is however tempting to make use of the d_{rr} nonlinear coefficient because in many materials it is significantly larger than other nonlinear coefficients. However, the d_{33} coefficient is only relevant if all the interacting waves have the same polarization, and for that case normal phase matching cannot be achieved.

A solution to overcome this limitation is quasi phasematching (QPM). The idea is the following: earlier we introduced the L_{coh} as the length over which the phasematching is fulfilled to such a degree as to lead to a coherent buildup of the frequency-converted field. If one had the possibility to rotate the crystal by 180° after a distance comparable to L_{coh} , one could reestablish good phasematching before the interacting fields became out of phase. By repeatedly doing that in regular distances, one can in principle keep up the phasematching even in very long crystals.

This can be achieved in the following way: if the material used is ferroelectric, one can apply a periodically modulated field to the crystal during the fabrication process. This will effectively invert the sign of d_{eff} periodically. This is illustrated in Fig. 16. If we use $d(z)$ to denote the spatial dependence of the effective nonlinear coefficient along

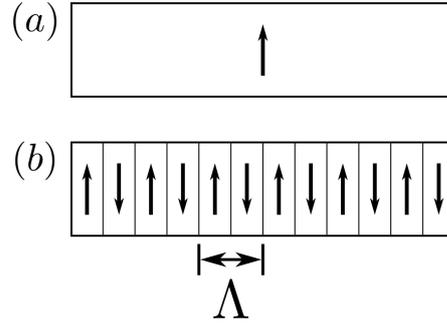


Figure 16: **Periodic poling.** (a) In a homogeneous nonlinear crystal, the crystal's c axis points in the same direction everywhere in the crystal. (b) In a periodically poled crystal, the orientation of the c axis is periodically flipped. The poling period is Λ .

the crystal, we can write $d(z)$ as a square wave:

$$d(z) = d_{\text{eff}} \text{sign} \left[\cos\left(z \frac{\Lambda}{2\pi}\right) \right]. \quad (2.30)$$

Sometimes a more complicated spatial dependence is chosen - for example, to achieve simultaneous phase matching for multiple processes. In order to describe the phase-matching process mathematically, we can replace d_{eff} in the coupled amplitude equations 2.15 and 2.16 by $d(z)$. To facilitate solving these equations, it is better to decompose $d(z)$ into a Fourier series:

$$d(z) = d_{\text{eff}} \sum_{m=-\infty}^{\infty} G_m \exp(iq_m z). \quad (2.31)$$

q_m are the grating wave vectors, and the Fourier coefficients fulfill:

$$G_m = \frac{2}{m\pi} \sin(m\pi/2). \quad (2.32)$$

One can then derive the corresponding coupled wave equations similar to our earlier derivation. In doing so, one assumes that the amplitudes A_i change slowly with z along the crystal, and that the phasematching process is dominated by one particular

component m in the decomposition in equation 2.31. The resulting equations are:

$$\begin{aligned}\frac{dA_1}{dz} &= \frac{2i\omega_1 d_Q}{n_1 c} A_3 A_2^* e^{-i(\Delta k_Q - 2q_m)z}, \\ \frac{dA_2}{dz} &= \frac{2i\omega_2 d_Q}{n_2 c} A_3 A_1^* e^{-i(\Delta k_Q - 2q_m)z}, \\ \frac{dA_3}{dz} &= \frac{2i\omega_3 d_Q}{n_3 c} A_1 A_2 e^{i\Delta k_Q z}.\end{aligned}\tag{2.33}$$

Here, we introduced the nonlinear coupling coefficient for the Fourier coefficient m :

$$d_Q = d_{\text{eff}} G_m.\tag{2.34}$$

and the wavevector mismatch for order m is given by:

$$\Delta k_Q = k_1 + k_2 - k_3 + q_m.\tag{2.35}$$

The main differences compared to our earlier set of coupled amplitude equations are (1) that we have d_Q instead of d_{eff} , and more importantly (2) the wavevector mismatch is modified.

Because d_Q typically decreases with increasing m , usually one tries to establish QPM using a first-order interaction ($m = -1$). In that case, we get:

$$\Delta k_Q = k_1 + k_2 - k_3 - \frac{2\pi}{\Lambda}, \quad d_Q = (2/\pi)d_{\text{eff}}.\tag{2.36}$$

By setting $\Delta k_Q = 0$, we can conclude that the optimal poling period Λ to achieve QPM is given by:

$$\Lambda = 2L_{\text{coh}} = 2\pi/(k_1 + k_2 - k_3).\tag{2.37}$$

2.5 Sum-frequency generation

We will use the mathematical formulations we derived earlier in this chapter in the next sections. Before we do that, let us first describe a simpler but instructive situation, which is called *frequency upconversion*. In particular, we will consider SFG in a situation where the pump field at frequency ω_2 is strong but the other pump field with ω_1 is weak. Using SFG, we can create a signal with $\omega_3 = \omega_1 + \omega_2$ where ω_2 is effectively a strong pump allowing us to convert the frequency of the input signal with ω_1 . For example, this can be useful if we want to detect a weak signal at ω_1 but we only have good detectors operating at ω_3 .

Let us describe this situation using our coupled amplitude equations 2.15 and 2.16. Because we assume that A_2 is strong, we can assume that it remains approximately

constant during the process. That simplifies our set of coupled equations to:

$$\frac{dA_1}{dz} = K_1 A_3 e^{-i\Delta k z}, \quad \frac{dA_3}{dz} = K_3 A_1 e^{i\Delta k z},$$

with the coupling quantities:

$$K_1 = \frac{2i\omega_1^2 d_{\text{eff}}}{k_1 c^2} A_2^*, \quad K_3 = \frac{2i\omega_3^2 d_{\text{eff}}}{k_3 c^2} A_2. \quad (2.38)$$

In the case of perfect phase matching ($\Delta k = 0$), this simplifies even further, and by eliminating A_3 , we get the ordinary differential equation:

$$\frac{d^2 A_1}{dz^2} = -\kappa^2 A_1. \quad (2.39)$$

Here, we introduced the (positive) coupling coefficient κ^2 as:

$$\kappa^2 \equiv -K_1 K_3 = \frac{4\omega_1^2 \omega_3^2 d_{\text{eff}}^2 |A_2|^2}{k_1 k_3 c^4}. \quad (2.40)$$

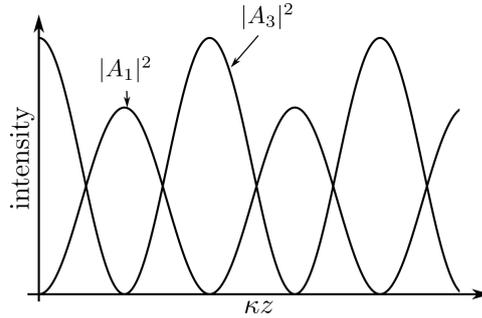


Figure 17: **Frequency conversion.** Using frequency conversion, we can convert the input field A_1 at ω_1 to the output field at ω_3 . The plot shows how the intensities of the two fields evolve along the crystal. Because we assume perfect phase matching, the plot shows perfect conversion efficiency.

The solutions of our coupled amplitude equations for this situation are:

$$\begin{aligned} A_1(z) &= B \cos(\kappa z) + C \sin(\kappa z), \\ A_r(z) &= -\frac{B\kappa}{K_1} \sin(\kappa z) + \frac{C\kappa}{K_1} \cos(\kappa z). \end{aligned} \quad (2.41)$$

To accurately describe the frequency conversion of a weak signal, we can choose the integration constants B, C such that the solutions fulfill the boundary conditions. In

particular, at the entry face of the crystal, the converted field should be zero - that means $A_3(0) = 0$. This implies that $C = 0$ and $B = A_1(0)$. Figure 17 shows how the intensities of the fields with ω_1 and ω_3 evolve along the crystal.

2.6 Second harmonic generation

In this case, we only have one input field with frequency ω_1 and an output field with frequency $\omega_2 = 2\omega_1$. The total electric field is given by:

$$\begin{aligned} E(z, t) &= E_1(z, t) + E_2(z, t) = e^{-i\omega_1 t} E_1(z) + e^{-i\omega_2 t} E_2(z) = \\ &= e^{-i\omega_1 t + ik_1 z} A_1(z) + e^{-i\omega_2 t + ik_2 z} A_2(z), \end{aligned} \quad (2.42)$$

where we assumed that we can focus on only the z component of the field, and we again assume that the A_j are slowly varying functions of z . The field E has to fulfill the wave equation in our nonlinear medium:

$$\frac{\partial^2 E_j(z, t)}{\partial z^2} - \frac{\epsilon(1)(\omega_j)}{c^2} \frac{\partial^2 E_j(z, t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 P_j(z, t)}{\partial t^2}, \quad (2.43)$$

where $P_j(z, t) = P_j(z)e^{-i\omega_j t} + \text{c.c.}$, and:

$$\begin{aligned} P_1(z) &= 4\epsilon_0 d_{\text{eff}} E_2 E_1^* = 4\epsilon_0 d_{\text{eff}} A_2 A_1^* e^{i(k_2 - k_1)z}, \\ P_2(z) &= 2\epsilon_0 d_{\text{eff}} E_1^2 = 2\epsilon_0 d_{\text{eff}} A_1^2 e^{2ik_1 z}. \end{aligned} \quad (2.44)$$

Note the different degeneracy factors 2 and 4 in the above equation.

We then arrive at the following coupled amplitude equations for SHG:

$$\begin{aligned} \frac{dA_1}{dz} &= \frac{2i\omega_1^2 d_{\text{eff}}}{k_1 c^2} A_2 A_1^* e^{-i\Delta k z}, \\ \frac{dA_2}{dz} &= \frac{i\omega_2^2 d_{\text{eff}}}{k_2 c^2} A_1^2 e^{i\Delta k z}, \end{aligned} \quad (2.45)$$

where $\Delta k = 2k_1 - k_2$.

To solve these equations, it is convenient to write the amplitudes as follows:

$$\begin{aligned} A_1 &= \left(\frac{I}{2n_1 \epsilon_0 c} \right)^{1/2} u_1 e^{i\phi_1}, \\ A_2 &= \left(\frac{I}{2n_2 \epsilon_0 c} \right)^{1/2} u_2 e^{i\phi_1}. \end{aligned} \quad (2.46)$$

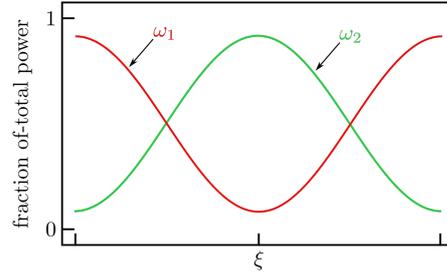


Figure 18: **Energy exchange.** Along the nonlinear crystal, power from the fundamental frequency ω_1 is converted to $\omega_2 = 2\omega_1$ over a typical length scale l . As ξ increases, the frequency conversion at some point saturates and then takes place from ω_2 to ω_1 instead.

We introduced the intensities $I = I_1 + I_2$ of the two fields:

$$I_j = 2n_j\epsilon_0c|A_j|^2. \quad (2.47)$$

$u_{1,2}$ are slowly varying functions of z . It is convenient to introduce a normalized distance parameter $\xi = z/l$, where

$$l = \left(\frac{2n_1^2n_2}{\epsilon_0cI} \right)^{1/2} \frac{c}{2\omega_1d_{\text{eff}}}. \quad (2.48)$$

This is the typical distance over which our two fields exchange energy. This exchange can be visualized as illustrated in Fig. 18.

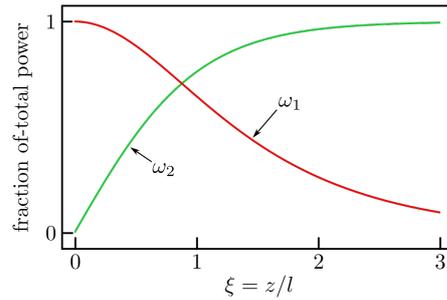


Figure 19: **Amplitudes of the fundamental field and the second harmonic field.** If $u_2 = 0$ in the beginning, then all the power will eventually be in u_1 for $\xi \rightarrow \infty$.

If one assumes the boundary conditions $u_1(0) = 1, u_2(0) = 0$, the solutions of these

coupled differential equations are:

$$\begin{aligned} u_1(\xi) &= \operatorname{sech}\xi = (\cosh \xi)^{-1}, \\ u_2(\xi) &= \tanh \xi. \end{aligned} \quad (2.49)$$

This case is shown in Fig. 19.

2.7 Difference frequency generation and parametric amplification

Consider the case illustrated in Fig. 20. One can simplify the analysis by assuming that the input field ω_3 is strong and that there is no significant depletion. In that case, the corresponding amplitude A_3 can be assumed to be roughly constant. By using a similar approach then as for SFG in section 2.2, one can derive coupled wave equations for the amplitudes $A_{1,2}$:

$$\begin{aligned} \frac{dA_1}{dz} &= \frac{2i\omega_1^2 d_{\text{eff}}}{k_1 c^2} A_3 A_2^* e^{i\Delta k z}, \\ \frac{dA_2}{dz} &= \frac{2i\omega_2^2 d_{\text{eff}}}{k_2 c^2} A_3 A_1^* e^{i\Delta k z}, \end{aligned} \quad (2.50)$$

where the wave vector mismatch is:

$$\Delta k = k_3 - k_1 - k_2. \quad (2.51)$$

In the case of perfect phase matching ($\Delta k = 0$), one can combine the above equations to yield:

$$\frac{d^2 A_2}{dz^2} = \frac{4\omega_1^2 \omega_2^2 d_{\text{eff}}^2}{k_1 k_2 c^4} A_3 A_3^* A_2 \equiv \kappa^2 A_2. \quad (2.52)$$

The real coupling constant κ is given by:

$$\kappa^2 = \frac{4d_{\text{eff}}^2 \omega_1^2 \omega_2^2}{k_1 k_2 c^4} |A_3|^2. \quad (2.53)$$

If one assumes the boundary condition $A_2(0) = 0$, the solution to that equation becomes:

$$\begin{aligned} A_1(z) &= A_1(0) \cosh(\kappa z) \\ A_2(z) &= i \left(\frac{n_1 \omega_2}{n_2 \omega_1} \right)^{1/2} \frac{A_3}{|A_3|} A_1^*(0) \sinh(\kappa z). \end{aligned} \quad (2.54)$$

There are two things we should note about these solutions: (1) one can see from

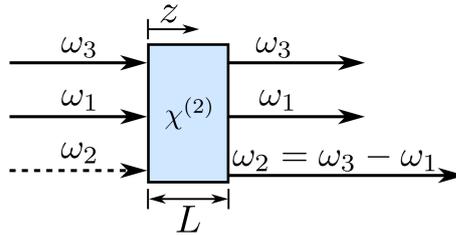


Figure 20: **Difference frequency generation (DFG)**. Typically there is no input with ω_2 but only inputs at ω_1 and ω_3 .

equations 2.54 that the solutions retain the phases of the initial fields, and (2) that both $A_1(z)$ as well as $A_2(z)$ increase monotonically. That means, there is no oscillation between the fields as in the case of SFG or SHG. One can intuitively understand the second point by considering the DFG process in terms of transitions between energy levels as illustrated in Fig. 21. After an excitation by a photon of frequency ω_3 to a virtual energy level, the decay back to the ground state can happen in two ways: (a) a field at ω_1 can stimulate the according transition, leading to the generation of an ω_2 photon. Or (b), the presence of a field at ω_2 can stimulate the transition that leads to the emission of an ω_1 photon.

The interesting thing about this is that, a higher intensity at one frequency will lead to a higher intensity at the other frequency. Because the generation of the ω_1 field is amplified by the ω_2 field, which is generated in the parametric process of DFG, this is known as *optical parametric amplification*¹. In this context, one says that the signal field at ω_1 is amplified by the nonlinear mixing process, and that the idler field at ω_2 is generated by this process.

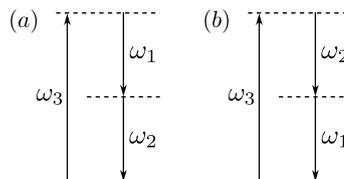


Figure 21: **Energy levels in DFG**. The decay from the excited state can occur in two ways shown in (a) and (b).

¹The difference between parametric and non-parametric processes is that the susceptibility in a parametric process is real and that the photon energy is conserved in the process[1].

2.8 Optical parametric oscillators (OPOs)

At the end of the last section, we introduced the concept of optical parametric amplification. If one places the nonlinear crystal used for parametric amplification inside a cavity resonant with ω_1 and/or ω_2 , one can enhance that amplification, and one does not require input beams at ω_1 or ω_2 but only at ω_3 . The resulting device is then known as an optical parametric oscillator (OPO). Fig. 22 illustrates the schematic layout of an OPO and the corresponding energy transitions. At some point, the assumption that the field at ω_3 is approximately constant will not be valid any longer. Due to energy conservation, the sum of the intensities of the output beams must, of course, be the same as the intensities of the input beams. BUT it is in principle possible to achieve very high efficiency in frequency conversion by “cavity-enhancing” nonlinear processes.

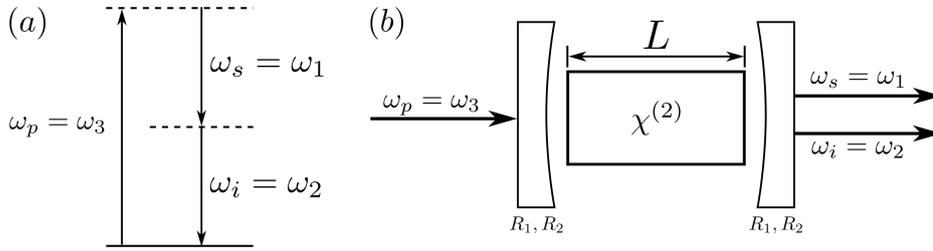


Figure 22: **Optical Parametric Oscillator.** (a) shows the energy levels as in DFG but with the notation of pump, signal and idler photons. (b) the nonlinear crystal inside a Fabry-Perot cavity with cavity mirrors that each have reflectivities $R_1 = |r_1|^2$ and $R_2 = |r_2|^2$ at the signal and idler wavelengths, respectively.

The coupled wave equations describing the evolution of the amplitudes along z in an OPO are the same as for DFG. That means, out coupled wave equations are identical to the ones in equation 2.50. The general solutions for these equations for $\Delta k = k_3 - k_1 - k_2$ are:

$$\begin{aligned} A_1(z) &= \left[A_1(0) \left(\cosh gz - \frac{i\Delta k}{2g} \sinh gz \right) + \frac{\kappa_1}{g} A_2^*(0) \sinh gz \right] e^{i\Delta kt/2}, \\ A_2(z) &= \left[A_1(0) \left(\cosh gz - \frac{i\Delta k}{2g} \sinh gz \right) + \frac{\kappa_2}{g} A_1^*(0) \sinh gz \right] e^{i\Delta kt/2}, \end{aligned} \quad (2.55)$$

with the gain g and the coupling constants κ_m given by:

$$g = [\kappa_1 \kappa_2^* - (\Delta k/2)^2]^{1/2} \quad \text{and} \quad \kappa_m = \frac{2i\omega_m^2 d_{\text{eff}} A_r}{k_m c^2}. \quad (2.56)$$

It is instructive to look at the special case, where we have perfect phase matching

($\Delta k = 0$) and the boundary condition $A_2(0) = 0$. Then the solutions become:

$$\begin{aligned} A_1(z) &= A_1(0) \cosh gz, \\ A_2(z) &= i \left(\frac{n_1 \omega_2}{n_2 \omega_1} \right) \frac{A_3}{|A_3|} A_1^*(0) \sinh gz. \end{aligned} \quad (2.57)$$

For large gz , both of these solutions asymptotically are proportional to $\exp(gz)$. That means that we have an exponential growth. Of course, that is only true as long as the pump field is not significantly depleted.

An OPO will only have a steady output at ω_1 and ω_2 , if the gain inside the cavity is larger than the losses. This defines the *threshold for optical parametric oscillation*. To be more precise, at the threshold the field amplitudes $A_{1,2}$ should not change in one round trip in the cavity. This condition on the two fields can be written as:

$$\begin{aligned} A_1(0) &= \left[A_1(0) \cosh gL + \frac{\kappa_1}{g} A_2^*(0) \sinh gL \right] (1 - l_1), \\ A_2^*(0) &= \left[A_2^*(0) \cosh gL + \frac{\kappa_2^*}{g} A_1(0) \sinh gL \right] (1 - l_2), \end{aligned} \quad (2.58)$$

where $l_m = 1 - R_m e^{-\alpha_m L}$ is the fractional amplitude loss per round trip, and α_m is the absorption coefficient for ω_m . Combining the equations above, one can conclude that:

$$\cosh gL = 1 + \frac{l_1 l_2}{2 - l_1 - l_2}. \quad (2.59)$$

3 Intensity-dependent refractive index

So far, we mostly concentrated on second-order nonlinear optical effects, which we described using the $\chi^{(2)}$ nonlinearity. It is a very common effect, though, that light passing through a material, especially if it has high intensity, can change the optical properties of materials. This is also known as the *optical Kerr effect*. A well-known example is the self-focusing of a beam. Here, we will discuss how to describe such effects, and then we will discuss some prominent examples and applications.

3.1 Describing the intensity-dependent refractive index

In many cases, the dependence of the refractive index of a material can be described via the relation

$$n = n_o + \bar{n}_2 \langle E^2 \rangle, \quad (3.1)$$

where n_o is the usual, unmodified refractive index, and \bar{n}_2 is sometimes called the second-order refractive index[1]. The bar on top of \bar{n}_2 is introduced to avoid confusing with other quantities we will introduce[1]. The angular brackets denote time average. If the electric field can be expressed as a plane wave

$$E(t) = E(\omega)e^{-i\omega t} + c.c., \quad (3.2)$$

then we can calculate the time average and get:

$$n = n_o + 2\bar{n}_2 |E(\omega)|^2. \quad (3.3)$$

We can express the effect as well in terms of the nonlinear polarization as we did earlier. Focusing only on the optical Kerr effect, the nonlinear polarization can be written as:

$$P^{(\text{NL})}(\omega) = 3\epsilon_0\chi^{(3)}(\omega = \omega + \omega - \omega)|E(\omega)|^2E(\omega). \quad (3.4)$$

Here, we assumed that all fields are linearly polarized, and we suppressed the tensor character of $\chi^{(3)}$.

The total polarization up to third order and assuming that there are no second-order effects, will then be given by:

$$P^{\text{TOT}}(\omega) = \epsilon_0\chi^{(1)}E(\omega) + 3\epsilon_0\chi^{(3)}|E(\omega)|^2E(\omega) \equiv \epsilon_0\chi_{\text{eff}}E(\omega) \quad (3.5)$$

with the effective susceptibility

$$\chi_{\text{eff}} = \epsilon_0 \chi^{(1)} + 3\epsilon_0 \chi^{(3)} |E(\omega)|^2. \quad (3.6)$$

If we then write $n^2 = 1 + \chi_{\text{eff}}$, we can conclude:

$$\begin{aligned} n_0 &= (1 + \chi^{(1)})^{1/2}, \\ \bar{n}_2 &= \frac{3\chi^{(3)}}{2n_0}. \end{aligned} \quad (3.7)$$

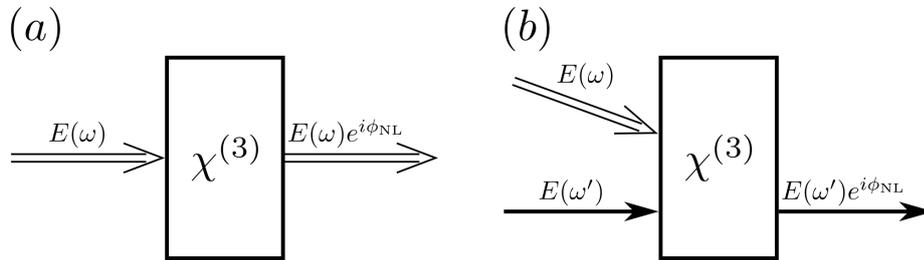


Figure 23: **Measuring the intensity-dependent refractive index.** (a) if the field $E(\omega)$ is sufficiently strong, it will modify the refractive index of a $\chi^{(3)}$ nonlinear medium and introduce a nonlinear phase shift ϕ_{NL} in the same field. (b) instead of observing changes to the strong field itself, one can measure the phase shift experienced by a non-collinear probe field $E(\omega')$ due to the change in refractive index caused by the strong field $E(\omega)$.

So far, we have assumed that we observe the changes to the refractive index due to the field $E(\omega)$ by its changes on the propagation of the same field. Another possibility is to use a weak probe field $E(\omega')$ to probe the changes to the refractive index caused by the stronger field $E(\omega)$. The nonlinear polarization at the frequency of the probe field can be written as:

$$P^{\text{NL}}(\omega') = 6\epsilon_0 \chi^{(3)}(\omega' = \omega' + \omega - \omega) |E(\omega)|^2 E(\omega'). \quad (3.8)$$

It is important to note that the degeneracy factor (6) in this case is larger than in the case where the same field is used as the probe. This is even true in the case where $\omega = \omega'$ because even then the two modes will be distinguishable because of their different directions of propagation (see Fig. 23). The refractive index encountered by the probe field will be:

$$n = n_0 + 2\bar{n}_2^{(\text{cross})} |E(\omega)|^2, \quad (3.9)$$

where:

$$\bar{n}_2^{(\text{cross})} = \frac{3\chi^{(3)}}{2n_0}, \quad (3.10)$$

which is twice as large as the \bar{n}_2 encountered in the case of Fig. 23(a).

One can also define the intensity-dependent refractive index as a function of the light intensity instead of the electric field. In this case, we can write:

$$n = n_0 + n_2 I, \quad (3.11)$$

where the intensity is given by:

$$I = 2n_0\epsilon_0 c |E(\omega)|^2, \quad (3.12)$$

and therefore the relation between \bar{n}_2 and n_2 will be:

$$n_2 = \frac{\bar{n}_2}{n_0\epsilon_0 c} = \frac{3}{4n_0^2\epsilon_0 c} \chi^{(3)}. \quad (3.13)$$

3.2 The third-order susceptibility tensor

In the preceding section, we described the intensity-dependent refractive index using the refractive index and the effective susceptibility. In a full treatment, $\chi_{ijkl}^{(3)}$ is of course a tensor with four indices. That means, it has 81 separate elements. In crystals without special symmetries, all or most of these elements can be independent, in principle. Many interesting third-order nonlinear effects occur even in isotropic media, though, like glass, air, liquids, or vapours. In such isotropic media, $\chi_{ijkl}^{(3)}$ has only three independent and non-zero elements.

One can see this as follows: because all coordinate axes must be equivalent, one can deduce that one must have:

$$\begin{aligned} \chi_{1111} &= \chi_{2222} = \chi_{3333}, \\ \chi_{1122} &= \chi_{1133} = \chi_{2211} = \chi_{2233} = \chi_{3311} = \chi_{3322}, \\ \chi_{1212} &= \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232}, \\ \chi_{1221} &= \chi_{1331} = \chi_{2112} = \chi_{2332} = \chi_{3113} = \chi_{3223}. \end{aligned} \quad (3.14)$$

Not all of these elements are independent. A rotation around an arbitrary axes must lead to the same physical results. It is convenient to choose an angle of 45 degrees for that purpose, and then one can show that one must have:

$$\chi_{1111} = \chi_{1122} + \chi_{1212} + \chi_{1221}. \quad (3.15)$$

Using these considerations, one sees that the nonlinear susceptibility has only three

non-zero and independent elements, and one can write:

$$\chi_{ijkl} = \chi_{1122}\delta_{ij}\delta_{kl} + \chi_{1212}\delta_{ik}\delta_{jl} + \chi_{1221}\delta_{il}\delta_{jk}. \quad (3.16)$$

In the case of third-harmonic generation, this simplifies even further. Similar to the case of inherent permutation symmetry in the case of SHG, one can conclude that one must have $\chi_{1122} = \chi_{1122} = \chi_{1221}$. Therefore, we are left with only one independent element, and we can write:

$$\chi_{ijkl}(3\omega = \omega + \omega + \omega) = \chi_{1122}(3\omega = \omega + \omega + \omega)(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}). \quad (3.17)$$

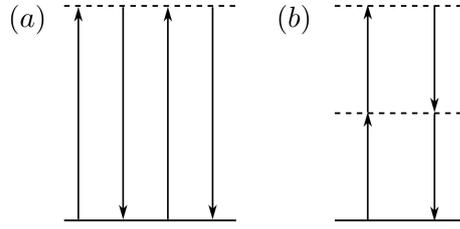


Figure 24: **Level diagrams for intensity-dependent refractive index.** Level representation of the two contributions to the nonlinear polarization for describing the intensity-dependent refractive index.

On the other hand, if we consider the nonlinear refractive index, the intrinsic permutation symmetry requires that $\chi_{1122} = \chi_{1212}$, and we can write:

$$\begin{aligned} \chi_{ijkl}(\omega = \omega + \omega - \omega) &= \chi_{1122}(\omega = \omega + \omega - \omega)(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + \\ &\chi_{1122}(\omega = \omega + \omega - \omega)\delta_{il}\delta_{jk}. \end{aligned} \quad (3.18)$$

Using this representation of the third-order susceptibility, the corresponding nonlinear polarization can be written as:

$$\begin{aligned} P_i(\omega) &= 3\epsilon_0 \sum_{jkl} \chi_{ijkl}(\omega = \omega + \omega - \omega) E_j(\omega) E_k(\omega) E_l(-\omega) \\ &= 6\epsilon_0 \chi_{1122} E_i(\vec{E} \cdot \vec{E}^*) + 3\epsilon_0 \chi_{1221} E_i^*(\vec{E} \cdot \vec{E}). \end{aligned} \quad (3.19)$$

By introducing the two coefficients A and B as

$$A = 6\chi_{1122} = 3\chi_{1122} + 3\chi_{1212} \text{ and} \quad B = 6\chi_{1221}, \quad (3.20)$$

one can write the nonlinear polarization as:

$$\vec{P} = \epsilon_0 A (\vec{E} \cdot \vec{E}^*) \vec{E} + \frac{1}{2} \epsilon_0 B (\vec{E} \cdot \vec{E}) \vec{E}^*. \quad (3.21)$$

Equation 3.21 consists of two distinct contributions associated with the A and B coefficients, respectively. An interesting thing to note is that one of these contributions behaves like \vec{E} and the other like \vec{E}^* . The “handedness” (right vs left) therefore remains the same for the A contribution but is opposite for the B contribution. Fig. 24 illustrates one-photon and two-photon resonant contributions to the nonlinear polarization. The one-photon process illustrated in Fig. 24(a) only contributes to coefficient A . Part (b) of the same figure can, in principle, contribute to A and B , but under some circumstances it only contributes to B . We will learn more about this later.

3.2.1 Propagation through an isotropic nonlinear medium

Following our short description of the two different contributions A and B to the nonlinear polarization, let us quickly discuss the propagation of light through such a nonlinear medium and how it affects the polarization of light. Given that the B contribution to the polarization in equation 3.21 has the opposite handedness than the incoming light, we can expect that there may be some phase shift between the different components of polarization in a beam transmitted through such a medium. In particular, we will see that only linearly polarized light and circularly polarized light will not experience a rotation upon travelling through such a medium.

Let us start by writing our electric field in the basis of circular polarization:

$$\vec{E} = E_+ \hat{\sigma}_+ + E_- \hat{\sigma}_-, \quad (3.22)$$

where the unit vectors for right-hand ($\hat{\sigma}_+$) and left-hand polarization ($\hat{\sigma}_-$) are given by:

$$\hat{\sigma}_\pm = \frac{\hat{x} \pm i\hat{y}}{\sqrt{2}}. \quad (3.23)$$

\hat{x} and \hat{y} denote the unit vectors along the x and y axis, respectively.

Using the definition in equation 3.22, we can write equation 3.21 as:

$$\vec{P}^{\text{NL}} = \epsilon_0 A (|E_+|^2 + |E_-|^2) \vec{E} + \epsilon_0 B (E_+ + E_-) \vec{E}^*. \quad (3.24)$$

If we decompose the polarization into a right-hand and a left-hand part as

$$\vec{P}^{\text{NL}} = P_+ \hat{\sigma}_+ + P_- \hat{\sigma}_-, \quad (3.25)$$

then we can write:

$$P_\pm \equiv \epsilon_0 \chi_\pm^{\text{NL}} E_{\text{pm}}. \quad (3.26)$$

Here, we introduced the effective nonlinear susceptibilities

$$\chi_{\pm}^{\text{NL}} = A|E_{\pm}|^2 + (A + B)|E_{\mp}|^2. \quad (3.27)$$

Let us now use these definitions in the wave equation that the electromagnetic field has to satisfy in the nonlinear medium:

$$\nabla^2 \vec{E}(z, t) = \frac{\epsilon^{(1)}}{c^2} \frac{\partial^2 \vec{E}(z, t)}{\partial t^2} + \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}^{\text{NL}}}{\partial t^2}. \quad (3.28)$$

Then one gets the following differential equations for the two components of the electric field polarization:

$$\nabla^2 E_{\pm}(z, t) = \frac{\epsilon_{\pm}^{\text{eff}}}{c^2} \frac{\partial^2 E_{\pm}(z, t)}{\partial t^2}, \quad (3.29)$$

where

$$\epsilon_{\pm}^{\text{eff}} = \epsilon^{(1)} + \chi_{\pm}^{\text{NL}}. \quad (3.30)$$

Equation 3.29 has plane-wave solutions travelling with phase velocities c/n^{\pm} with $n_{\pm}^2 = \epsilon_{\pm}^{\text{eff}}$. That means that plane waves with those polarizations experience different refractive indices:

$$\begin{aligned} n_{\pm}^2 &= n_0^2 + \chi_{\pm}^{\text{NL}} = n_0^2 \left(1 + \frac{1}{n_0^2} [A|E_{\pm}|^2 + (A + B)|E_{\mp}|^2] \right) \\ &\approx n_0 + \frac{1}{2n_0} [A|E_{\pm}|^2 + (A + B)|E_{\mp}|^2]. \end{aligned} \quad (3.31)$$

The + and – polarization components will therefore experience a relative phase shift due to a difference Δn in the corresponding refractive indices:

$$\Delta n = n_+ - n_- = \frac{B}{2n_0} (|E_-|^2 - |E_+|^2). \quad (3.32)$$

From this equation we can conclude the following: (a) linear polarization as well as circular polarization are unaffected (consider: why is that?) while light with elliptical polarization will experience a rotation of polarization, and (b) this effect only depends on the B coefficient. The latter meant that only two-photon processes illustrated in Fig. 24 contribute to this effect. The rotation angle for elliptical polarization is given by:

$$\theta = \frac{1}{2} \Delta n \frac{\omega}{c} z. \quad (3.33)$$

3.3 Self-focusing of light and other self-action effects

The change in the refractive index of a nonlinear material caused by high-intensity light can lead to self-focusing. In this case, the refractive index is higher in the regions of higher intensity, and therefore the intense light effectively generates a lens-like distribution of the refractive index in the material. At the same time, the diffraction of light will tend to lead to a growth in the beam diameter. When these two effects balance exactly, the beam is said to trap itself. This occurs at the critical power for self trapping P_{cr} [1]

$$P_{\text{cr}} = \frac{\pi(0.61)^2 \lambda_0^2}{8n_0 n_2}, \quad (3.34)$$

where λ_0 is the laser wavelength in vacuum. Self-focusing will only occur if the power P is greater than P_{cr} . If the power is much larger ($P \gg P_{\text{cr}}$), then imperfections in the laser front will lead to the beam splitting up into many filaments, each of them approximately carrying a power of P_{cr} . We will discuss all of these effects in more detail in the following.

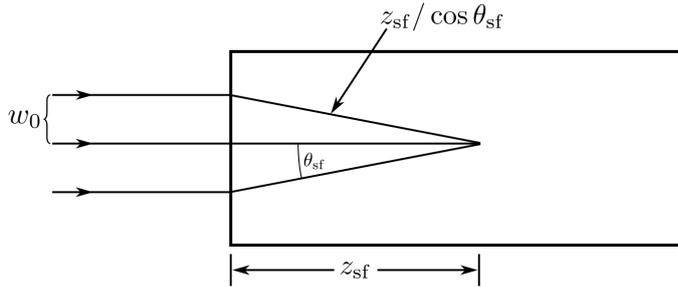


Figure 25: **Self-focusing.** Assuming ray optics propagation, we can predict the focal distance for self-focusing using Fermat's principle.

We will begin by considering the self-focusing of light following the description in Ref. [1], where they describe the self-focusing of a beam in the ray-optics regime - that means, we will neglect diffraction effects in this context. This approximation can be made if the beam diameter or the beam intensity is sufficiently large. We will include the effects of diffraction later on. A sketch accompanying our argument is provided in Fig. 25. Let us assume that the linear refractive index of the material is n_0 , and that the nonlinear refractive index along the central ray is $n_0 + n_2 I_0$, where I_0 is the intensity of the beam. According to Fermat's principle the travel time of light along the three lines shown in Fig. 25 needs to be identical. That means, we require:

$$(n_0 + n_2 I) z_{\text{sf}} = n_0 z_{\text{sf}} / \cos \theta_{\text{sf}}. \quad (3.35)$$

z_{sf} and θ_{sf} are defined in Fig. 25.

If we assume that $\cos \theta_{\text{sf}} \approx 1 - \theta_{\text{sf}}^2$, we can solve equation 3.35 to yield:

$$\theta_{\text{sf}} = \sqrt{2n_2 I / n_0}. \quad (3.36)$$

This “self-focusing angle” is typically a small quantity, which motivates the approximation above, and this is the typical angle by which a beam is deflected due to self-focusing. The self-focusing distance then is given by (for $P \gg P_{\text{cr}}$):

$$z_{\text{sf}} = w_0 \sqrt{\frac{n_0}{2n_2 I}} = \frac{2n_0 w_0^2}{\lambda_0} \frac{1}{\sqrt{P/P_{\text{cr}}}}. \quad (3.37)$$

Above, we wrote that this is valid for $P \gg P_{\text{cr}}$ because in this case the self-focusing is much stronger than the diffraction. If one takes diffraction into account, then the angular spread resulting from diffraction will counter-act the self-focusing such that the overall focusing angle becomes approximately $\theta = (\theta_{\text{sf}}^2 - \theta_{\text{diff}}^2)^{1/2}$, where

$$\theta_{\text{diff}} = \frac{0.61\lambda_0}{n_0 d} \quad (3.38)$$

is the diffraction angle for a beam with diameter d . Then the focusing distance becomes[1]:

$$z_{\text{sf}} = \frac{2n_0 w_0^2}{\lambda_0} \frac{1}{\sqrt{P/P_{\text{cr}} - 1}}. \quad (3.39)$$

For arbitrary powers and arbitrary beam-waist position, the self-focusing distance can be shown to be [1]:

$$z_{\text{sf}} = \frac{k w^2 / 2}{(P/P_{\text{cr}} - 1)^{1/2} + 2z_{\text{min}} / k w_0^2}, \quad (3.40)$$

where $k = n - 0\omega/c$, w is the waist of the beam when it enters the crystal, and z_{min} is the distance from the crystal face to the beam waist if there was no medium.

3.3.1 Self-trapping of light

Roughly speaking, self-trapping of light occurs if we have

$$\theta_{\text{diff}} = \theta_{\text{sf}}. \quad (3.41)$$

That means, if the diffraction angle is equal to the self-focusing angle. In this case, the spreading of the beam resulting from diffraction is balanced by the self-focusing of the light due to its intensity modifying the refractive index of the material.

If we insert the expressions for these two angles from equations 3.36 and 3.38, respec-

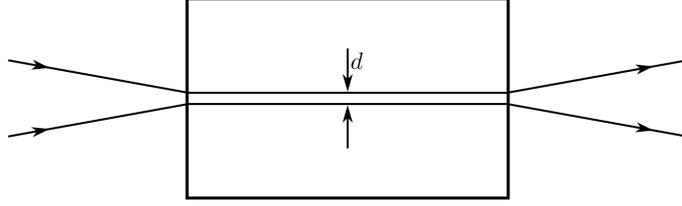


Figure 26: **Self-trapping.** If the incoming beam has the critical power P_{cr} , the light will trap itself transversally and travel along the crystal with some constant diameter d .

tively, into equation 3.41, we can calculate the intensity required for self trapping:

$$I = \frac{(0.61)^2 \lambda_0^2}{2n_2 n_0 d^2}, \quad (3.42)$$

where d is the diameter of the self-trapped beam as we illustrate it in Fig. 26. If we express the intensity in terms of the beam's power, we find that self-trapping occurs only if the power is equal to the critical power we defined earlier without proof:

$$P_{\text{cr}} = \frac{\pi(0.61)^2 \lambda_0^2}{8n_0 n_2} \approx \frac{\lambda_0^2}{8n_0 n_2}. \quad (3.43)$$

An interesting thing to note about equation 3.43 is that the beam diameter d dropped out. The effect of self-trapping only depends on the power but not on the intensity. The beam diameter will then simply depend on how we focus that power into the material.

A possible alternative way to understand this effect was illustrated by Chiao et al. [4, 1]. The argument is based on the simplifying assumption that the laser beam has a flat-top intensity distribution that will lead to a modified refractive index $n_0 + \delta n$ inside the medium. n_0 is the normal refractive index of the medium. This will effectively create a waveguide inside which light will be trapped if it fulfills the condition required for total internal reflection. This situation is illustrated in Fig. 27. In this picture, self-trapping occurs if the angle θ is less than the critical angle θ_{tot} for total internal reflection. This critical angle is given by:

$$\cos \theta_{\text{tot}} = \frac{n_0}{n_0 + \delta n}. \quad (3.44)$$

Because δn will typically be much smaller than unity, the angle $\cos \theta_{\text{tot}}$ will be small, and we can do a Taylor expansion of the cosine up to second order. Then we get:

$$\theta_{\text{tot}} = \left(\frac{2\delta n}{n_0} \right)^{1/2}. \quad (3.45)$$

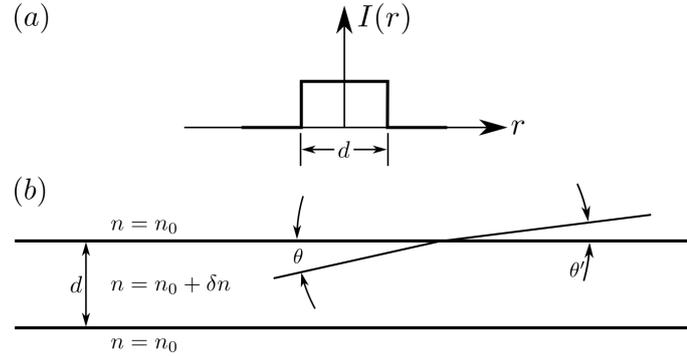


Figure 27: **Self-trapping of a beam with flat-topped intensity.** (a) the flat-top radial intensity distribution. (b) a ray of light originating inside the higher-index region is diffracted into the low-index region at an angle $\theta' < \theta$. Total reflection can trap the ray inside the waveguide.

If we have a beam of light with diameter d , the rays inside that beam will have an angular distribution with angles of at most θ_{diff} given in equation 3.38. Setting this equal to θ_{tot} , we can conclude that self-trapping will occur if the deviation of the refractive index fulfills:

$$\delta n = \frac{1}{2} n_0 \left(\frac{0.61 \lambda_0}{d n_0} \right)^2. \quad (3.46)$$

On the other hand, if δn is given, then this becomes a condition on the beam diameter:

$$d = \frac{0.61 \lambda_0}{\sqrt{2 n_0 \delta n}}. \quad (3.47)$$

If we now replace δn with $n_2 I$, then we can use this in combination with equation 3.47 to get the corresponding beam power:

$$P_{\text{cr}} = \frac{\pi}{4} d^2 I = \frac{\pi (0.61)^2 \lambda_0^2}{8 n_0 n_2}, \quad (3.48)$$

which is exactly the same result as we got earlier.

Mathematically describing self-action effects

To describe the effects of self-focusing and self-trapping more rigorously, let us look at the paraxial wave equation. This is appropriate because the angles resulting from self-focusing or self-trapping will be very small, and therefore the transverse components of the wavevectors will be much smaller than the longitudinal component along the beam

axis z . The paraxial wave equation for the slowly varying amplitude of our beam will be:

$$2ik_0 \frac{\partial A}{\partial z} + \nabla_T^2 A = -\frac{\omega^2}{\epsilon_0 c^2} p_{\text{NL}}. \quad (3.49)$$

For a purely third-order nonlinear resonance, the amplitude of the nonlinear polarization will be [1]:

$$p_{\text{NL}} = 3\epsilon_0 \chi^{(3)} |A|^2 A. \quad (3.50)$$

This equation can be used to find steady-state solutions for self trapping - that means, this applies for continuous-wave laser beams, not for pulsed laser beams.

Let us consider that wave equation for a case where we only have one transverse dimension. That means, the light is propagating, e.g., in a planar waveguide. Then equation 3.49 simplifies to:

$$2ik_0 \frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3\chi^{(3)} \frac{\omega^2}{c^2} |A|^2 A. \quad (3.51)$$

Here, A is a function only of x and z .

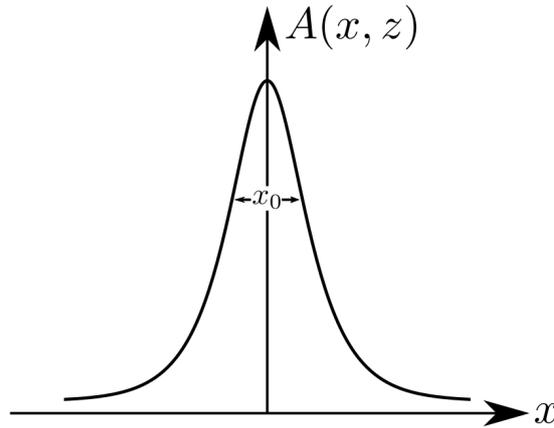


Figure 28: **Spatial soliton.** If there is only one transverse dimension x , then one solution of the paraxial wave equation is a spatial soliton as plotted here for an arbitrary amplitude, and for an arbitrary position along z . The width of the soliton is determined by x_0 .

A specific solution of equation 3.51 is:

$$A(x, z) = A_0 \text{sech}(x/x_0) e^{i\gamma z}, \quad (3.52)$$

where $\text{sech}(x) = (\cosh x)^{-1}$. The width of this peaked function is determined by x_0 ,

which is given by the relation:

$$x_0 = \frac{1}{k_0} \left(\frac{n_0}{2\bar{n}_2|A_0|^2} \right)^{1/2}, \quad (3.53)$$

where $\bar{n}_2 = 3\chi^{(2)}/4n_0$ is the same as in equation 3.7.

The solution given in equation 3.52 is sometimes called a “transverse soliton”. Fig. 28 shows the shape of this wavepacket. It can travel long distances without broadening. While the width will stay nearly constant, equation 3.52 shows that the wavepacket will acquire a phase during its propagation through the nonlinear medium. The rate of phase acquisition is given by:

$$\gamma = \frac{k_0\bar{n}_2|A_0|^2}{n_0}. \quad (3.54)$$

In higher dimensions, finding such solutions is not straight forward. They are often unstable. For a cylindrically symmetric beam, a solution was found by Chiao et al.[4].

3.3.2 Laser beam breakup

When a beam travels through a medium, impurities in that medium can cause distortions to the beam’s wavefront. If the intensity of the beam is much larger than P_{cr} , then the distorted parts of the wavefront can still have a larger power than P_{cr} and then that part of the beam will self-trap. For that reason, impurities in a nonlinear crystal can lead to the breaking up of a high-power optical beam into clearly separate parts that will then be self-trapped. Effectively, this looks as if the total beam would break apart into separate filaments. We illustrate this phenomenon in Fig. 29.

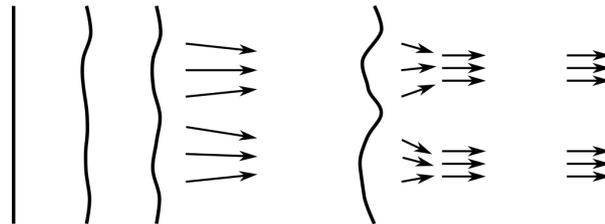


Figure 29: **Beam break-up.** Impurities in the crystal can distort the wavefront. If parts of that distorted wavefront still have sufficient power, these parts can self trap into separate filaments.

3.4 Optical phase conjugation

Aberration is an effect where different parts of an electromagnetic wavefront experience different delays as they pass through a medium or an optical system. The effects of aberration can be overcome by using optical phase conjugation. The principle behind phase conjugation is illustrated in Fig. 30. Part (a) of that figure shows a distorted wavefront reflected by a normal mirror. Parts of the wavefront that are preceding other parts will still remain foremost after the reflection. In this sense, a normal mirror mirrors the wavefront. The goal of a phase-conjugated mirror (PCM) is to reflect the wavefront such that parts that were lagging before the reflection will still be lagging behind the other parts of the wavefront after the reflection. This is illustrated in part (b) of Fig. 30.

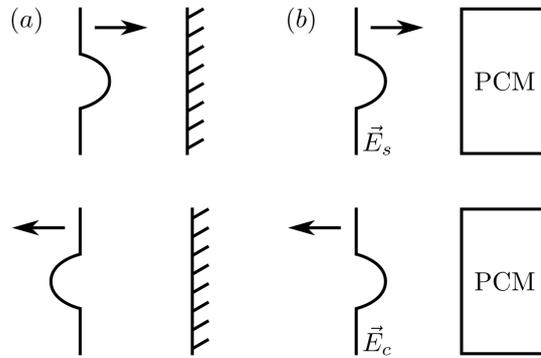


Figure 30: **Phase-conjugated mirrors (PCMs)**. (a) shows the reflection of a distorted wavefront from a normal mirror. (b) shows the reflection of the same wavefront from a PCM. In the latter case, parts of the wavefront that were lagging behind still do so after the reflection from the PCM. In (a), this is not the case.

Let us assume that the incoming field can be described by the vector:

$$\vec{E}_s(\vec{r}, t) = \vec{E}_s(\vec{r})e^{-i\omega t} + c.c.. \quad (3.55)$$

After the reflection from the PCM, the field should then be:

$$\vec{E}_c(\vec{r}, t) = r\vec{E}_s^*(\vec{r})e^{-i\omega t} + c.c., \quad (3.56)$$

where r is the reflection coefficient of the PCM.

To better understand the meaning of taking the conjugate of \vec{E}_s in equation 3.56, let us split the field into its amplitude, which is only slowly varying in space, the field's

polarization, and a part that is quickly oscillating in space:

$$\vec{E}_s(\vec{r}, t) = \hat{\epsilon}_s A_s(\vec{r}) e^{i\vec{k}_s \cdot \vec{r}}. \quad (3.57)$$

Here, $\hat{\epsilon}_s$ denotes the polarization unit vector, and \vec{k}_s is the wave vector of the incoming light.

Taking the complex conjugate gives:

$$\vec{E}_s^*(\vec{r}, t) = \hat{\epsilon}_s^* A_s^*(\vec{r}) e^{-i\vec{k}_s \cdot \vec{r}}. \quad (3.58)$$

From that we can conclude that the action of an ideal PCM implies the following:

- the polarization unit vector is replaced by its conjugate. That means, circularly polarized light will remain its handedness upon reflection: right-hand circular polarized light will remain right-hand polarized etc. Upon reflection from a metallic mirror, the handedness is reversed.
- $A_s(\vec{r})$ is replaced by its conjugate. This implies that the wavefront is reversed as illustrated in Fig. 30.
- \vec{k}_s is replaced with $-\vec{k}_s$. That means the wave is reflected back. In terms of ray optics, this means that each incident ray is reflected back exactly the way it came. In other words, the reflected angle is the inverse of the angle of incidence, not the same as would be in a normal mirror.

From equation 3.56, we can conclude that, apart from the reflection coefficient, the action of a PCM is essentially a time reversal because:

$$\vec{E}_c(\vec{r}, t) = \vec{E}_s(\vec{r}, -t). \quad (3.59)$$

This may already make clear, that it is no simple feat to realize a perfect PCM. Typically, real elements that are referred to as PCMs do not fulfill some of the characteristics of a perfect PCM. For example, they might not act on polarization as a perfect PCM would.

3.4.1 Aberration correction using phase conjugation

Assume that we have a plane wave travelling through a possibly inhomogeneous optical medium. This will introduce varying delays to different parts of the wavefront. A simple argument why the reflection at a PCM can correct this aberration is the following: the PCM will time-reverse the incoming wavefront, and then it will travel backwards through the same optical medium, experiencing the same inhomogeneities but in the opposite direction. The wave exiting the medium will then look exactly like the original incoming wave. We illustrate this in Fig. 31.

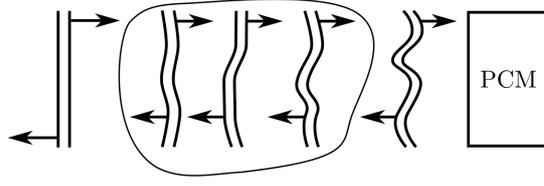


Figure 31: **Aberration correction by a PCM.** An incoming plane wavefront is distorted by its propagation through an inhomogeneous medium. After a PCM reflects the wavefront, it travels backwards through the same medium, resulting in the correction of the aberration it experienced.

In more formal terms, let us assume that the medium the incoming wave passes through has the non-homogeneous refractive index $n(\vec{r}) = \epsilon(\vec{r})^{1/2}$, and that our field satisfies the scalar waveequation:

$$\nabla^2 E - \frac{\epsilon(\vec{r})}{c^2} \frac{\partial^2 E}{\partial t^2} = 0. \quad (3.60)$$

Let us write our incoming field as:

$$E(\vec{r}, t) = A(\vec{r})e^{i(kz - \omega t)} + \text{c.c.} \quad (3.61)$$

The field amplitude $A(\vec{r})$ is assumed to be slowly varying in space. If our field is travelling in the z direction, and if the transverse components of the wavevector are sufficiently small, it is convenient to use the transverse form of the paraxial form of the wave equation. In particular, let us first split the Laplacian into its longitudinal and transverse parts, and then let us neglect the second derivative in z because we assume that the amplitude of the field only varies slowly. Then the wave equation becomes:

$$\nabla_T^2 A + \left[\frac{\omega^2 \epsilon(\vec{r})}{c^2} - k^2 \right] A + 2ik \frac{\partial A}{\partial z} = 0. \quad (3.62)$$

If this equation holds, so must its complex conjugate. That means, we must have:

$$\nabla_T^2 A^* + \left[\frac{\omega^2 \epsilon(\vec{r})}{c^2} - k^2 \right] A^* - 2ik \frac{\partial A^*}{\partial z} = 0, \quad (3.63)$$

where we assumed that $\epsilon(\vec{r})$ is real (no loss).

A solution to this equation is the wave: Let us write our incoming field as:

$$E_c(\vec{r}, t) = A^*(\vec{r})e^{i(-kz - \omega t)} + \text{c.c.} \quad (3.64)$$

Given that this field agrees with the incoming wave at any one time, this boundary condition will ensure that the above wave is the complex conjugate of the incoming wave *everywhere*. The boundary condition could for example be that the two solutions agree on the surface of the PCM.

3.4.2 Phase conjugation by degenerate four-wave mixing

While it is difficult to realize a perfect phase-conjugated mirror (PCM), people found ways to realize devices that implement some of its features if not all already quite some time ago. An example for that is phase conjugation via degenerate four-wave mixing. We will first describe the rough idea of how the process works before looking at it in a bit more detail. Fig. 32 illustrates the basic layout of realizing a PCM using degenerate four-wave mixing (DFWM) in a $\chi^{(3)}$ nonlinear medium. This approach has already been suggested by Hellwarth et al and by Yariv et al in 1977 (CITATIONS). This approach was demonstrated experimentally by Bloom and Bjorklund in the same year (CITATION).

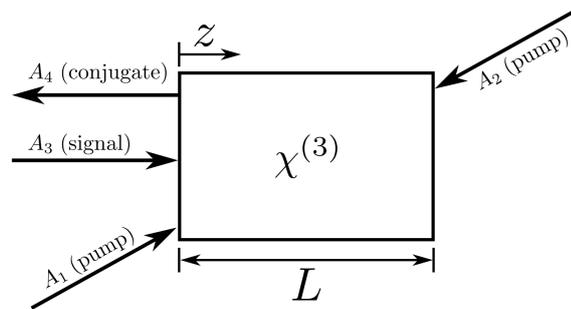


Figure 32: **Realizing a PCM via four-wave mixing.** A crystal is pumped by two fields $A_{1,2}$, which propagate at exactly opposite directions. The input field (signal) is the field A_3 . If the geometry is chosen properly, the output field A_4 will be the conjugate of the input field. The figure is based on Ref. [1].

Let us first discuss the working principle of this process. We will write the four fields as we are used to:

$$\begin{aligned} E_u(\vec{r}, t) &= E_i(\vec{r})e^{-i\omega t} + \text{c.c.} \\ &= A_i(\vec{r})e^{i(\vec{k}_i \cdot \vec{r} - \omega t)} + \text{c.c.}, \end{aligned} \quad (3.65)$$

where $i = 1, 2, 3, 4$, and the A_i are varying only slowly. We assume scalar quantities because the geometry will be fixed for the purpose of DFWM. While the nonlinear polarization will have many terms, in principle, we will focus on the following:

$$P^{\text{NL}} = 6\epsilon_0\chi^{(3)}E_1E_2E_3^* = 6\epsilon_0\chi^{(3)}A_1A_2A_3^*e^{i(\vec{k}_1 + \vec{k}_2 - \vec{k}_3) \cdot \vec{r}}. \quad (3.66)$$

This already assumes that the four frequencies match, that means that they are all $\omega \equiv \omega_i$, and $\chi^{(3)} = \chi^{(3)}(\omega = \omega + \omega - \omega)$. Then the time-dependent part in the exponent will vanish.

If we assume that the fields E_1 and E_2 are counter-propagating, we will have:

$$\vec{k}_1 + \vec{k}_2 = 0, \quad (3.67)$$

and then the term above becomes:

$$P^{\text{NL}} = 6\epsilon_0\chi^{(3)}A_1A_2A_3^*e^{-i\vec{k}_3\cdot\vec{r}}. \quad (3.68)$$

This shows that the nonlinear polarization contains a term that can act as a phase-matched source for the output field E_4 that will have the conjugate wavevector of the input field:

$$\vec{k}_4 = -\vec{k}_3. \quad (3.69)$$

At the same time, the amplitude of the output field will be proportional to $A_1A_2A_3^*$. If A_1 and A_2 are constant in space (plane waves), or if $A_1 = A_2^*$, then the product A_1A_2 will be proportional to the real quantity $|A_1|^2$.

Another way of looking at this process is that the interference of A_3 with one of the pump fields (e.g. A_1) will create an interference pattern inside the nonlinear crystal. This then will create a 3D grating of varying refractive index inside the material. The second pump field will then diffract from that grating to create the output wave A_4 .

Let us now look at the process in more detail. The total electric field will be the sum of the four field E_i , and this field will generate the third-order nonlinear polarization:

$$P = 3\epsilon_0\chi^{(3)}E^2E^*. \quad (3.70)$$

This will have 4^3 different terms, but the ones that are most interesting to us are the ones with the spatial dependence

$$e^{i\vec{k}_i\cdot\vec{r}} \quad (3.71)$$

for $i = 1, 2, 3, 4$. These terms will act as phase-matched source terms for the output field. The corresponding polarization amplitudes for these phase-matched contributions are[1]:

$$\begin{aligned} P_1 &= 3\epsilon_0 [E_1^2E_1^* + 2E_1E_2E_2^* + 2E_1E_3E_3^* + 2E_1E_4E_4^* + 2E_3E_4E_2^*], \\ P_2 &= 3\epsilon_0 [E_2^2E_2^* + 2E_2E_1E_1^* + 2E_2E_3E_3^* + 2E_2E_4E_4^* + 2E_3E_4E_1^*], \\ P_3 &= 3\epsilon_0 [E_3^2E_3^* + 2E_3E_1E_1^* + 2E_3E_2E_2^* + 2E_3E_4E_4^* + 2E_1E_2E_4^*], \\ P_4 &= 3\epsilon_0 [E_4^2E_4^* + 2E_4E_1E_1^* + 2E_4E_2E_2^* + 2E_4E_3E_3^* + 2E_1E_2E_3^*]. \end{aligned} \quad (3.72)$$

To simplify our treatment, we will assume that the pump fields $E_{1,2}$ are much stronger

than the signal and output fields $E_{3,4}$. Using this assumption, we can neglect all terms in equation 3.72 that depend on more than one of these weak fields, and we get:

$$\begin{aligned} P_1 &= 3\epsilon_0 [E_1^2 E_1^* + 2E_1 E_2 E_2^*], \\ P_2 &= 3\epsilon_0 [E_2^2 E_2^* + 2E_2 E_1 E_1^*], \\ P_3 &= 3\epsilon_0 [2E_3 E_1 E_1^* + 2E_3 E_2 E_2^* + 2E_1 E_2 E_4^*], \\ P_4 &= 3\epsilon_0 [2E_4 E_1 E_1^* + 2E_4 E_2 E_2^* + 2E_1 E_2 E_3^*]. \end{aligned} \quad (3.73)$$

Because the corresponding equations are much simpler, we can first concentrate on how the propagation through the medium affects $E_{1,2}$, and once we have solutions for that, we can enter them in the remaining equations to analyze the evolution of the weaker fields $E_{3,4}$. Each of the fields has to fulfill the wave equation:

$$\nabla^2 E_i - \frac{\epsilon}{c^2} \frac{\partial^2 E_i}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 P_i}{\partial t^2}. \quad (3.74)$$

If we introduce a new spatial coordinate z' along the direction of \vec{k}_1 , and that the field amplitudes vary only slowly, and if we also assume that the pump fields $E_{1,2}$ are plane waves, then the amplitude A_1 must fulfill the equation:

$$\begin{aligned} &\left[\left(-k_1^2 + 2ik_1 \frac{d}{dz'} + \frac{\epsilon\omega^2}{c^2} \right) A_1 \right] e^{i(k_1 z' - \omega t)} \\ &= -\frac{\omega^2}{c^2} 3\chi^{(3)} [|A_1|^2 + 2|A_2|^2] A_1 e^{i(k_1 z' - \omega t)} \end{aligned} \quad (3.75)$$

Doing the equivalent thing for A_2 , one arrives at two first-order ordinary differential equations for A_1 and A_2 . The solutions are:

$$A_1(z') = A_1(0)e^{i\kappa z'}, \quad A_2(z') = A_2(0)e^{i\kappa z'}. \quad (3.76)$$

This should not be terribly surprising since we assumed that the two pump fields are plane waves. The quantities $\kappa_{1,2}$ are coupling constants. If the two pump fields have equal intensities, we will have $\kappa_1 = \kappa_2$, and the product of the two pump amplitudes will then also be constant:

$$A_1(z')A_2(z') = A_1(0)A_2(0). \quad (3.77)$$

If we enter our solutions for $A_{1,2}$ into our wave equation 3.74, we arrive at the following

coupled differential equations for $A_{3,4}$:

$$\begin{aligned}\frac{dA_3}{dz} &= i\kappa_3 A_3 + i\kappa A_4^*, \\ \frac{dA_4}{dz} &= -i\kappa_3 A_4 - i\kappa A_3^*,\end{aligned}\quad (3.78)$$

where we introduced the coupling coefficients:

$$\begin{aligned}\kappa_3 &= \frac{3\omega}{nc} \chi^{(3)} (|A_1|^2 + |A_2|^2), \\ \kappa &= \frac{3\omega}{nc} \chi^{(3)} A_1 A_2.\end{aligned}\quad (3.79)$$

We can simplify these equations by introducing new functions $A'_{3,4}$ defined via the relations:

$$\begin{aligned}A_3 &= A'_3 e^{i\kappa_3 z}, \\ A_4 &= A'_4 e^{-i\kappa_3 z}.\end{aligned}\quad (3.80)$$

Then equations 3.78 become:

$$\begin{aligned}\frac{dA'_3}{dz} &= i\kappa A'_4, \\ \frac{dA'_4}{dz} &= -i\kappa A'_3.\end{aligned}\quad (3.81)$$

From the last equation, we can see why DFWM leads to phase conjugation. The field amplitude A'_4 is only driven by the input field amplitude.

One can combine the two equations 3.81 to find solutions for the field amplitudes $A'_{3,4}$. If we assume that we know the values of $A'_3(0)$ and $A'_4(L)$, then the solutions will be:

$$\begin{aligned}A'_3(z) &= -\frac{i|\kappa|}{\kappa} \frac{\sin |\kappa| z}{\cos |\kappa| L} A'_4(L) + \frac{\cos[|\kappa|(z-L)]}{\cos |\kappa| L} A'_3(L) \\ A'_4(z) &= \frac{\cos |\kappa| z}{\cos |\kappa| L} A'_4(L) - \frac{i\kappa}{|\kappa|} \frac{\sin[|\kappa|(z-L)]}{\cos |\kappa| L} A'_3(L)\end{aligned}\quad (3.82)$$

If we want to achieve optical phase conjugation, we usually will not insert a field A_4 at the end face of the crystal. That means, we have $A'(L) = 0$, and what we are mostly

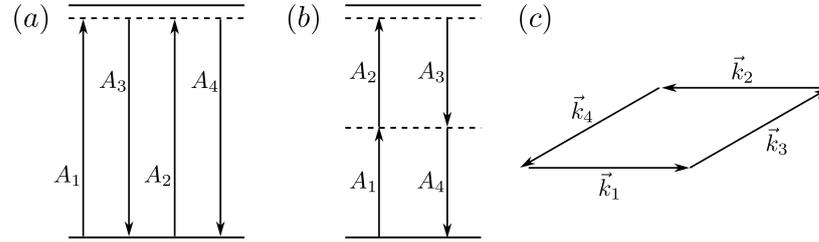


Figure 33: **Level diagrams and phase matching for DFWM.** (a) and (b) are the level diagrams for two possible interactions leading to the DFWM process described in the text. In (a) $A_{1,2}$ pump from the ground state close to a one-photon transition, and A_3 and A_4 correspond to the one-photon decay from that near resonance back to the ground state. (b) describes the case where we are near resonance with a two-photon transition. Two photons from A_1 and A_2 pump close to that transition, and then we have a two-photon decay generating the fields A_3 and A_4 . (c) shows the phase matching of the four wave vectors.

interested in are the values of the fields as they leave the crystal:

$$\begin{aligned}
 A_3^*(L) &= \frac{A_3^*(0)}{\cos |\kappa|L}, \\
 A_4(0) &= \frac{i\kappa}{|\kappa|} (\tan |\kappa|L) A_3^*(0).
 \end{aligned} \tag{3.83}$$

That means the output field A_4' will be the conjugate of the input field A_3' . Another interesting thing to note is that the intensities of the output fields can take on any value from zero to infinity. This is due to the fact that we are continuously pumping the crystal with the fields $E_{1,2}$. Of course, our results here are only valid as long as our initial assumption holds that the fields $E_{3,4}$ are much weaker than the fields $E_{1,2}$.

3.4.3 Phase conjugation - the conclusion

In the preceding sub-section we showed that it is possible to realize a PCM fulfilling the condition $\vec{k} \rightarrow -\vec{k}$ and that the output wave will have a conjugated wavefront. So far, we have not discussed the matter of polarization. There have been theoretical works as well as experiments showing that one can realize a PCM that also fulfills the condition of compensating any non-uniform changes to the polarization of the incoming wavefront. To achieve that, one performs four-wave mixing in an atomic system utilizing two-photon resonances. An important prerequisite in this context is that the two counter-propagating pump fields $E_{1,2}$ have to be circularly polarized and counter-rotating[1].

4 Nonlinear optical effects in optical fibers

So far, we have studied nonlinear optical effects for light passing through bulk media. If light is travelling along a waveguide, all its power can pass through a very small cross section. Consider, for example, a case where we have 1 W of light in an optical fiber with a fiber core diameter of $10\ \mu\text{m}$. In this case, the intensity of the light will be $1\ \text{MW}/\text{cm}^2$. In itself, this is nothing special. We could just take a beam and focus it tightly to a waist of that size. The important thing is that this power will travel along the waveguide, and the field will maintain that high intensity all the way. This makes optical fibers and, more in general, optical waveguides an ideal playground for nonlinear optics.

4.1 A figure of merit for nonlinear effects in fibers

We can make this comparison between nonlinear effects in bulk media and in fibers more precise and derive a figure of merit for comparing these two cases. In both cases, the nonlinear effects will become stronger if we have higher intensity I and if we have a longer interaction length L . We can therefore say that we want to maximize the product $I \cdot L$ in order to maximize nonlinear effects.

In the case of bulk optics, simply increasing the length of the nonlinear medium is not sufficient because diffraction will lead to the beam waist to change along the direction of beam propagation (z axis). In particular, the waist $w(z)$ of a Gaussian beam behaves as follows:

$$w(z) = w_0 \sqrt{1 + \left(\frac{\lambda z}{\pi n w_0^2} \right)^2}. \quad (4.1)$$

Then the optical intensity at position z will be:

$$I_B(z) = \frac{P}{\pi w(z)^2}, \quad (4.2)$$

where B denotes the case of bulk optics. The case for fibers, we will denote with an F . Then we can define the product $[I \cdot L]$ for our figure of merit by integrating over z :

$$[I \cdot L]_B = \int_{-\infty}^{\infty} dz I_B(z) = \pi \frac{nP}{\lambda}. \quad (4.3)$$

From this, we can see that the only way to increase the magnitude of our nonlinear interaction will be to increase the power. Of course, this is not taking into account the possibility of optimizing the material parameters. But these we can also optimize for waveguides.

Now let us compare that with the case where we have our light in an optical fiber. In that case, the waist w_0 will stay constant over the interaction length, but we should also take into account the attenuation along the fiber. The latter is determined by the attenuation coefficient α . The intensity of light inside the fiber will be:

$$I_F(z) = \frac{P \exp(-\alpha z)}{\pi w_0^2}, \quad (4.4)$$

and our product of intensity and length becomes:

$$[I \cdot L]_F = \int_{-\infty}^{\infty} dz I_F(z) = \frac{P}{\pi w_0^2} L_{\text{eff}}. \quad (4.5)$$

Here, we defined the effective interaction length

$$L_{\text{eff}} = \frac{[1 - \exp(-\alpha L)]}{\alpha}. \quad (4.6)$$

If the interaction length L becomes very large, we can write $L_{\text{eff}} \approx 1/\alpha$, and the ratio between those two $[I \cdot L]$ products will become:

$$\frac{[I \cdot L]_F}{[I \cdot L]_B} = \frac{\lambda}{(\pi w_0)^2 n \alpha}. \quad (4.7)$$

As an example, consider the case where we have the following experimental parameters: $P = 1 \text{ W}$, $w_0 = 10 \mu\text{m}$, $n = 1.5$, $\lambda = 1 \mu\text{m}$, and $\alpha = 2.3 \times 10^{-4} \text{ m}^{-1} = 1 \text{ dB/km}$. For these parameters, the ratio in equation 4.7 becomes approximately 5×10^7 . That means, we could expect the same nonlinear effects in a fiber with 1 W of power as in a bulk crystal with 50 MW. This is assuming that we could even have a nonlinear bulk crystal as long as a fiber.

4.2 The intensity-dependent refractive index in an optical fiber

Given the figure of merit we just discussed, some nonlinear optical effects we encountered earlier but dismissed as small effects may become significantly more relevant. For that reason, let us consider how the optical Kerr effect can influence our optical signals if we send them through a fiber. Often, the optical fibers we will be interested in consist of

silica glass (SiO_2). The nonlinearity for that material is typically around $\bar{n}_2 = 3.18 \times 10^{-20} \text{ m}^2/\text{W}$. There are several interesting nonlinear effects we can discuss in the context of silica fibers: (a) optical solitons, (b) optical pulse compression, and (c) modulational instabilities.

4.2.1 Self-phase modulation

Consider a short optical pulse we send through a fiber. The instantaneous phase of the pulse will change rapidly as it travels along the fiber. Due to the optical Kerr effect, the refractive index of the medium will change according to:

$$n(\omega_0, |E|^2) = \frac{c\beta}{\omega_0} = n_0(\omega_0) + \bar{n}_2|E|^2, \quad (4.8)$$

where we used β to denote the (instantaneous) wavenumber of the light inside the fiber. The optical phase of our optical pulse then is defined as $\Phi = \omega_0 t - \beta z$, and the instantaneous frequency of our pulse will be:

$$\omega(t) = \frac{\partial \Phi}{\partial t} = \omega_0 - \frac{\omega_0 \bar{n}_2}{c} z \frac{\partial |E|^2}{\partial t}. \quad (4.9)$$

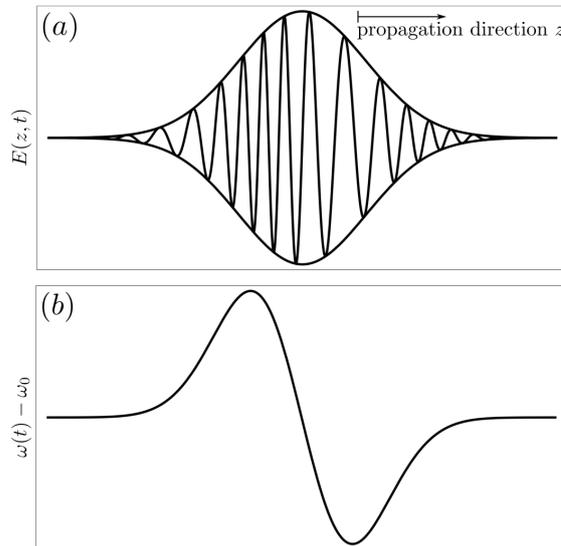


Figure 34: **Self-phase modulation of an optical pulse.** (a) shows the oscillation of the field amplitude and the slowly varying envelop. (b) shows the change in the instantaneous frequency.

This last equation shows that a non-zero nonlinear coefficient \bar{n}_2 will lead to a variation of the frequency as the pulse travels along. This is called self-phase modulation (SPM), and we illustrate the effect in Fig. 34. Part (b) of that figure shows that the instantaneous frequency of the light at first decreases for the leading part of the pulse, and then the frequency increases for the trailing part of the pulse. In tele-communication one often uses zero-dispersion fibers. Such fibers are denoted as dispersion-shifted fibers (DSFs). These will be designed such that there is zero dispersion around a given design wavelength λ_0 . Typical design wavelengths are 1300 nm and 1550 nm. The wavelength range with $\lambda < \lambda_0$ is called the *normal dispersion region*. In that regime, the dispersion in the fiber will be normal (group velocity increases with wavelength). The region with $\lambda > \lambda_0$ is called the *anomalous dispersion region*, where the group velocity decreases with increasing wavelength.

Because the frequency decreases for the leading edge of the pulse, the group velocity will also decrease. At the same time, for the trailing edge of the pulse the frequency will be larger than ω_0 , and that corresponds to a higher group velocity. That means the leading edge will be delayed while the trailing edge will catch up. Effectively, this means that the pulse will be compressed due to this SPM effect. If this compression of the pulse exactly cancels the pulse broadening due to dispersion, the pulse width will remain constant as the pulse travels along the fiber. That means, we will have an *optical soliton*. To be more precise, the envelope of the pulse will keep its shape. Such solitons are therefore sometimes called *envelope solitons*.

In the normal dispersion region, there is also another solution to the wave equation: a *dark soliton*. This describes a solitary wave that results from cutting a portion of the continuous wave, which we will discuss in subsection 4.2.4.

4.2.2 Temporal pulse spreading and the nonlinear Schrödinger equation

Now let us describe the electromagnetic field in a nonlinear optical fiber a bit more precisely. We will see, that the equation governing that propagation can be reduced to an equation often referred to as the nonlinear Schrödinger equation. For simplicity, let us only take into account the propagation direction z and time t for now and not the transverse dimensions x, y , and we want to use this equation to describe the propagation of an optical pulse through a nonlinear medium. We will write the electric field as:

$$E(z, t) = A(z, t)e^{i(k_0 z - \omega_0 t)} + \text{c.c.}, \quad (4.10)$$

where $A(z, t)$ here describes the envelope of the pulse. The wave equation describing the propagation of the pulse through our nonlinear medium is:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 E}{\partial t^2} = 0, \quad (4.11)$$

where D is the total displacement including linear and nonlinear contributions. Let us express $E(z, t)$ and $D(z, t)$ in terms of their Fourier transforms:

$$E(z, t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} E(z, \omega), \quad D(z, t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} D(z, \omega).$$

Then the Fourier components of E and D fulfill the relation:

$$D(z, \omega) = \epsilon_0 \epsilon(\omega) E(z, \omega)$$

with $\epsilon(\omega)$ being the effective dielectric constant.

Using these expressions, we can write the wave equation as:

$$\frac{\partial^2 E(z, \omega)}{\partial z^2} + \epsilon(\omega) \frac{\omega^2}{c^2} E(z, \omega) = 0. \quad (4.12)$$

If we express the Fourier transform of the field in terms of the Fourier transform of the slowly varying field amplitude $A(z, t)$, we get:

$$\begin{aligned} E(z, \omega) &= A(z, \omega - \omega_0) e^{ik_0 z} + A(z, \omega + \omega_0) e^{-ik_0 z} \\ &\approx A(z, \omega - \omega_0) e^{ik_0 z}. \end{aligned} \quad (4.13)$$

The approximation holds true if the amplitude $A(z, t)$ is slowly varying, because in that case there cannot be high-frequency components at $\omega + \omega_0$. If we insert that into the wave equation 4.12, and we drop the second derivatives in z once more, we get:

$$2ik_0 \frac{\partial A}{\partial z} + (k^2 - k_0^2) A = 0, \quad (4.14)$$

where the frequency dependent wavenumber fulfills:

$$k(\omega) = \sqrt{\epsilon(\omega)} \omega / c. \quad (4.15)$$

If k is close to k_0 , we can write $k^2 - k_0^2 \approx 2k_0(k - k_0)$, and by using this we can reduce 4.14 to the *reduced wave equation*:

$$\frac{\partial A(z, \omega - \omega_0)}{\partial z} - i(k - k_0) A(z, \omega - \omega_0) = 0. \quad (4.16)$$

In this context, it is important to keep in mind that k is not simply a variable in the equation above. The wavenumber will depend on the refractive index and therefore on the intensity of the light field. We can take this into account by expanding k in a power series:

$$k = k_0 + \Delta k_{\text{NL}} + k_1(\omega - \omega_0) + \frac{1}{2} k_2(\omega - \omega_0)^2, \quad (4.17)$$

where we introduced the following quantities:

$$\begin{aligned}\Delta k_{\text{NL}} &= \Delta n_{\text{NL}}\omega_0/c = n_2 I\omega_0/c, \\ k_1 &= \left. \frac{dk}{d\omega} \right|_{\omega=\omega_0} = \frac{1}{c} \left[n_{\text{lin}} + \omega \frac{dn_{\text{lin}}}{d\omega} \right]_{\omega=\omega_0} \equiv \frac{1}{v_g}(\omega_0), \\ k_2 &= \left. \frac{d^2k}{d\omega^2} \right|_{\omega=\omega_0} = \frac{d}{d\omega} \left[\frac{1}{v_g(\omega)} \right]_{\omega=\omega_0} = \left[-\frac{1}{v_g^2} \frac{dv_g}{d\omega} \right]_{\omega=\omega_0}.\end{aligned}\quad (4.18)$$

k_1 is the inverse of the *group velocity*, and k_2 is the *group velocity dispersion*.

We then insert that into the reduced wave equation, and then we transform the equation to the time domain by multiplying both sides with $\exp[-i(\omega - \omega_0)t]$ and then integrating everything over all values of $\omega - \omega_0$. This results in the following equation:

$$\frac{\partial A}{\partial z} + k_1 \frac{\partial A}{\partial t} + \frac{1}{2} i k_2 \frac{\partial^2 A}{\partial t^2} - i \Delta k_{\text{NL}} A = 0. \quad (4.19)$$

The second term on the left-hand side describes the spreading of the pulse due to group-velocity dispersion, while the term on the right-hand side describes the spreading of the pulse due to self-phase modulation. As we stated in the beginning of this section, this equation is sometimes referred to as the nonlinear Schrödinger equation.

As a next step, let us coordinate transformation introducing the “retarded time” τ :

$$\tau = t - \frac{z}{v_g} = t - k_1 z. \quad (4.20)$$

If we introduce a new function $A_s(z, \tau) = A(z, t)$ to describe our pulse, then equation 4.19 becomes:

$$\frac{\partial A_s}{\partial z} + \frac{1}{2} i k_2 \frac{\partial^2 A_s}{\partial \tau^2} - i \Delta k_{\text{NL}} A_s = 0. \quad (4.21)$$

By rewriting the nonlinear contribution to the propagation constant as:

$$\Delta k_{\text{NL}} = n_2 \frac{\omega_0}{c} I = 2n_0 \epsilon_0 n_2 \omega_0 |A_s|^2 \equiv \gamma |A_s|^2, \quad (4.22)$$

we can rewrite equation 4.21 as:

$$\frac{\partial A_s}{\partial z} + \frac{1}{2} i k_2 \frac{\partial^2 A_s}{\partial \tau^2} = i \gamma |A_s|^2 A_s. \quad (4.23)$$

4.2.3 Temporal solitons in a fiber

So far, we have treated this as a 1-dimensional problem. Even in a fiber, however, the field will have radial components of course. Let us quickly investigate how this affects our analysis. If we are considering a *single-mode fiber* (SMF), the fiber will only support

HE_{11}^x modes. This is simply a Gaussian mode. In this case, the H and E fields satisfy:

$$\begin{aligned} H_y &= n_1 \epsilon_0 c E_x, \\ E_y &= H_x = 0, \end{aligned} \quad (4.24)$$

where we assumed $k = k_0 n_1$ (light in the core of the fiber, and we use n_0 for the fiber cladding).

The (linear) propagation of light in step-index fibers was covered in the Photonics I course, but to be consistent with the notation here, we need to define a few quantities. In particular, the relative refractive index difference between the refractive index of the core (n_1) and the cladding (n_0) is given by:

$$\Delta = \frac{n_1^2 - n_0^2}{2n_1^2} \approx \frac{n_1 - n_0}{n_1}, \quad (4.25)$$

which is often expressed in percent. The following relation holds between the numerical aperture NA of the fiber and the maximum angle θ_{\max} a ray can have with the propagation direction and still be guided along the fiber:

$$\text{NA} = \theta_{\max} \approx n_1 \sqrt{2\Delta}. \quad (4.26)$$

Within the fiber, the maximum angle for a ray is $\phi_{\max} \approx \theta_{\max}/n_1 \approx \sqrt{2\Delta}$.

For a ray that has an angle ϕ with the z axis inside the fiber, the *propagation constants* along the propagation direction (z) and perpendicular to that (x) are defined as:

$$\begin{aligned} \beta &= k_0 n_1 \cos \phi \\ \kappa &= k_0 n_1 \sin \phi \end{aligned} \quad (4.27)$$

If a mode of order m is to be guided along the fiber, it has to fulfill the following phase-matching condition:

$$v \equiv k_0 n_1 a \sqrt{2\Delta} = \frac{\cos^{-1} \xi + m\pi/2}{\xi}, \quad (4.28)$$

where v is called the normalized frequency, and we use the definition:

$$\xi = \frac{\sin \phi}{\sqrt{2\Delta}}. \quad (4.29)$$

Then the instantaneous peak power of our mode is:

$$P = \int_0^{2\pi} \int_0^\infty dr d\theta r (E_x H_y^* - E_y H_x^*) = n_1 \epsilon_0 c \int_0^{2\pi} \int_0^\infty dr d\theta r |E_x|^2. \quad (4.30)$$

If we assume that the peak power is achieved for $\tau = 0$, then we can replace E_x with $R(r, \theta)A_s(0, 0)$ in the above equation, and we get:

$$P = n_1 \epsilon_0 c |A_s^0|^2 \int_0^{2\pi} \int_0^\infty dr d\theta r |R|^2.$$

This is related to the cross-sectional area occupied by the light field in the fiber. In particular, this *effective area* is defined as[5]:

$$A_{\text{eff}} = \frac{\left[\int_0^{2\pi} \int_0^\infty dr d\theta r |R|^2 \right]^2}{\int_0^{2\pi} \int_0^\infty dr d\theta r |R|^4}. \quad (4.31)$$

If we use this definition, and if we define:

$$\eta = \frac{\int_0^{2\pi} \int_0^\infty dr d\theta r |R|^4}{\int_0^{2\pi} \int_0^\infty dr d\theta r |R|^2}, \quad (4.32)$$

then we can write P as:

$$P = \eta n_1 \epsilon_0 c |A_p|^2 A_{\text{eff}} \approx \frac{1}{2} n_1 \epsilon_0 c |A_p|^2 A_{\text{eff}}, \quad (4.33)$$

where we introduced the peak amplitude $A_p = A_s(0, 0)$. The approximation on the right-hand side assumes that $\eta \approx 0.5$. This is the case for the “normal” operation of optical fibers. In that case, the normalized frequency v is between 1.5 and 2.5. That is the range of single-mode operation (or at least not very multi-mode).

Let us now reconsider the nonlinear Schrödinger equation 4.23 we derived. In the context of self trapping, we derived equation 3.51, which has similar features to the nonlinear Schrödinger equation and had spatial solitons as a solution. It should therefore not surprise us that equation 4.23 has a similar solution of the form:

$$A_s(z, \tau) = A_s^0 \text{sech}(\tau/\tau_0) e^{i\zeta z}, \quad (4.34)$$

where the pulse amplitude A_s^0 and the pulse width τ_0 must fulfill:

$$|A_s^0|^2 = \frac{-k_2}{\gamma \tau_0^2} = \frac{-k_2}{2n_0 \epsilon_0 n_2 \omega_0 \tau_0^2}, \zeta = -k_2/2\tau_0^2 = \frac{1}{2} \gamma |A_s^0|^2.$$

ζ is the phase shift experienced by the soliton pulse during propagation - similar to the phase shift we had in the case of the spatial soliton.

4.2.4 Dark solitons

Bibliography

- [1] Robert W. Boyd. *Nonlinear Optics*. Academic Press, Inc., USA, 3rd edition, 2008. [2](#), [3](#), [4](#), [5](#), [6](#), [7](#), [10](#), [11](#), [12](#), [13](#), [14](#), [15](#), [17](#), [21](#), [28](#), [29](#), [32](#), [35](#), [52](#), [55](#), [61](#), [62](#), [63](#), [65](#), [70](#), [71](#), [74](#)
- [2] D. Meschede. *Optics, Light and Lasers*. Wiley-VCH, Weinheim, Germany, 2017. [24](#), [25](#), [36](#)
- [3] Ilias Pappas, Stylianos Siskos, and Charalambos A. Active-Matrix Liquid Crystal Displays - Operation, Electronics and Analog Circuits Design. In *New Developments in Liquid Crystals*. InTech, nov 2009. [36](#)
- [4] R. Y. Chiao, E. Garmire, and C. H. Townes. Self-Trapping of Optical Beams. *Physical Review Letters*, 13(15):479–482, oct 1964. [63](#), [66](#)
- [5] K. Okamoto. *Fundamentals of Optical Waveguides*. Elsevier Academic Press, 2006. [82](#)