

Nonlinear Optics

Lecture Notes

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The Nonlinear Optical Susceptibility

1.1 The wave equation and the polarization vector

Nonlinear optics is a branch in optics which considers the following:

1. modification of the optical properties of a medium by light
2. optical wave equations which include non linear terms in the field (e.g. $E^2, E^3\dots$)
3. processes that generate new optical frequencies (see Fig.1.1)



Fig. 1.1 Generation of new frequency components

The response of any material (even vacuum) to light is essentially non-linear. However, to observe nonlinear phenomena we need to apply high optical intensities. Let us start with the wave equation for the electric field:

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (1.1)$$

\mathbf{P} is the polarization which is the source term in the equation.

$\varepsilon_0 = 8.85e - 12$ F/m is the vacuum permittivity

$c = 3e8$ m/s is the speed of light

All the terms in the equation depend on both the coordinates $\mathbf{r} = (x, y, z)$ and time t .

In writing Eq.1.1 we have assumed that $\nabla \cdot \mathbf{E} = 0$ which is correct for transverse infinite plane waves, while this is only an approximation for the general case in nonlinear optics.

The polarization is usually split into linear and nonlinear terms:

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{NL}, \quad (1.2)$$

where $\mathbf{P}^{(1)}$ depends linearly on the field.

Using the displacement field $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$ and separating it also to a linear and nonlinear terms:

$$\mathbf{D} = \mathbf{D}^{(1)} + \mathbf{P}^{NL} \quad (1.3)$$

where $\mathbf{D}^{(1)} = \varepsilon_0 \mathbf{E} + \mathbf{P}^{(1)}$ we can set the wave equations as:

$$\nabla^2 \mathbf{E} - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2}{\partial t^2} \mathbf{D}^{(1)} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2} \quad (1.4)$$

For a lossless and dispersion-less medium (or at least for CW fields) we can relate the displacement to the field through:

$$\mathbf{D}^{(1)} = \varepsilon_0 \boldsymbol{\epsilon}^{(1)} \cdot \mathbf{E}, \quad (1.5)$$

where $\boldsymbol{\epsilon}^{(1)}$ is the *dielectric tensor*. For an isotropic material this last relation is reduced to :

$$\mathbf{D}^{(1)} = \varepsilon_0 \varepsilon^{(1)} \mathbf{E}, \quad (1.6)$$

where now $\varepsilon^{(1)}$ is a scalar related to the index of refraction through:

$$\varepsilon^{(1)} = n^2. \quad (1.7)$$

So, for a dispersion-less, isotropic material the wave equation is given with:

$$\nabla^2 \mathbf{E} - \frac{n^2}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2} \quad (1.8)$$

It is convenient in many cases to separate the field into a sum of modes around different (temporal and spatial) carrier frequencies:

$$\mathbf{E} = \sum_n \tilde{\mathbf{E}}_n(\mathbf{r}, t) = \sum_{n; \omega_n > 0} \mathbf{E}_n(\mathbf{r}, t) e^{-i\omega_n t} + c.c \quad (1.9)$$

$$= \sum_{n; \omega_n > 0} \mathbf{A}_n(\mathbf{r}, t) e^{i(\mathbf{k}_n \cdot \mathbf{r} - \omega_n t)} + c.c \quad (1.10)$$

where c.c. stand for complex conjugate, and we have used: $\mathbf{E}_n(\mathbf{r}, t) = \mathbf{A}_n(\mathbf{r}, t) e^{i\mathbf{k}_n \cdot \mathbf{r}}$.

An alternative notation uses $\mathbf{E}_n(\mathbf{r}, t) = \mathbf{E}(\omega_n; \mathbf{r}, t)$ and $\mathbf{A}_n(\mathbf{r}, t) = \mathbf{A}(\omega_n; \mathbf{r}, t)$ with which we can write the complex conjugate part, using: $\mathbf{E}(-\omega_n; \mathbf{r}, t) = \mathbf{E}(\omega_n; \mathbf{r}, t)^*$ and $\mathbf{A}(-\omega_n; \mathbf{r}, t) = \mathbf{A}(\omega_n; \mathbf{r}, t)^*$. With this notation we can write:

$$\mathbf{E} = \sum_n \mathbf{E}(\omega_n; \mathbf{r}, t) e^{-i\omega_n t} = \sum_n \mathbf{A}(\omega_n; \mathbf{r}, t) e^{i(\mathbf{k}_n \cdot \mathbf{r} - i\omega_n t)} \quad (1.11)$$

In cases where the modes are monochromatic (CW): $\mathbf{E}_n = \mathbf{E}_n(\mathbf{r})$ and $\mathbf{A}_n = \mathbf{A}_n(\mathbf{r})$.

For the polarization, the separation to a superposition of modes can be applied to both the linear and nonlinear terms. For example, for the nonlinear term:

$$\mathbf{P}^{NL} = \sum_{n; \omega_n > 0} \tilde{\mathbf{P}}_n^{NL}(\mathbf{r}, t) e^{i(\mathbf{k}_n \cdot \mathbf{r} - \omega_n t)} = \sum_{n; \omega_n > 0} \mathbf{P}_n^{NL}(\mathbf{r}, t) e^{-i\omega_n t} \quad (1.12)$$

$$= \sum_n \mathbf{P}^{NL}(\omega_n; \mathbf{r}, t) e^{-i\omega_n t} \quad (1.13)$$

The same applies for the linear term.

For CW modes: $\tilde{\mathbf{P}}_n^{NL} = \tilde{\mathbf{P}}_n^{NL}(\mathbf{r})$, $\mathbf{P}_n^{NL} = \mathbf{P}_n^{NL}(\mathbf{r})$ and $\mathbf{P}^{NL}(\omega_n) = \mathbf{P}^{NL}(\omega_n; \mathbf{r})$ and the same, again, applies for the linear terms.

Further, for CW modes:

$$\mathbf{P}_n^{(1)}(\mathbf{r}) = \varepsilon_0 \chi^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) \quad (1.14)$$

where $\chi^{(1)}$ is a second rank tensor - it connects between any single component of the field to a single component of the polarization - thus is connect between pairs of components, which explains its rank. This also means that this tensor needs to have two indices to describe its components. Generally, the number of components of tensor equals to its rank. This can be written explicitly (for CW) at a given point $\mathbf{r} = \mathbf{r}_0$:

$$P_i(\omega_n) = \sum_j \chi_{ij}^{(1)}(\omega_n) E_j(\omega_n) \quad (1.15)$$

Here the indices i and j stands for the three cartesian directions. $\chi^{(1)}$ is made of 9 different components, which can be represented by a 3X3 matrix.

We would see below the generalization of this description for the case of modes which are time dependent .

Now we can write (for CW):

$$\mathbf{D}_n^{(1)}(\mathbf{r}) = \varepsilon_0 (1 + \chi^{(1)}(\omega_n)) \cdot \mathbf{E}_n(\mathbf{r}) = \varepsilon_0 \epsilon^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) \quad (1.16)$$

Here $\epsilon^{(1)}$ is the dielectric tensor. For an isotropic material this tensor reduces to a scalar and is related to the index of refraction n through: $n^2 = \epsilon^{(1)}$.

With this expression we can set Eq.1.4 for a single CW mode using eq.1.9 to be:

$$\nabla^2 \mathbf{E}_n(\mathbf{r}) + \frac{\omega_n^2}{c^2} \epsilon^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) = -\frac{\omega_n^2}{\varepsilon_0 c^2} \mathbf{P}_n^{NL}(\mathbf{r}). \quad (1.17)$$

As was mentioned, the polarization is a function of the field \mathbf{E} . In case that the maximum applied field is much smaller than the fields already existing between the charges in a given material (e.e. the coulomb field binding an electron to the atom's nucleus) the polarization can be expanded, by virtue of perturbation theory as a power sum in the field. For a model in which all fields and polarizations are

acing only along a given direction (can be represented as scalars) and in which the material response to the field is instantaneous and homogeneous, the polarization is written as:

$$P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} E^2 + \varepsilon_0 \chi^{(3)} E^3 + \dots \quad (1.18)$$

with $P = P(\mathbf{r}, t)$, $E = E(\mathbf{r}, t)$ and $\chi^{(n)}$ are the n-th order optical susceptibilities.

Let us assume a simple case in which $E = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c.$ and consider the 2nd order nonlinear response:

$$\varepsilon_0 \chi^{(2)} E^2 = \varepsilon_0 \chi^{(2)} [E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_1^* e^{i\omega_1 t} + E_2^* e^{i\omega_2 t}]^2 \quad (1.19)$$

We get 16 different terms here that we can sort using the notation in Eq.1.13.

Terms where the same frequency is added together are known as Second-Harmonic-Generation (SHG):

$$SHG : \quad P(2\omega_1) = \varepsilon_0 \chi^{(2)} E_1^2; \quad P(2\omega_2) = \varepsilon_0 \chi^{(2)} E_2^2 \quad (1.20)$$

The term in which different frequencies are added is known as Sum-Frequency-Generation (SFG):

$$SFG : \quad P(\omega_1 + \omega_2) = \varepsilon_0 \chi^{(2)} E_1 E_2 \quad (1.21)$$

(essentially this term and its conjugate give together the total *real* SFG polarization)

The term in which different frequencies are subtracted is known as Difference-Frequency-Generation (DFG):

$$DFG : \quad P(\omega_1 - \omega_2) = \varepsilon_0 \chi^{(2)} E_1 E_2^*. \quad (1.22)$$

Finally, the term where the same frequency is subtracted to give a term "oscillating" at zero frequency is known as optical rectification (OR):

$$OR : \quad P(0) = \varepsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*). \quad (1.23)$$

We will consider now more complicated models for the polarization.

1.2 Temporal description of the susceptibility

The model we consider first considers only the temporal behavior of the susceptibility. For this reason, the model is homogenous and scalar. Homogeneous: we assume there is no dependency on the coordinate \mathbf{r} . Scalar: we ignore the vectorial

nature of all the fields, so we treat only cases in which the fields and the induced polarization are along the same direction.

The temporal model assumes that the material response to applied fields is non-instantaneous, that is - the response of the material at a given time depends on past events.

1.2.1 Linear response

Starting with the linear response at a given point r :

$$P^{(1)}(t) = \varepsilon_0 \int_0^\infty R^{(1)}(\tau) E(t - \tau) d\tau \quad (1.24)$$

$R^{(1)}(\tau)$ is the temporal linear response function. It describes the linear response of the material at time t due to the field applied at earlier times $t - \tau$. Due to causality τ cannot be negative: $R^{(1)}(\tau) = 0; \forall \tau < 0$.

To get the corresponding expression in the frequency domain we use the Fourier pair:

$$E(\omega) = \int_{-\infty}^{\infty} E(t) e^{i\omega t} dt \quad (1.25)$$

$$E(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega) e^{-i\omega t} d\omega \quad (1.26)$$

To get:

$$\begin{aligned} P^{(1)}(t) &= \varepsilon_0 \int_0^\infty d\tau \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} R^{(1)}(\tau) E(\omega) e^{-i\omega(t-\tau)} \\ &= \varepsilon_0 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \int_0^\infty d\tau R^{(1)}(\tau) e^{i\omega\tau} E(\omega) e^{-i\omega t} \end{aligned} \quad (1.27)$$

Defining the frequency response of the system as

$$\chi^{(1)}(\omega) \triangleq \int_0^\infty d\tau R^{(1)}(\tau) e^{i\omega\tau} \quad (1.28)$$

we get that:

$$P^{(1)}(t) = \varepsilon_0 \frac{1}{2\pi} \int_{-\infty}^{\infty} \chi^{(1)}(\omega) E(\omega) e^{-i\omega t} d\omega \quad (1.29)$$

Because we can also write:

$$P^{(1)}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} P^{(1)}(\omega) e^{-i\omega t} d\omega \quad (1.30)$$

we get that:

$$P^{(1)}(\omega) = \varepsilon_0 \chi^{(1)}(\omega) E(\omega) \quad (1.31)$$

We note that another acceptable notation for the susceptibility is as follows:
 $\chi^{(1)} = \chi^{(1)}(\omega; \omega) = \chi^{(1)}$ (response at frequency ω ; due to field at frequency ω)

If the material response was instantaneous: $R^{(1)}(\tau) = \chi_0^{(1)} \delta(\tau)$ which leads to:
 $P^{(1)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} \chi_0^{(1)} \delta(\tau) E(t - \tau) d\tau = \varepsilon_0 \chi_0^{(1)} E(t)$,
while in the frequency domain: $\chi^{(1)}(\omega) = \int_0^{\infty} d\tau \chi_0^{(1)} \delta(\tau) e^{i\omega\tau} = \chi_0^{(1)}$ which gives
 $P^{(1)}(\omega) = \varepsilon_0 \chi_0^{(1)} E(\omega)$. Which means there is no dispersion in this case.

1.2.2 2nd order nonlinear response

Here:

$$P^{(2)}(t) = \varepsilon_0 \int_0^{\infty} d\tau_1 \int_0^{\infty} d\tau_2 R^{(2)}(\tau_1, \tau_2) E(t - \tau_1) E(t - \tau_2) \quad (1.32)$$

Here $R^{(2)}(\tau_1, \tau_2)$ connects the contribution from the field at two earlier times to the polarization at time t .

Causality dictates that $R^{(2)}(\tau_1, \tau_2) = 0 ; \forall \tau_1, \tau_2 < 0$.

Writing the field using its Fourier transform:

$$P^{(2)}(t) = \varepsilon_0 \int_0^{\infty} d\tau_1 \int_0^{\infty} d\tau_2 R^{(2)}(\tau_1, \tau_2) \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} E(\omega_1) E(\omega_2) e^{i\omega_1 \tau_1} e^{i\omega_2 \tau_2} e^{-i\omega_1 t} e^{-i\omega_2 t} \quad (1.33)$$

We define:

$$\chi^{(2)}(\omega_1, \omega_2) \triangleq \int_0^{\infty} d\tau_1 \int_0^{\infty} d\tau_2 R^{(2)}(\tau_1, \tau_2) e^{i(\omega_1 \tau_1 + \omega_2 \tau_2)} \quad (1.34)$$

to get:

$$P^{(2)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \chi^{(2)}(\omega_1, \omega_2) E(\omega_1) E(\omega_2) e^{-i\omega_\sigma t}, \quad (1.35)$$

where $\omega_\sigma = \omega_1 + \omega_2$.

We can use the notation $\chi^{(2)} = \chi^{(2)}(\omega_\sigma; \omega_1, \omega_2)$.

The instantaneous case would be represented by $R^{(2)} = \chi_0^{(2)} \delta(\tau_1) \delta(\tau_2)$ which leads to

$$P^{(2)}(t) = \varepsilon_0 \chi_0^{(2)} (E(t))^2 \quad (1.36)$$

while in the frequency domain: $\chi^{(2)}(\omega_1, \omega_2) = \chi_0^{(2)}$ which gives:

$$P^{(2)}(\omega) = \int_{-\infty}^{\infty} P^{(2)}(t) e^{i\omega t} dt = \varepsilon_0 \chi_0^{(2)} E(\omega) * E(\omega), \quad (1.37)$$

where $*$ stands for convolution.

For a bi-chromatic field

$$\begin{aligned} E(\omega) &= \sum_{n=1,2} \tilde{E}(\omega_n) \delta(\omega - \omega_n) + \tilde{E}(-\omega_n) \delta(\omega + \omega_n) \\ &= \sum_{n=1,2} \tilde{E}(\omega_n) \delta(\omega - \omega_n) + \tilde{E}^*(\omega_n) \delta(\omega + \omega_n), \end{aligned} \quad (1.38)$$

we get a product of the amplitudes in the spectral domain because in this case, when the two fields are at frequencies ω_1 and ω_2 we get:

$$\begin{aligned} E(\omega) * E(\omega) &= 2\tilde{E}_1(\omega_1)\tilde{E}_2(\omega_2)\delta(\omega - (\omega_1 + \omega_2)) + 2\tilde{E}_1^*(\omega_1)\tilde{E}_2^*(\omega_2)\delta(\omega + (\omega_1 + \omega_2)) \\ &\quad + 2\tilde{E}_1(\omega_1)\tilde{E}_2^*(\omega_2)\delta(\omega - (\omega_1 - \omega_2)) + 2\tilde{E}_1^*(\omega_1)\tilde{E}_2(\omega_2)\delta(\omega + (\omega_1 - \omega_2)) \\ &\quad + (\tilde{E}_1(\omega_1))^2 \delta(\omega - 2\omega_1) + (\tilde{E}_1^*(\omega_1))^2 \delta(\omega + 2\omega_1) \\ &\quad + (\tilde{E}_2(\omega_2))^2 \delta(\omega - 2\omega_2) + (\tilde{E}_2^*(\omega_2))^2 \delta(\omega + 2\omega_2) \\ &\quad + 2(|E_1(\omega_1)|^2 + |E_2(\omega_1)|^2)\delta(0). \end{aligned} \quad (1.39)$$

Overall we have $4 \times 4 = 16$ terms at 5 distinct frequencies $\omega_1 + \omega_2, \omega_1 - \omega_2, 2\omega_1, 2\omega_2$ and 0.

Inversion symmetry

In the context of the temporal description of the 2nd order nonlinear response, it is appropriate to mention the effect that a system with inversion symmetry has on any even order nonlinear polarization term. A system with inversion symmetry is also known as a centrosymmetric - this means the system possesses a center of inversion. Every physical property of the medium should be oblivious to the transformation: $\mathbf{r} \rightarrow -\mathbf{r}$. If we have a field acting on the system and this field is polarized along some direction $\hat{\mathbf{x}}$, operating with the inversion operator just inverts the sign of the field: $\mathbf{E} = E(t)\hat{\mathbf{x}} \rightarrow -\mathbf{E} = E(t)(-\hat{\mathbf{x}}) = (-E(t))\hat{\mathbf{x}}$. Let us assume that the material system possesses an instantaneous nonlinear susceptibility giving rise to nonlinear polarization. We can write a scalar relationship between the field and nonlinear polarization:

$$P(t) = \varepsilon_0 \chi^{(2)} E(t)^2. \quad (1.40)$$

If we operate now with the inversion operator it should change the sign of both the applied field and polarization (because they both have a specific direction - defined by their polarization).

At the same time the symmetry of the system assures us that the susceptibility does not change. Overall:

$$-P(t) = \varepsilon_0 \chi^{(2)} [-E(t)]^2 = P(t), \quad (1.41)$$

which can only be true if $P(t) = 0$ for any applied non zero field. This requires that $\chi^{(2)} = 0$.

This result is true for any nonlinear response of an even order.

We note that this symmetry is true as long as the Lorentz magnetic force $q\mathbf{v} \times \mathbf{B}$ can be neglected (which is for most cases we are interested at). In first order nonlinear optics the velocity $\propto \mathbf{E}$ giving rise to a nonlinear polarization term of the form $\mathbf{E} \times \mathbf{B}$. However because under $\mathbf{r} \rightarrow -\mathbf{r}$, while $\mathbf{E} \rightarrow -\mathbf{E}$ the magnetic field does not change $\mathbf{B} \rightarrow \mathbf{B}$ because it is an axial (pseudo) vector. This means that overall $\mathbf{E} \times \mathbf{B} \rightarrow -\mathbf{E} \times \mathbf{B}$ and so the nonlinear susceptibility associated with this term does not vanish in a centrosymmetric media. By the way, a simple way to observe that \mathbf{B} is a pseudo-vector is to notice that in the expression for the Lorentz force $\mathbf{F} = q(\mathbf{E} + \mathbf{v} \times \mathbf{B})$ the force \mathbf{F} and the velocity \mathbf{v} are real vectors (that change sign upon inversion). This means that \mathbf{B} cannot change sign under inversion.

1.2.3 3rd order nonlinear response

Similarly to the previous cases, the 3rd order polarization is:

$$P^{(3)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \chi^{(3)}(\omega_1, \omega_2, \omega_3) E(\omega_1) E(\omega_2) E(\omega_3) e^{-i\omega_\sigma t}, \quad (1.42)$$

where $\omega_\sigma = \omega_1 + \omega_2 + \omega_3$ and:

$$\chi^{(3)}(\omega_\sigma; \omega_1, \omega_2, \omega_3) \triangleq \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 R^{(3)}(\tau_1, \tau_2, \tau_3) e^{i(\omega_1 \tau_1 + \omega_2 \tau_2 + \omega_3 \tau_3)}. \quad (1.43)$$

1.3 Tensorial description of the susceptibility for 2nd order nonlinear response

Here we take into account the vectorial nature of the fields and the possible response of the material to fields applied in different directions. All other complications are set aside - so the model is homogenous and instantaneous and the applied

field is monochromatic, or consists of a discrete superposition of monochromatic components:

$$\mathbf{E} = \sum_n \mathbf{E}(\omega_n; \mathbf{r}) e^{-i\omega_n t} = \sum_n \mathbf{A}(\omega_n; \mathbf{r}) e^{i(\mathbf{k}_n \cdot \mathbf{r} - i\omega_n t)} \quad (1.44)$$

and the polarization is:

$$\mathbf{P} = \sum_n \mathbf{P}(\omega_n; \mathbf{r}) e^{-i\omega_n t} \quad (1.45)$$

1.3.1 Form of the 2nd order nonlinear polarization

For the following we consider the nonlinear response of a medium at a given point $\mathbf{r} = \mathbf{r}_0$.

As a 3rd rank tensor $\chi^{(2)}$ connects between any two different components of the field to a single component of the polarization. Thus it makes connection between triplets of components. Generally for a nonlinear optical process of order n , there is a tensor describing it with rank $n+1$. For the 2nd order process we are considering here we can write:

$$P_i(\omega_n + \omega_m) = \varepsilon_0 \sum_{jk} \sum_{(nm)} \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m). \quad (1.46)$$

Here i, j, k are cartesian coordinates. The notation (nm) indicates that we should sum on all values of ω_n and ω_m as long as we keep $\omega_n + \omega_m$ constant.

Summation over the (nm) indices results in a number D known as the *degeneracy factor*. With which we can write:

$$P_i(\omega_n + \omega_m) = \varepsilon_0 D \sum_{jk} \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) E_j(\omega_n) E_k(\omega_m). \quad (1.47)$$

Regarding the frequencies on which the nonlinear susceptibility is dependent - there is another common notation: $\chi^{(2)}(\omega_3; \omega_2, \omega_1)$ with $\omega_3 = \omega_2 + \omega_1$.

For a specific set of the frequencies the tensor $\chi^{(2)}$ has a total of $3 \cdot 3 \cdot 3 = 27$ components.

However, on top of that, for an interaction leading to the polarization term of Eq. 1.46 there are 12 different combinations of the frequencies, stemming from the relation $\omega_3 = \omega_2 + \omega_1$ when we also note that each frequency can be related to a different component of the field and that the equation is still valid when we negate both sides. So there are 6 ways to arrange three frequencies in three directions, and we multiply this number by 2 for the negative frequency components.

Let's see a few examples:

$$\omega_3 = \omega_2 + \omega_1 \rightarrow \chi^{(2)}(\omega_3; \omega_2, \omega_1)$$

$$\begin{aligned}\omega_3 &= \omega_1 + \omega_2 \rightarrow \chi^{(2)}(\omega_3; \omega_1, \omega_2) \\ -\omega_3 &= (-\omega_2) + (-\omega_1) \rightarrow \chi^{(2)}(-\omega_3; -\omega_2, -\omega_1)\end{aligned}$$

In total we have then 12 different tensors, each containing 27 components, that is - we need to know $27 \cdot 12 = 324$ numbers to describe all aspect of the second order interaction. Luckily, different symmetries greatly reduce the number of independent numbers defining the nonlinear susceptibility. We'll consider these symmetries next.

1.3.2 Symmetries of the 2nd order nonlinear response

Reality of the fields

We remind that the reality of the fields forces for our nomenclature that: $\mathbf{E}(\omega_n) = \mathbf{E}(-\omega_n)^*$

This is also true for the polarization and in particular to any component of the field and of the polarization. With regards to Eq. 1.46 this sets: $P_i(-\omega_n - \omega_m) = P_i(\omega_n + \omega_m)^*$, $E_j(-\omega_n) = E_j(\omega_n)^*$ and $E_k(-\omega_m) = E_k(\omega_m)^*$. Using these relations in Eq. 1.46, it forces the symmetry:

$$\chi_{ijk}^{(2)}(-\omega_n - \omega_m; -\omega_n, -\omega_m) = \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m)^* \quad (1.48)$$

Intrinsic permutation symmetry

The polarization in Eq. 1.46 is dependent on multiplication of the form $E_j(\omega_n)E_k(\omega_m)$. Now, it should not really matter what is the order of the fields in this multiplication, so $E_k(\omega_m)E_j(\omega_n)$ should work exactly the same. But these two terms there come with two different components of the susceptibility:

$$\begin{aligned}E_j(\omega_n)E_k(\omega_m) &\rightarrow \chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) \\ E_k(\omega_m)E_j(\omega_n) &\rightarrow \chi_{ikj}^{(2)}(\omega_n + \omega_m; \omega_m, \omega_n)\end{aligned}$$

To reconcile this, we must have $\chi_{ijk}^{(2)}(\omega_n + \omega_m; \omega_n, \omega_m) = \chi_{ikj}^{(2)}(\omega_n + \omega_m; \omega_m, \omega_n)$.

So - we can permute between the indices (except for i) as long as we carry the frequencies together with them and the associate value of the susceptibility should remain the same.

Full permutation symmetry

Developing a quantum mechanical model for the nonlinear susceptibility leads to two important results when the interacting frequencies in a nonlinear medium are far off any resonance:

- The nonlinear susceptibility is real - we neglect absorption (but still retain dispersion)

- There is a full permutation symmetry - a symmetry under permutation of *all* the indices of the nonlinear susceptibility when the frequencies are carried with the indices.

The full permutation symmetry states that, for example:

$$\chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{jki}^{(2)}(-\omega_1 = \omega_2 - \omega_3) \quad (1.49)$$

Another relation we can find is by using the last equation together with the symmetry due to the reality of the fields Eq. 1.48, leading here to $\chi_{jki}^{(2)}(-\omega_1 = \omega_2 - \omega_3) = \chi_{jki}^{(2)}(\omega_1 = -\omega_2 + \omega_3)^*$, together with the reality of the nonlinear susceptibility in the lossless case, giving $\chi_{jki}^{(2)}(\omega_1 = -\omega_2 + \omega_3)^* = \chi_{jki}^{(2)}(\omega_1 = -\omega_2 + \omega_3)$ to find that:

$$\chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{jki}^{(2)}(\omega_1 = -\omega_2 + \omega_3) \quad (1.50)$$

Kleinman's Symmetry

If the frequencies involved in the interaction are much smaller than the lowest resonance of the material - the nonlinear susceptibility becomes independent of frequency. Essentially this means it is instantaneous (see section 1.2.2). If we consider such a dispersion-less behavior together with the full permutation symmetry - we can permute the indices *without* carrying the frequencies with them, leading for example to :

$$\chi_{ijk}^{(2)}(\omega_3 = \omega_1 + \omega_2) = \chi_{jki}^{(2)}(\omega_3 = \omega_1 + \omega_2) \quad (1.51)$$

This result is known as Kleinman's symmetry.

1.3.3 Contracted notation

We define now the "d" tensor: $d_{ijk} = \frac{1}{2}\chi_{ijk}^{(2)}$, with which we write the polarization:

$$P_i(\omega_n + \omega_m) = 2\epsilon_0 \sum_{jk} \sum_{(nm)} d_{ijk} E_j(\omega_n) E_k(\omega_m). \quad (1.52)$$

Now, if $\omega_n = \omega_m$ (a case known as second-harmonic-generation) it is obvious that $d_{ijk} = d_{ikj}$ because we cannot differentiate between the fields. This symmetry also exists when there is Kleinman's Symmetry - as we can permute any indices that we want. In these cases there is no need for three indices in the description of the d-tensor, as the last two are degenerate. Because of that we can adopt a contracted notation d_{il} according to the rule:

$$\begin{array}{llllll} jk : & 11 & 22 & 33 & 23, 32 & 31, 13 & 12, 21 \\ l : & 1 & 2 & 3 & 4 & 5 & 6 \end{array} \quad (1.53)$$

So instead of using $3 \cdot 3 \cdot 3 = 27$ numbers for the susceptibility we can use $3 \cdot 6 = 18$ numbers. We can arrange these numbers in a 3×6 matrix form:

$$\begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \quad (1.54)$$

However, we have not finished exhausting all the options given with the existence of Kleinman symmetry. For example:

$$d_{12} \equiv d_{122} = d_{212} \equiv d_{26} \quad (1.55)$$

and:

$$d_{14} \equiv d_{123} = d_{213} \equiv d_{25} \quad (1.56)$$

If we keep on in this manner we find that of the 18 elements of the contracted notation only 10 are independent:

$$\begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{bmatrix} \quad (1.57)$$

1.3.4 Sum-frequency generation

With sum-frequency generation (SFG) we consider a process in which two input fields with frequencies ω_1 and ω_2 give rise to photons at the sum frequency $\omega_3 = \omega_1 + \omega_2$. In this case the (nm) summation in Eq.1.46 gives:

$$P_i(\omega_3) = \varepsilon_0 \sum_{jk} [\chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2) + \chi_{ijk}^{(2)}(\omega_3; \omega_2, \omega_1) E_j(\omega_2) E_k(\omega_1)]. \quad (1.58)$$

As the j, k indices are just dummy indices - we can switch between them, but we would do so only in the second expression (formally we make a change in the name of the indices $j \rightarrow k$ and $k \rightarrow j$). Then we would also make use of the intrinsic permutation symmetry (which is always true):

$\chi_{ikj}^{(2)}(\omega_3; \omega_2, \omega_1) = \chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_2)$ which together would yield the result:

$$P_i(\omega_3) = 2\varepsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2). \quad (1.59)$$

If both input fields are polarized along a specific direction, say x , we'll get:

$$P_i(\omega_3) = 2\epsilon_0 \chi_{ixx}^{(2)}(\omega_3; \omega_1, \omega_2) E_x(\omega_1) E_x(\omega_2). \quad (1.60)$$

Using the matrix form of the contracted notation of the susceptibility we can write all of the polarization component together for SFG:

$$\begin{bmatrix} P_x(\omega_3) \\ P_y(\omega_3) \\ P_z(\omega_3) \end{bmatrix} = 4\epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_x(\omega_1) E_x(\omega_2) \\ E_y(\omega_1) E_y(\omega_2) \\ E_z(\omega_1) E_z(\omega_2) \\ E_y(\omega_1) E_z(\omega_2) + E_z(\omega_1) E_y(\omega_2) \\ E_x(\omega_1) E_z(\omega_2) + E_z(\omega_1) E_x(\omega_2) \\ E_x(\omega_1) E_y(\omega_2) + E_y(\omega_1) E_x(\omega_2) \end{bmatrix} \quad (1.61)$$

1.3.5 Second-harmonic generation

Second-harmonic generation (SHG) is a special case of SFG in which $\omega_1 = \omega_2$ and so $\omega_3 = 2\omega_1$. Returning to Eq.1.46 the (nm) summation has only one single term and so:

$$P_i(\omega_3) = \epsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_1) E_j(\omega_1) E_k(\omega_1). \quad (1.62)$$

If the input field is polarized along a specific direction, say x , we get:

$$P_i(\omega_3) = \epsilon_0 \chi_{ixx}^{(2)}(\omega_3; \omega_1, \omega_1) E_x(\omega_1)^2. \quad (1.63)$$

We note the factor of 2 between the polarization for SHG and SFG. This is explained by anticipating that the nonlinear polarization produced by two different fields would be larger than that produced by a single field (when all the fields have the same amplitudes).

Using the matrix form of the contracted notation of the susceptibility we can write all of the polarization component together for SHG:

$$\begin{bmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_x(\omega)^2 \\ E_y(\omega)^2 \\ E_z(\omega)^2 \\ 2E_y(\omega) E_z(\omega) \\ 2E_x(\omega) E_z(\omega) \\ 2E_x(\omega) E_y(\omega) \end{bmatrix} \quad (1.64)$$

1.3.6 Effective nonlinear coefficient

Generally, for a given propagation direction in a specific material with given polarization of the fields, it is possible to write a scalar relationship between the size

of the fields $E(\omega) = |\mathbf{E}(\omega)|$ and polarization $P(\omega) = |\mathbf{P}(\omega)|$ with the help of an effective nonlinear coefficient d_{eff} .

For SFG, we would write:

$$P(\omega_3) = 4\epsilon_0 d_{eff} E(\omega_1) E(\omega_2) \quad (1.65)$$

and for SHG:

$$P(2\omega) = 2\epsilon_0 d_{eff} E(\omega)^2 \quad (1.66)$$

For each material (crystal symmetry class) there are specific expressions for calculating the effective nonlinearity as a function of the propagation direction of the pump beam with respect to the crystal principal axes.

1.4 The classical anharmonic oscillator - for the 2nd order nonlinear optical susceptibility

The most accurate physical models for the optical susceptibility is based on quantum mechanics which is beyond our scope. Here we use an extension of the Lorentz model which treats the electron in an atom as an harmonic oscillator. The extension modifies the potential to be anharmonic - which would generate the desired nonlinear response.

The major deficiency of the classical model with respect to the quantum model is that the Lorentz model has a single resonance frequency for the system while in quantum mechanics we have infinite resonances (as we have an infinite number of eigen-energies).

1.4.1 The harmonic oscillator

We start with the original Lorentz model for which the displacement of the electron in the atom is modeled by a real quantity $x(t)$ obeying the equation of motion:

$$m \frac{d^2x}{dt^2} = -kx - 2\gamma m \frac{dx}{dt} - eE(t). \quad (1.67)$$

The forces on the right hand side of the equation are the restoring force $-kx$ with k being the spring constant, a damping force $-2\gamma m \frac{dx}{dt}$ and the force due to an external electric field $-eE(t)$.

We describe the spring constant with the resonance frequency ω_0 using: $\omega_0^2 = k/m$, with which we can rewrite the equation of motion as:

$$\frac{d^2x}{dt^2} + 2\gamma \frac{dx}{dt} + \omega_0^2 x = -\frac{e}{m} E(t). \quad (1.68)$$

If the external field is zero, we say that the oscillator is un-driven and the solution to the equation is:

$$x(t) = x(t_0)e^{i\phi_0}e^{-\gamma(t-t_0)}e^{-i\omega_0(t-t_0)} + c.c. \quad (1.69)$$

where $x(t_0)$ and ϕ_0 are real values determined through the initial conditions of the electron.

If the oscillator is driven with a CW field:

$$E(t) = Ee^{-i\omega t} + c.c. \quad (1.70)$$

then we first guess a solution of the form:

$$x(t) = x(\omega)e^{-i\omega t} + c.c. \quad (1.71)$$

where $x(\omega)$ can contain a phase (it is a complex number).

If we substitute the form of the solution and the driving field and compare all the terms that are multiplied with $e^{-i\omega t}$ we are left with the following equation:

$$-\omega^2 x(\omega) - 2i\gamma\omega x(\omega) + \omega_0^2 x(\omega) = -\frac{e}{m}E, \quad (1.72)$$

from which the solution is found to be:

$$x(\omega) = -\frac{e}{m} \frac{E}{\omega_0^2 - \omega^2 - 2i\gamma\omega} = -\frac{e}{m} \frac{E}{D(\omega)}, \quad (1.73)$$

where $D(\omega) = \omega_0^2 - \omega^2 - 2i\gamma\omega$.

The dipole moment for a single electron-atom system is simply $p = -ex(t)$. For a density of N atoms per unit volume in a given medium the macroscopic polarization is now given with

$$\begin{aligned} P(t) &= -eNx(t) = -eNx(\omega)e^{-i\omega t} + c.c. \\ &= P(\omega)e^{-i\omega t} + c.c. \end{aligned} \quad (1.74)$$

where ω is the frequency of oscillation of the driving field.

We can write:

$$P(\omega) = -eNx(\omega). \quad (1.75)$$

We remind that the linear susceptibility in the frequency domain can be written as:

$$P^{(1)}(\omega) = \varepsilon_0 \chi^{(1)}(\omega) E(\omega) \quad (1.76)$$

Comparing Eq.1.75 and Eq.1.76 we get:

$$\varepsilon_0 \chi^{(1)}(\omega) E = -eN x(\omega), \quad (1.77)$$

by rearranging terms:

$$\chi^{(1)}(\omega) = \frac{-eN x(\omega)}{\varepsilon_0 E} \quad (1.78)$$

and with Eq.1.73 we finally get:

$$\chi^{(1)}(\omega) = \frac{Ne^2}{\varepsilon_0 m (\omega_0^2 - \omega^2 - 2i\gamma\omega)} = \frac{Ne^2}{\varepsilon_0 m D(\omega)}. \quad (1.79)$$

Separating the susceptibility to real and imaginary parts:

$$\chi^{(1)}(\omega) = \frac{Ne^2(\omega_0^2 - \omega^2)}{\varepsilon_0 m [(\omega_0^2 - \omega^2)^2 + 4\gamma^2\omega^2]} + i \frac{2\gamma\omega Ne^2}{\varepsilon_0 m [(\omega_0^2 - \omega^2)^2 + 4\gamma^2\omega^2]} \quad (1.80)$$

The real and imaginary parts are shown in Fig.1.2. It can be seen that the real part, responsible for dispersion, is an asymmetric function while the imaginary part, responsible for absorption, is a symmetric Lorentzian.

We remind that the relation between the susceptibility and the index of refraction is

$$n = \sqrt{1 + \chi^{(1)}} \simeq 1 + \frac{1}{2}\chi^{(1)} = n_R + in_I \quad (1.81)$$

for which $n_R = 1 + \frac{1}{2}\Re e\{\chi^{(1)}\}$ and $n_I = \frac{1}{2}\Im m\{\chi^{(1)}\}$.

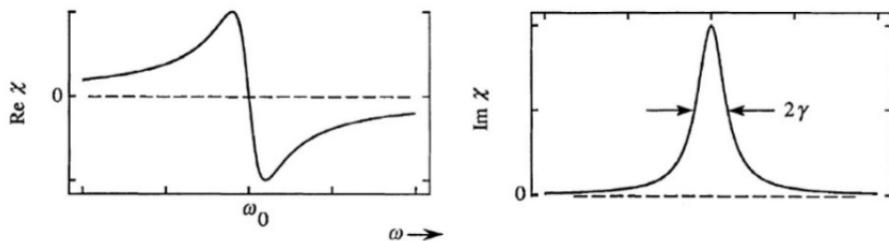


Fig. 1.2 Form of the linear susceptibility

1.4.2 The anharmonic oscillator for a non-centrosymmetric media

We now extend the classical oscillator model by adding a quadratic nonlinear term:

$$\frac{d^2x}{dt^2} + 2\gamma \frac{dx}{dt} + \omega_0^2 x + ax^2 = -\frac{e}{m} E(t). \quad (1.82)$$

The parameter a characterizes the strength of the nonlinearity. Now the total restoring force acting on the electron is:

$$F_{restoring} = -m\omega_0^2x - max^2. \quad (1.83)$$

This force is derived from the potential:

$$U(x) = - \int F_{restoring} dx = \frac{1}{2}m\omega_0^2x^2 + \frac{1}{3}max^3, \quad (1.84)$$

which is now comprised from the harmonic (parabolic) term and an anharmonic component.

Clearly this potential is non-centrosymmetric :

$$U(-x) \neq U(x). \quad (1.85)$$

The form of the potential is shown in Fig.1.3.

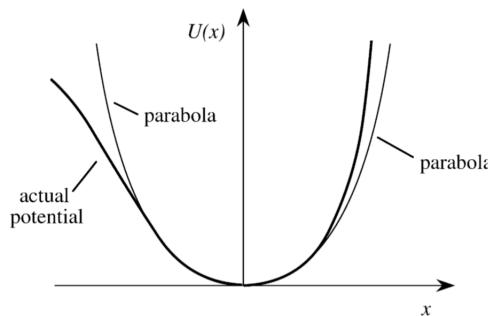


Fig. 1.3

The potential energy of a non-centrosymmetric medium. The parabola is the form of the potential energy for the harmonic oscillator.

We assume that the total field is given with:

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c. \quad (1.86)$$

We also assume that the nonlinear induced term in the equation of motion ax^2 which is driven by the field is weak - this mean that also the field is weak such that the force it exerts on the electron is weak compared with the force exerted on it by the harmonic part of the potential. As such the applied field can be considered as a small perturbation, for which we can use perturbation theory to solve the equation of motion. For this we first replace $E(t)$ with $\lambda E(t)$ where $0 \leq \lambda \leq 1$ (at the end of the procedure we would set $\lambda = 1$) characterizes the strength of the perturbation. We now seek a solution to the equation of motion which is a power series in λ :

$$x(t) = \lambda x^{(1)}(t) + \lambda^2 x^{(2)}(t) + \lambda^3 x^{(3)}(t) + \dots \quad (1.87)$$

We require that this form of the solution would be valid for any value of λ . Due to this requirement after substituting this solution we require that terms proportional to any specific power of λ satisfy the equation by themselves. That is, the terms proportional to λ should satisfy:

$$\lambda : \quad \frac{d^2x^{(1)}}{dt^2} + 2\gamma \frac{dx^{(1)}}{dt} + \omega_0^2 x^{(1)} = -\frac{e}{m} E(t). \quad (1.88)$$

Those proportional to λ^2 :

$$\lambda^2 : \quad \frac{d^2x^{(2)}}{dt^2} + 2\gamma \frac{dx^{(2)}}{dt} + \omega_0^2 x^{(2)} + a[x^{(1)}]^2 = 0, \quad (1.89)$$

Those to λ^3 :

$$\lambda^3 : \quad \frac{d^2x^{(3)}}{dt^2} + 2\gamma \frac{dx^{(3)}}{dt} + \omega_0^2 x^{(3)} + 2ax^{(1)}x^{(2)} = 0, \quad (1.90)$$

and so on.

The equation for the lowest order $x^{(1)}$ is just the regular linear equation that we analyzed in section 1.4.1. Assuming that the form of the solution (in steady state) is similar to the form of the field:

$$x^{(1)}(t) = x^{(1)}(\omega_1)e^{-i\omega_1 t} + x^{(2)}(\omega_1)e^{-i\omega_2 t} + c.c. \quad (1.91)$$

and following the procedure outlined in section 1.4.1 we easily find that:

$$x^{(1)}(\omega_j) = -\frac{e}{m} \frac{E_j}{D(\omega_j)}, \quad (1.92)$$

with $D(\omega_j) = \omega_0^2 - \omega_j^2 - 2i\gamma\omega_j$, $j = 1, 2$.

Using again the relations for the linear polarization as in the previous section:

$$P^{(1)}(\omega_j) = \varepsilon_0 \chi^{(1)}(\omega_j) E(\omega_j) = -N e x^{(1)}(\omega_j) \quad (1.93)$$

We get that the linear susceptibility is :

$$\chi^{(1)}(\omega_j) = \frac{-e N x(\omega_j)}{\varepsilon_0 E_j} = \frac{N e^2}{\varepsilon_0 m D(\omega_j)}. \quad (1.94)$$

We consider the lowest order nonlinearity in this model by substituting the result of $x^{(1)}(t)$ into Eq.1.89, where it is squared and so contains terms oscillating at frequencies $\pm 2\omega_1$, $\pm 2\omega_2$, $\pm(\omega_1 + \omega_2)$, $\pm(\omega_1 - \omega_2)$ and 0.

To continue we assume that the general solution for $x^{(2)}(t)$ contains components oscillating at the same frequencies:

$$\begin{aligned} x^{(2)}(t) = & x^{(2)}(2\omega_1)e^{-2i\omega_1 t} + x^{(2)}(2\omega_2)e^{-2i\omega_2 t} + x^{(2)}(\omega_1 + \omega_2)e^{-i(\omega_1 + \omega_2)t} \\ & + x^{(2)}(\omega_1 - \omega_2)e^{-i(\omega_1 - \omega_2)t} + x^{(2)}(0) + c.c. \end{aligned} \quad (1.95)$$

We continue by equating terms oscillating at the same frequency, from which we get the following solutions:

$$x^{(2)}(2\omega_1) = \frac{-a(e/m)^2 E_1^2}{D(2\omega_1)D^2(\omega_1)}, \quad (1.96)$$

$$x^{(2)}(2\omega_2) = \frac{-a(e/m)^2 E_2^2}{D(2\omega_2)D^2(\omega_2)}, \quad (1.97)$$

$$x^{(2)}(\omega_1 + \omega_2) = \frac{-2a(e/m)^2 E_1 E_2}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)}, \quad (1.98)$$

$$x^{(2)}(\omega_1 - \omega_2) = \frac{-2a(e/m)^2 E_1 E_2^*}{D(\omega_1 - \omega_2)D(\omega_1)D(-\omega_2)}. \quad (1.99)$$

$$x^{(2)}(0) = \frac{-2a(e/m)^2 E_1 E_1^*}{D(0)D(\omega_1)D(-\omega_1)} + \frac{-2a(e/m)^2 E_2 E_2^*}{D(0)D(\omega_2)D(-\omega_2)}. \quad (1.100)$$

The nonlinear polarization at the second harmonic of ω_1 is given with:

$$P^{(2)}(2\omega_1) = \varepsilon_0 \chi^{(2)}(2\omega_1, \omega_1, \omega_1) E^2(\omega_1). \quad (1.101)$$

On the other hand, using the oscillator model:

$$P^{(2)}(2\omega_1) = -N e x^{(2)}(2\omega_1). \quad (1.102)$$

So:

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{-N e x^{(2)}(2\omega_1)}{\varepsilon_0 E^2(\omega_1)}. \quad (1.103)$$

Using Eq.1.96 we get:

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{N(e^3/m^2)a}{\varepsilon_0 D(2\omega_1)D^2(\omega_1)} \frac{E_1^2}{E^2(\omega_1)} \quad (1.104)$$

with $E(\omega_1) = E_1$ we finally get:

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{N(e^3/m^2)a}{\varepsilon_0 D(2\omega_1)D^2(\omega_1)} \quad (1.105)$$

We can describe the nonlinear susceptibility using the expression for the linear susceptibility (Eq.1.94):

$$\chi^{(2)}(2\omega_1, \omega_1, \omega_1) = \frac{\varepsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(2\omega_1) [\chi^{(1)}(\omega_1)]^2 \quad (1.106)$$

Very similar expression is derived for $\chi^{(2)}(2\omega_2, \omega_2, \omega_2)$ by trivially replacing $\omega_1 \rightarrow \omega_2$.

Next, for SFG:

$$P^{(2)}(\omega_1 + \omega_2) = 2\varepsilon_0 \chi^{(2)}(\omega_1 + \omega_2, \omega_1, \omega_2) E(\omega_1) E(\omega_2). \quad (1.107)$$

Notice the factor of 2 above is due to the degeneracy factor (see the discussion after Eq.1.46 and section 2.5).

At the same time, from the oscillator model:

$$P^{(2)}(\omega_1 + \omega_2) = -Nex^{(2)}(\omega_1 + \omega_2). \quad (1.108)$$

Using the last two equations together with Eq.1.98 we get:

$$\chi^{(2)}(\omega_1 + \omega_2, \omega_1, \omega_2) = \frac{N(e^3/m^2)a}{\varepsilon_0 D(\omega_1 + \omega_2) D(\omega_1) D(\omega_2)}. \quad (1.109)$$

This also can be written as a product of linear susceptibilities:

$$\chi^{(2)}(\omega_1 + \omega_2, \omega_1, \omega_2) = \frac{\varepsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(\omega_1 + \omega_2) \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2). \quad (1.110)$$

In a similar manner we get the other susceptibilities:

For DFG:

$$\begin{aligned} \chi^{(2)}(\omega_1 - \omega_2, \omega_1, -\omega_2) &= \frac{N(e^3/m^2)a}{\varepsilon_0 D(\omega_1 - \omega_2) D(\omega_1) D(-\omega_2)} \\ &= \frac{\varepsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(\omega_1 - \omega_2) \chi^{(1)}(\omega_1) \chi^{(1)}(-\omega_2). \end{aligned} \quad (1.111)$$

And for optical rectification:

$$\begin{aligned} \chi^{(2)}(0, \omega_1, -\omega_1) &= \frac{N(e^3/m^2)a}{\varepsilon_0 D(0) D(\omega_1) D(-\omega_1)} \\ &= \frac{\varepsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(0) \chi^{(1)}(\omega_1) \chi^{(1)}(-\omega_1), \end{aligned} \quad (1.112)$$

and we also have a similar $\chi^{(2)}(0, \omega_2, -\omega_2)$ expression, giving together the total susceptibility for optical rectification.

Note that for all of the expression above when the involved frequencies of the fields are much lower than the resonance frequency $\omega_j \ll \omega_0$ all the $D(\omega)$ become frequency independent $D(\omega) \simeq \omega_0^2$. This means that far below the resonance the

susceptibility (both linear and nonlinear) is independent of frequency, in accordance with Kleinman's symmetry discussed in section 1.3.2.

Miller's Rule

Looking again at the expressions above where the various $\chi^{(2)}$ components are described as product of $\chi^{(1)}$ terms, we note an empirical rule known as *Miller's Rule*: the following quantity:

$$\frac{\chi^{(2)}(\omega_1 + \omega_2, \omega_1, \omega_2)}{\chi^{(1)}(\omega_1 + \omega_2)\chi^{(1)}(\omega_1)\chi^{(1)}(\omega_2)} \quad (1.113)$$

is nearly constant for most noncentrosymmetric crystals.

In fact this quantity will be constant if the combination:

$$\frac{\varepsilon_0^2 ma}{N^2 e^3} \quad (1.114)$$

is constant. But N is about the same for most condensed matter ($10^{22} cm^{-3}$), and so is the strength of the nonlinearity a . We can estimate the latter by assuming that the linear and nonlinear contributions to the restoring force:

$$F_{restoring} = -m\omega_0^2 x - max^2. \quad (1.115)$$

would become about the same when the displacement x would be about the size of the separation of the atoms in the crystal which we denote with d (in a one dimensional lattice it is the lattice constant). So:

$$m\omega_0^2 d = mad^2 \rightarrow a = \frac{\omega_0^2}{d} \quad (1.116)$$

We note that ω_0 and d are about the same for most solids, for which $\omega_0 = 10^{16} rad/s$, $d = 3 \text{ \AA}$. If we further approximate $D(\omega) \simeq \omega_0^2$ and $N \simeq 1/d^3$ we can approximate:

$$\chi^{(2)} = \frac{e^3}{\varepsilon_0 m^2 \omega_0^4 d^4} \simeq 6.9 \cdot 10^{-12} m/V = 6.9 pm/V. \quad (1.117)$$

Later on, in Section 3.3 we would come back to the classical model of the anharmonic oscillator for describing the 3rd order nonlinear susceptibility.

Second-order processes with monochromatic fields

For simplicity, in this chapter, we would ignore the tensorial nature of the susceptibility and reduce all material response to scalar form. Additionally we would consider only one dimensional problems, essentially reducing all vectors to scalars. The fields would be assumed to be polarized along the same (unspecified) direction and dependent only on the z coordinate. We also assume the material is lossless—so the susceptibilities are all real.

2.1 Coupled wave equations for three wave mixing

We consider here the case where the total field is the superposition of three CW terms:

$$E = \sum_{n=1}^3 E_n(z) e^{-i\omega_n t} = \sum_{n=1}^3 A_n(z) e^{i(k_n z - \omega_n t)} + c.c. \quad (2.1)$$

while energy conservation dictates that: $\omega_3 = \omega_1 + \omega_2$.

Each field component E_n obeys its own wave equation, which is the scalar version of Eq.1.17:

$$\frac{d^2}{dz^2} E_n(z) + \frac{\omega_n^2}{c^2} \varepsilon^{(1)}(\omega_n) E_n(z) = -\frac{\omega_n^2}{\varepsilon_0 c^2} P_n^{NL}(z). \quad (2.2)$$

Each of the components of the field can be generated in a 2nd order nonlinear interaction dependent on the two other components.

E_1 is related to the process $\omega_1 = \omega_3 - \omega_2$ (DFG) and is generated by $P_1 = 4\varepsilon_0 d_{eff} E_3 E_2^*$

E_2 is related to the process $\omega_2 = \omega_3 - \omega_1$ (DFG) and is generated by $P_2 = 4\varepsilon_0 d_{eff} E_3 E_1^*$

E_3 is related to the process $\omega_3 = \omega_1 + \omega_2$ (SFG) and is generated by $P_3 = 4\varepsilon_0 d_{eff} E_1 E_2$

Notice that for example $E_2^* = E(-\omega_2) = E^*(\omega_2)$

Above we used the same d_{eff} for all interactions, which means that we assumed

full permutation symmetry.

In SFG two photons with frequencies ω_1 and ω_2 are destroyed and a photon at frequency ω_3 is created. In an energy level diagram (See Fig.2.1) the two annihilated photons are absorbed to bring the energy of an electron in a medium from the ground state to a virtual energy level, and the decay to the ground state is made through the emission of the ω_3 photon. As the transition is to a virtual level, this process is non-resonant (it does not connect actual energy levels). Such processes occur very fast in order to comply with the uncertainty principle when taking into account the energy uncertainty associated with the transition to the virtual level.

In the DFG process in which E_1 is generated, in order to emit a photon at frequency ω_1 , a photon at frequency ω_3 is destroyed and an additional photon at frequency ω_2 is emitted (see Fig.2.2. So this process increases also the number of ω_2 photons. For this reason DFG is also known as *optical parametric amplification*. A *parametric process* is one in which the medium at the start and end of the process is at exactly the same state - it is not left at some form of an excited state. So, for a parametric process there is no transfer of energy from electromagnetic form (photons) to another form (like material vibrations for example), only to other photons.

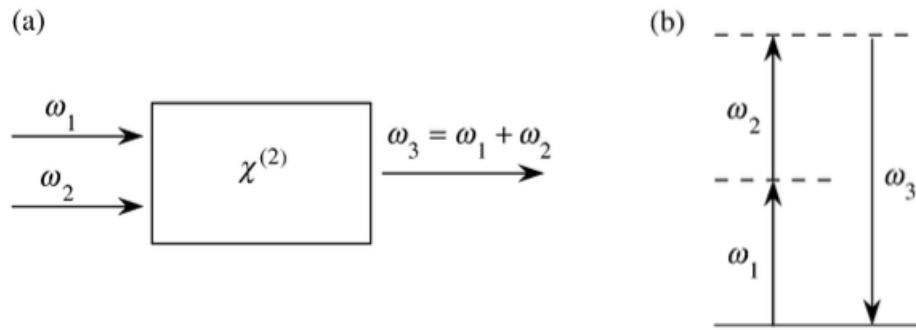


Fig. 2.1 Sum frequency generation

Now, substituting P_3 in eq.2.2 we get that the wave equation for $E_3(z) = A_3(z)e^{ik_3z}$ is:

$$\left[\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} - k_3^2 A_3 + \frac{\omega_3^2}{c^2} \varepsilon^{(1)}(\omega_3) A_3 \right] e^{ik_3 z} = -\frac{4d_{eff}\omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2)z} \quad (2.3)$$

Using $k_3^2 = \varepsilon_0^{(1)}(\omega_3)\omega_3^2/c^2$ and dividing both sides by $e^{ik_3 z}$ we get:

$$\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} = -\frac{4d_{eff}\omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2-k_3)z} \quad (2.4)$$

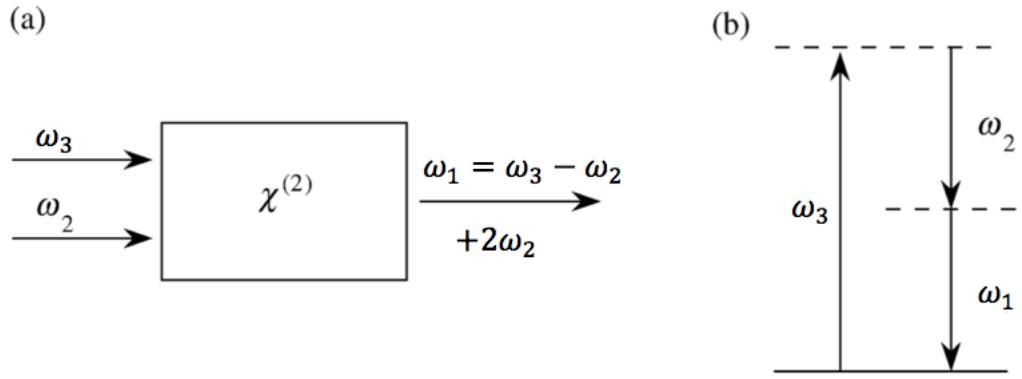


Fig. 2.2

Difference frequency generation

If the envelope A_3 is changing slowly compared to the spatial carrier frequency k_3 we can make an approximation known as the *slowly varying envelope approximation* (SVEA).

To understand this let us consider some field $A_0(z)e^{ik_0z}$. We note that we can get the rate of change of some carrier by simply using differentiation, with: $\frac{d}{dz}e^{ik_0z} = ik_0e^{ik_0z}$ we get that:

$$k_0 = \frac{\left| \frac{d}{dz}e^{ik_0z} \right|}{\left| e^{ik_0z} \right|} \quad (2.5)$$

Now, on similar grounds we can use the same operation on the envelope and require that the ensuing rate would be much smaller than the carrier rate :

$$k_0 \gg \frac{\left| \frac{d}{dz}A_0(z) \right|}{\left| A_0(z) \right|} \quad (2.6)$$

This is the first order SVEA.

Similarly we can define:

$$k_0 \gg \frac{\left| \frac{d^2}{dz^2}A_0(z) \right|}{\left| \frac{d}{dz}A_0(z) \right|} \quad (2.7)$$

as a second order SVEA. Usually in the literature the 2nd order SVEA is simply known as the SVEA.

If we apply the SVEA to Eq.2.4 we get:

$$\frac{dA_3}{dz} = \frac{2id_{eff}\omega_3^2}{k_3c^2} A_1 A_2 e^{i\Delta kz} \quad (2.8)$$

where we have introduced the *phase mismatch*:

$$\Delta k = k_1 + k_2 - k_3. \quad (2.9)$$

The phase mismatch is also known as the momentum mismatch or the wavevector mismatch. It represents the momentum imbalance of the process for every triplet of photons - the photon momenta at the end of the nonlinear process minus the photon momenta at the start of the process. When $\Delta k = 0$ momentum is completely conserved in the process and the interaction is considered to be macroscopically efficient. Otherwise the interaction is efficient only over the distance $\pi/\Delta k$. The interpretation is simple -although momentum is not exactly conserved, the process can allow for Δk uncertainty in momentum as long as the process is restricted to uncertainty of $\pi/\Delta k$ in location. We'll consider this in more detail in section 2.4.

If we repeat now the step leading to Eq.2.4 for the A_1 and A_2 amplitudes we would get:

$$\frac{dA_1}{dz} = \frac{2id_{eff}\omega_1^2}{k_1c^2} A_3 A_2^* e^{-i\Delta kz} \quad (2.10)$$

$$\frac{dA_2}{dz} = \frac{2id_{eff}\omega_2^2}{k_2c^2} A_3 A_1^* e^{-i\Delta kz} \quad (2.11)$$

Together with the equation we already wrote:

$$\frac{dA_3}{dz} = \frac{2id_{eff}\omega_3^2}{k_3c^2} A_1 A_2 e^{i\Delta kz} \quad (2.12)$$

We have the complete coupled wave equations for three-wave-mixing.

2.2 The Manley–Rowe relations

In the MKS system of units the relation between the intensity I and field $Ee^{i\omega t} + c.c.$ is:

$$I = 2n\varepsilon_0 c |E|^2, \quad (2.13)$$

where n is the index of refraction, $\varepsilon_0 = 8.85 \cdot 10^{-12} F/m$ is the vacuum permittivity and $c = 3 \cdot 10^8 m/s$ is the speed of light and the units are: $[E] = V/m$ and $[I] = W/m^2$.

The intensity associated with each of the three coupled waves Eq.2.10-2.12 is:

$I_i = 2n_i \varepsilon_0 c A_i A_i^*$ for $i = 1, 2, 3$ which we can use to find the variation in the intensity:

$$\frac{dI_i}{dz} = 2n_i \varepsilon_0 c \left(A_i^* \frac{dA_i}{dz} + A_i \frac{dA_i^*}{dz} \right) \quad (2.14)$$

Applying this equation to Eq.2.10-2.12 results in:

$$\frac{dI_1}{dz} = -8\varepsilon_0 d_{eff} \omega_1 \text{Im} (A_3 A_1^* A_2^* e^{-i\Delta kz}). \quad (2.15)$$

$$\frac{dI_2}{dz} = -8\varepsilon_0 d_{eff} \omega_2 \text{Im} (A_3 A_1^* A_2^* e^{-i\Delta kz}). \quad (2.16)$$

$$\frac{dI_3}{dz} = 8\varepsilon_0 d_{eff} \omega_3 \text{Im} (A_3 A_1^* A_2^* e^{-i\Delta kz}). \quad (2.17)$$

So adding them together:

$$\frac{dI}{dz} = \frac{dI_1}{dz} + \frac{dI_2}{dz} + \frac{dI_3}{dz} \quad (2.18)$$

$$= -8\varepsilon_0 d_{eff} (\omega_1 + \omega_2 - \omega_3) \text{Im} (A_3 A_1^* A_2^* e^{-i\Delta kz}) = 0, \quad (2.19)$$

because of energy conservation $\omega_3 = \omega_1 + \omega_2$.

This result implies that the total intensity in a lossless medium stays constant while the waves propagate and exchange energy in the medium.

The three equations Eq.2.15-2.17 also give:

$$\frac{d}{dz} \left(\frac{I_1}{\omega_1} \right) = \frac{d}{dz} \left(\frac{I_2}{\omega_2} \right) = -\frac{d}{dz} \left(\frac{I_3}{\omega_3} \right) \quad (2.20)$$

known as the *Manley-Rowe relations* relating to quantities of the form I_i/ω_i which is proportional to number of photons per unit area per unit time.

We can also write the Manley-Rowe relations in an alternative form:

$$\frac{d}{dz} \left(\frac{I_2}{\omega_2} + \frac{I_3}{\omega_3} \right) = 0, \quad \frac{d}{dz} \left(\frac{I_1}{\omega_1} + \frac{I_3}{\omega_3} \right) = 0, \quad \frac{d}{dz} \left(\frac{I_1}{\omega_1} - \frac{I_2}{\omega_2} \right) = 0. \quad (2.21)$$

which implies that the rate at which ω_1 photons are created is equal to the rate at which ω_2 photons are created and the rate at which ω_3 photons are destroyed, which is consistent with the energy conservation looked upon at the photon level: $\hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2$.

2.3 The non-depletion approximation

Under the non-depletion approximation we assume that two of the coupled fields are much stronger than the third one all along the interaction, thus they hardly lose energy and they can be taken to be constant. Let us assume that A_1 and A_2 are constant. We are left with a single equation for SFG:

$$\frac{dA_3}{dz} = \frac{2id_{eff}\omega_3^2}{k_3c^2} A_1 A_2 e^{i\Delta kz} \quad (2.22)$$

which we can now easily integrate from 0 to some distance L :

$$A_3(L) = \frac{2id_{eff}\omega_3^2 A_1 A_2}{k_3c^2} \int_0^L e^{i\Delta kz} dz = \frac{2id_{eff}\omega_3^2 A_1 A_2}{k_3c^2} \left(\frac{e^{i\Delta k L} - 1}{i\Delta k} \right). \quad (2.23)$$

Now, we can write:

$$e^{i\Delta k L} - 1 = e^{i\Delta k L/2} \left(e^{i\Delta k L/2} - e^{-i\Delta k L/2} \right) = 2ie^{i\Delta k L/2} \sin(\Delta k L/2). \quad (2.24)$$

which we can use to write:

$$A_3(L) = \kappa e^{i\Delta k L/2} L \frac{\sin \Delta k L/2}{\Delta k L/2} = \kappa e^{i\Delta k L/2} L \text{sinc} \left(\frac{\Delta k L}{2} \right) \quad (2.25)$$

where we have thrown all the coefficients into $\kappa = \frac{2id_{eff}\omega_3^2 A_1 A_2}{k_3c^2}$.

Using the expression for the intensity $I_i = 2n_i \varepsilon_0 c |A_i|^2$, $i = 1, 2, 3$ for the various fields, we have:

$$I_3 = \tilde{\kappa} L^2 \text{sinc}^2 \left(\frac{\Delta k L}{2} \right) \quad (2.26)$$

with $\tilde{\kappa} = \frac{2d_{eff}^2 \omega_3^2 I_1 I_2}{n_1 n_2 n_3 \varepsilon_0 c^3}$.

We see that for perfect phase matching $\Delta k = 0$ the intensity is growing quadratically with the distance L .

2.4 Phase Matching and Quasi-Phase-Matching

Let us consider the phase mismatch Δk in more details. The phase mismatch can be regarded as a momentum imbalance between the interacting photons. Symbolically it can be described with an arrows diagram, depicting momenta, as can be seen in Fig.2.3.

Considering again eq.2.26, but this time the effect of the phase mismatch Δk ,

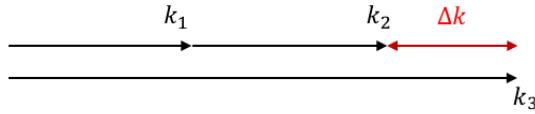


Fig. 2.3

Momentum representation of the phase mismatch for SFG.

we can see that for a given interaction length L the intensity drops very fast with growing phase mismatch. This can be appreciated with Fig.2.4 which shows the term dependent on the phase mismatch.

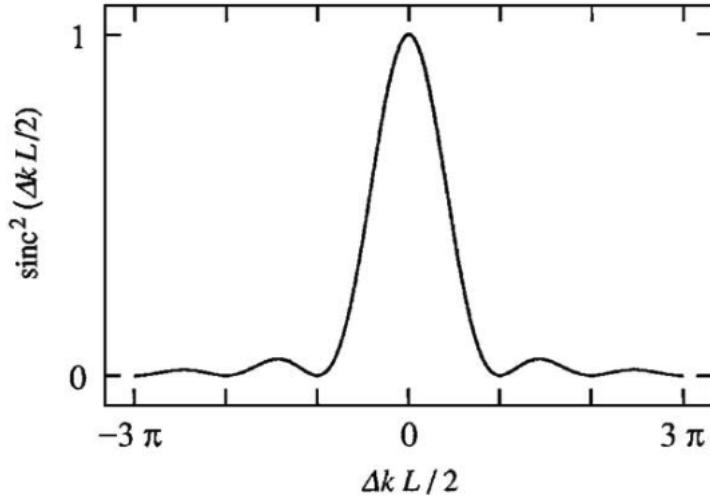


Fig. 2.4

SFG as a function of phase mismatch.

If we consider on the other hand the dependence on the coordinate z (by parametrizing $L \rightarrow z$) we should write eq.2.26 as:

$$I_3(z) = \frac{2\tilde{\kappa}}{\Delta k^2} \sin^2\left(\frac{\Delta kz}{2}\right) = \frac{\tilde{\kappa}}{\Delta k^2} (1 - \cos(\Delta kz)). \quad (2.27)$$

So the intensity is oscillating with frequency Δk , and the field envelope is oscillating with frequency $\Delta k/2$ (which is also clear from eq.2.23-2.24. The evolution of the field as a function of coordinate is shown in Fig.2.5.

There is a simple interpretation for this oscillatory behavior: the phase mismatch in the polarization indicates that the nonlinear radiation emitted at different location are having different phases. This phase accumulates as a function of distance. So two nearby locations would emit at almost the same phase, but as the distance

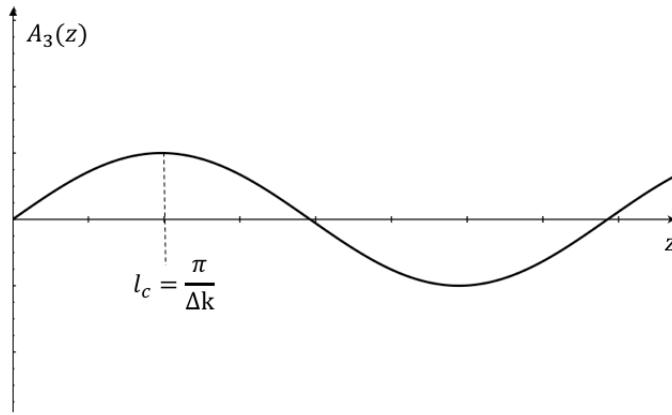


Fig. 2.5 SFG as a function of coordinate.

grows between the initial point and z we would get to a coordinate in which the emission would be exactly out of phase - there would be a π phase difference. We denote such a distance with $l_c = \pi/\Delta k$ and call it the *coherence length*. Within this length different emitters in the material are added in phase - leading to a coherent build up the total radiation. For longer distances new emitters are coherently added in a destructive interference, and so the total radiation is gradually reduced to zero, and then is starting to get build up again, and so on.

For an efficient interaction we would like to minimize as much as possible the phase mismatch. For this we first need to understand that the source for the phase mismatch in bulk crystals is material dispersion. Usually the dispersion is *normal dispersion* - the index of refraction grows with frequency (otherwise it is called *anomalous dispersion*.)

For simplicity let us consider the special case of SHG, and write down explicitly the phase mismatch. For this we'll denote the pump frequency with ω , the SH frequency with 2ω , and the corresponding wave vectors using the notation $k_\omega = k(\omega)$.

$$\Delta k = k_{2\omega} - 2k_\omega = \frac{2\omega}{c}n(2\omega) - 2\frac{\omega}{c}n(\omega) = \frac{2\omega}{c}\Delta n, \quad (2.28)$$

with $\Delta n = n(2\omega) - n(\omega)$ being the mismatch in dispersion. So, in bulk material we would like to minimize the dispersion mismatch.

2.4.1 Phase matching in birefringent crystals

In many crystals the index of refraction of a propagating beam depends on the polarization and propagation direction of the beam. This allows in principle to phase match some processes when the direction and/or polarization of the input and output beams in the process are different.

In terms of their linear optical properties crystals are classified into three different classes, based on the symmetry of their dielectric tensor (see equation 1.16) $\epsilon^{(1)}$ which can be represented by a 3X3 matrix.

Using electromagnetic theory the following can be shown:

- Materials with negligible absorption and no gain and lacking magnetic response have a real valued symmetric dielectric tensor (see Landau and Lifshitz Electrodynamics of continuous media, chapter 13 and also Yariv and Yeh, Photonics, section 1.7). Using matrix theory it can be shown that symmetric matrices can be diagonalized, with the matrix eigenvalues appearing on the diagonal.
- Materials with no absorption and gain but having a magnetic response would generally have an Hermitian dielectric tensor - that can also be diagonalized to a matrix with real values (the matrix eigenvalues) in its diagonal.
- Materials with gain or loss would have a non-Hermitian dielectric tensor - leading to complex eigenvalues whose imaginary parts correspond to gain or loss.

For cases in which the dielectric tensor can be diagonalized - the directions (coordinate basis) in which the dielectric tensor is diagonal are called the *principal axes* of the crystal. Let us denote $\epsilon_{ii}^{(1)} = \epsilon_i$ for $i = 1, 2, 3$ in this coordinate system. Then we can classify the crystals as follows:

- *Isotropic crystals.* $\epsilon_1 = \epsilon_2 = \epsilon_3$. In this case the index of refraction is the same regardless of polarization and direction of propagation.
- *Uniaxial crystals.* $\epsilon_1 = \epsilon_2 \neq \epsilon_3$. For these crystals there is a single direction, called the *optical axis*, whereas all directions perpendicular to it (or at a given angle to it) are optically equivalent. Thus rotating the material around this axis does not change its optical behavior.
- *Biaxial crystals.* ϵ_i are all different from each other.

Uniaxial crystals have reduced symmetry with respect to the isotropic crystals while they are still more symmetrical than biaxial crystals - allowing to change the index of refraction of propagating light in a simple manner. So we concentrate here on uniaxial crystals. In these crystals, for linearly polarized light: light whose polarization is perpendicular to the optic axis is governed by a refractive index $n_o = n_1 = n_2$ (for "ordinary"). Light whose polarization is in the direction of the optical axis sees an optical index $n_e = n_3$ (for "extraordinary"). See also the discussion on the index ellipsoid in section 2.10.1.

When $n_o > n_e$ the uniaxial crystal is called *negative*. When $n_o < n_e$ it is called *positive*.

Light whose polarization has some component along the optical axis has an index of refraction which is between the values of n_e and n_o . Specifically, for light whose polarization is at the same plane containing its wave-vector and the optical axis and where the angle between the optical axis and the wave vector is θ (see Fig.2.6) the index of refraction can be calculated to be (see Saleh and Teich, Fundamentals of Photonics):

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

Now, with normal dispersion $n(2\omega) > n(\omega)$, so we can try to fix this by using a negative uniaxial crystal and first assign the FH to the higher index of refraction $n_o = n_o(\omega)$, that is - setting it as the ordinary beam as depicted in Fig.2.6.

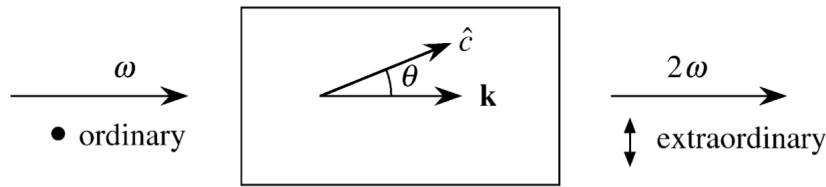


Fig. 2.6

SHG in a negative uniaxial crystal. \hat{c} is the optical axis.

Notice that we assume the two photons of the pump beam have the same polarization. This is called *Type I* interaction. If we had split the pump beam to two orthogonal polarizations then we would have called this a *Type II* interaction. This terminology is also used for SFG, relating to the polarization states of the two lower frequency terms.

Now, we can hope to match the indices of refraction of the FH and the SH by tuning the angle of propagation of the SH when it is the extraordinary beam. Formally, we write this phase matching condition as:

$$n(\theta, 2\omega) = n_o(\omega). \quad (2.30)$$

So, we need to find an angle θ that satisfies the last equation with eq.2.136 set for the SH. Combining these two conditions we get:

$$\frac{1}{n_o^2(\omega)} = \frac{\cos^2(\theta)}{n_o^2(2\omega)} + \frac{\sin^2(\theta)}{n_e^2(2\omega)}. \quad (2.31)$$

So assuming we have the information about the dispersion of the crystal and know the terms $n_o(\omega)$, $n_o(2\omega)$ and $n_e(2\omega)$ we can try to solve for the angle θ .

We can simply isolate $\sin(\theta)$ from the last equation:

$$\sin^2(\theta) = \frac{\frac{1}{n_o^2(\omega)} - \frac{1}{n_o^2(2\omega)}}{\frac{1}{n_e^2(2\omega)} - \frac{1}{n_o^2(2\omega)}} \quad (2.32)$$

The trouble with this condition is that if the birefringence is too small or the dispersion is too large the right side of the equation would be larger than 1 - and we cannot find a solution. So we cannot phase match the process.

There are further issues with birefringence phase matching that limit its performance. First, whenever the angle θ is different than 90° or 0° ¹, the direction of propagation for the FH and SH is not the same. This means that they would overlap only over a restricted length of the interaction which limits the efficiency of the interaction. This phenomenon is called *walkoff*. We note that if we are resolved to keep $\theta = 90^\circ$ we can still tune the indices of refraction by changing the temperature of the crystal. This can also accomplish phase matching in some cases, known as phase matching by *temperature tuning* as opposed to the *angle tuning* discussed above.

Second, because the FH and SH are of different polarization we cannot use for the interaction a component of the susceptibility like d_{11} or d_{33} where the polarization of all waves is the same (see section 1.3.3), but those tend to be the largest components (for example for LiNbO₃, $d_{33} = 31.5\text{pm}/V$ is the largest, compared with, say $d_{31} = -4.52\text{pm}/V$).

2.4.2 Quasi Phase Matching

Quasi phase matching (QPM) is a method in which the condition for momentum matching (phase matching) is replaced with a less strict condition, allowing the nonlinear optical process to be efficient over a long interaction distance. Basically, the requirement for momentum conservation (or matching in our terminology) is a consequence of a very specific symmetry - continuous translational symmetry. In such a system momentum needs to be conserved. We can lessen this strict condition by simply limiting the system (or interaction) in space. In this case the uncertainty in momentum would increase due to the certainty in location. The consequence is that the phase mismatch curve shown in Fig.2.4 would get wider but also lower. However, there is another way in which we can change the condition of momentum conservation - and that is by breaking the continuous translational symmetry to a discrete translational symmetry, replacing momentum conservation with a less strict condition. If we modulate a relevant parameter of the process (it can be the index of refraction or the nonlinear susceptibility) in a periodic manner with period Λ , we replace the exact requirement for momentum conservation $\Delta k = 0$ with the less strict condition of quasi-momentum conservation: $\Delta k = m \frac{2\pi}{\Lambda}$ with m being an integer. (Using solid state physics terminology - the eigenstates of the system are now Bloch modes, for which the quasi-momenta are invariant to shifts which are multiples of the length of the Brillouin zone.)

As a model for QPM we would consider modulation of the nonlinear coupling coefficient $d_{eff} \rightarrow d(z) = d_{eff} \cdot g(z)$ which is equivalent to modulating a component of the nonlinear susceptibility described with a function $g(z) \in [-1, 1]$.

To understand the effect of such a modulation let us go back to SFG under the non depletion approximation:

¹ In this case the Poynting vector is not parallel to the wave-vector, resulting in a beam moving in a different direction than its wave-vector.

$$\frac{dA_3}{dz} = \frac{2id(z)\omega_3^2}{k_3c^2} A_1 A_2 e^{i\Delta kz} \quad (2.33)$$

and, similar to before, let us integrate the equation, but from $-\infty$ to $+\infty$:

$$A_3 = \frac{2id_{eff}\omega_3^2 A_1 A_2}{k_3 c^2} \int_{-\infty}^{\infty} g(z) e^{i\Delta kz} dz = \kappa G(\Delta k), \quad (2.34)$$

where $\kappa = \frac{2id_{eff}\omega_3^2 A_1 A_2}{k_3 c^2}$ and

$$G(\Delta k) = \mathcal{F}_{\Delta k}\{g(z)\} = \int_{-\infty}^{\infty} g(z) e^{i\Delta kz} dz, \quad (2.35)$$

is simply the Fourier transform of the function $g(z)$ in the variable Δk .

Without QPM, we still restrict the interaction to the dimensions of the crystal of a given length L , for which we can write that:

$$g(z) = \text{rect}(z/L) = \begin{cases} 1 & ; |z| \leq L/2 \\ 0 & ; \text{else} \end{cases} \quad (2.36)$$

The Fourier transform of which is $L \text{sinc}(\Delta k L/2)$ and we get

$$A_3(L) = \kappa L \text{sinc}\left(\frac{\Delta k L}{2}\right) \quad (2.37)$$

which is identical to Eq.2.25 apart from a phase factor as here the integration was taken within $[-L/2, L/2]$ while before we used $[0, L]$.

Now, for QPM we consider the following modulation:

$$g(z) = \text{sign}[\cos(2\pi z/\Lambda)] \cdot \text{rect}(z/L). \quad (2.38)$$

The rect function limits the length of the interaction, while the rectangular (square) wave is the periodic modulation applied to the interaction.

Physically a modulation of the sign of the nonlinear coefficient is possible in some crystals (known as ferroelectric) by applying a one-time strong DC electric field.

It is useful to expand the periodic modulation in a Fourier series:

$$\text{sign}[\cos(2\pi z/\Lambda)] = \sum_{m=-\infty}^{\infty} G_m e^{-ik_m z} \quad (2.39)$$

where the frequencies of the expansion are $k_m = 2\pi m/\Lambda$ (with m being an integer) and it is straightforward to show that the Fourier coefficients are given with: $G_m = (2/m\pi)\sin(m\pi/2)$. We note that these coefficients vanish for even orders of m , and that the odd orders are dropping in magnitude with m : $G_1 =$

$2/\pi, G_2 = 0, G_3 = 2/3\pi, G_4 = 0, G_5 = 2/5\pi, \dots$. Additionally note that due to the 50% duty cycle - there is no DC term in the expansion $G_0 = 0$.

We can now use this expansion to find the Fourier transform of $g(z)$:

$$\begin{aligned}
 G(\Delta k) &= \mathcal{F}_{\Delta k} \left\{ \sum_{m=-\infty}^{\infty} G_m e^{-ik_m z} \right\} * \mathcal{F}_{\Delta k} \{ \text{rect}(z/L) \} \\
 &= \left(\sum_{m=-\infty}^{\infty} \int_{-\infty}^{\infty} G_m e^{-ik_m z} e^{i\Delta k z} dz \right) * L \text{sinc} \left(\frac{\Delta k L}{2} \right) \\
 &= \left(\sum_{m=-\infty}^{\infty} G_m 2\pi \delta(\Delta k - k_m) \right) * L \text{sinc} \left(\frac{\Delta k L}{2} \right) \\
 &= \sum_{m=-\infty}^{\infty} 2\pi G_m L \text{sinc} \left(\frac{(\Delta k - k_m)L}{2} \right). \tag{2.40}
 \end{aligned}$$

Here $*$ stands for convolution. This particular case of modulation for QPM is shown in Fig.2.7.

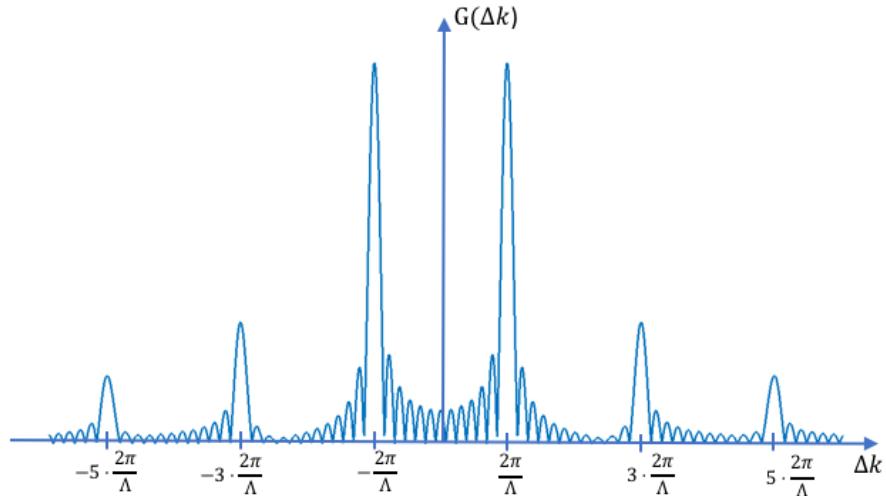


Fig. 2.7 QPM spectrum for a finite length rectangular wave modulation.

The conclusion is that the process can now be efficient provided that one of the Fourier components in the spectrum of the modulation (shown as peaks in the figure) matches the phase mismatch.

It is interesting to see how the amplitude of the field evolves for the three different cases: no phase matching, perfect phase matching and QPM. We already saw that when there is some phase mismatch the envelope of the field oscillates (Fig.2.5).

Also it is obvious that when there is perfect phase matching conditions the field amplitude would grow linearly with the coordinate. QPM would also lead to an overall growth of the field but it would be accompanied with some oscillations due to the terms that exists in the modulation and do not contribute to achieving phase matching (these are just terms that oscillate exactly like in the case of no phase matching - but each term oscillates at a different rate). All of these cases are shown in Fig.2.8.

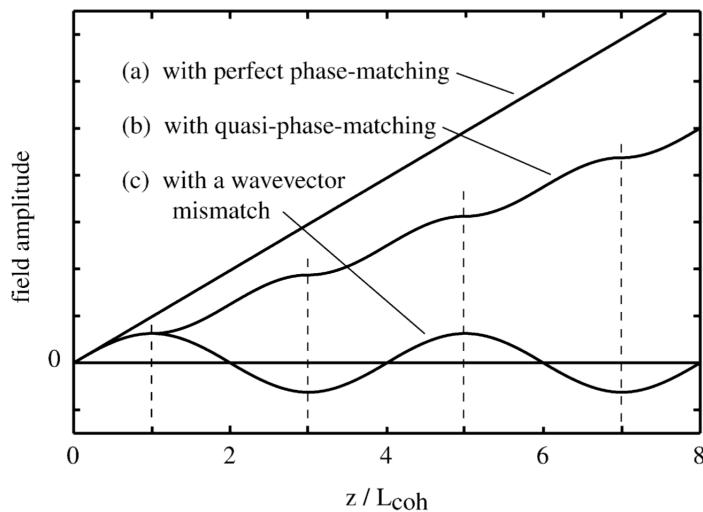


Fig. 2.8

Evolution of the field's amplitude for conditions of: phase matched interaction, not phased matched interactions and QPM. Here $L_{coh} = l_c$ is the coherence length.

2.5 Sum-Frequency Generation, strong pump and weak signal approximation

Let us return to the coupled wave equation for three-wave-mixing:

$$\frac{dA_1}{dz} = \frac{2id_{eff}\omega_1^2}{k_1 c^2} A_3 A_2^* e^{-i\Delta kz} \quad (2.41)$$

$$\frac{dA_2}{dz} = \frac{2id_{eff}\omega_2^2}{k_2 c^2} A_3 A_1^* e^{-i\Delta kz} \quad (2.42)$$

$$\frac{dA_3}{dz} = \frac{2id_{eff}\omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta kz} \quad (2.43)$$

We now assume that one of the input waves, at frequency ω_2 let's say, is very strong - it would stay undepleted during the interaction, so we can assume that A_2 is constant. The other input wave, ω_1 would be much weaker. We would consider how this weaker signal exchange its energy with the wave at frequency ω_3 which initially can be set to zero. Schematically this configuration is shown in Fig.2.9.

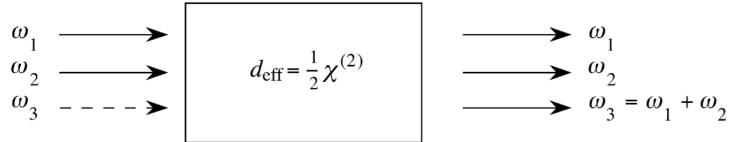


Fig. 2.9 SFG with a strong pump and a weak signal.

Under this approximation we are left with two equations for A_1 and A_3 :

$$\frac{dA_1}{dz} = \kappa_1 A_3 e^{-i\Delta kz} \quad (2.44)$$

$$\frac{dA_3}{dz} = \kappa_3 A_1 e^{i\Delta kz} \quad (2.45)$$

with $\kappa_1 = \frac{2id_{eff}\omega_1^2}{k_1 c^2} A_2^*$ and $\kappa_3 = \frac{2id_{eff}\omega_3^2}{k_3 c^2} A_2$ and we remember that $\Delta k = k_1 + k_2 - k_3$. There are two reasons to learn this specific approximation - the first one is that the situations it describes is quite common in practical scenarios of frequency conversion. The second reason is that these two coupled-mode equations are ubiquitous in physics, and they can describe many different situations which all have the same mathematical description. Examples are - a system of coupled springs, evolution of a quantum two level system, exchange of energy between two optical modes in a waveguide (modes in the same wavelength - a problem in linear optics) and so on.

The first case we consider is when there is no phase mismatch $\Delta k = 0$. To solve the coupled equations we differentiate Eq.2.44 and substitute $\frac{dA_3}{dz}$ from that equation into Eq.2.45 to get:

$$\frac{d^2 A_1}{dz^2} = -\kappa^2 A_1, \quad (2.46)$$

where $\kappa^2 = -\kappa_1 \kappa_3$. The solution to this equation is of course:

$$A_1(z) = B \cos(\kappa z) + C \sin(\kappa z). \quad (2.47)$$

Getting now back to Eq.2.44 we find that:

$$A_3(z) = -B \frac{\kappa}{\kappa_1} \sin(\kappa z) + C \frac{\kappa}{\kappa_1} \cos(\kappa z). \quad (2.48)$$

Using the boundary conditions $A_1(z = 0) = A_1(0)$ and $A_3(z = 0) = 0$ we solve for the coefficients and find that:

$$A_1(z) = A_1(0) \cos(\kappa z) \quad (2.49)$$

and

$$A_3(z) = -A_1(0) \frac{\kappa}{\kappa_1} \sin(\kappa z). \quad (2.50)$$

We notice that:

$$\left| \frac{\kappa}{\kappa_1} \right| = \sqrt{\frac{n_1 \omega_3}{n_3 \omega_1}} \simeq \sqrt{\frac{\omega_3}{\omega_1}} > 1, \quad (2.51)$$

because $\omega_3 = \omega_1 + \omega_2 > \omega_1$. This means that A_3 changes with a greater amplitude than A_1 as can be seen in Fig.2.10.

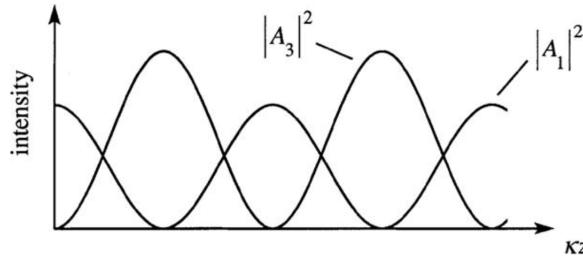


Fig. 2.10

Intensity of A_1 and A_3 for conditions of undepleted pump, weak signal and perfect phase matching.

Next, we consider the case where there is some phase mismatch $\Delta k \neq 0$.

For this case we make the following ansatz:

$$A_1(z) = (F e^{igz} + G e^{-igz}) e^{-i\Delta kz/2}, \quad (2.52)$$

$$A_3(z) = (C e^{igz} + D e^{-igz}) e^{i\Delta kz/2}, \quad (2.53)$$

We substitute these equations in Eq.2.44-2.45 and equate the coefficients of the terms $e^{\pm igz}$ to get four equations:

$$F \left(ig - \frac{1}{2} i \Delta k \right) = \kappa_1 C, \quad (2.54)$$

$$-G \left(ig + \frac{1}{2} i \Delta k \right) = \kappa_1 D, \quad (2.55)$$

$$C \left(ig + \frac{1}{2} i \Delta k \right) = \kappa_3 F, \quad (2.56)$$

$$- D \left(ig - \frac{1}{2} i \Delta k \right) = \kappa_3 G. \quad (2.57)$$

Now, if we take the two equations for the constants F and C (or we can do the same with D and G and get the same result) we can write them together in matrix form:

$$\begin{bmatrix} i(g - \frac{1}{2} \Delta k) & -\kappa_1 \\ -\kappa_3 & i(g + \frac{1}{2} \Delta k) \end{bmatrix} \begin{bmatrix} F \\ C \end{bmatrix} = 0. \quad (2.58)$$

A non trivial solution exist if the determinant of the matrix is zero, giving:

$$g^2 = -\kappa_1 \kappa_3 + \frac{1}{4} \Delta k^2. \quad (2.59)$$

We again make use of the definition: $\kappa^2 = -\kappa_1 \kappa_3$ to set the evolution rate of the amplitudes with:

$$g = \sqrt{\kappa^2 + \frac{1}{4} \Delta k^2}. \quad (2.60)$$

Notice that it does not matter if we take the negative or positive square root as our ansatz contains both terms with e^{-igz} and e^{igz} .

Next we substitute the boundary conditions $A_1(z=0) = A_1(0)$ and $A_3(z=0) = A_3(0)$ in Eq.2.52-2.53 to get:

$$A_1(0) = F + G, \quad (2.61)$$

$$A_3(0) = C + D. \quad (2.62)$$

These two together with Eq.2.54-2.55 give us four equations in the four unknowns C, D, F, G , which is easily solved so we can use them back in Eq.2.52-2.53 to get the final result:

$$A_1(z) = \left[A_1(0) \cos(gz) + \left(\frac{\kappa_1}{g} A_3(0) + \frac{i \Delta k}{2g} A_1(0) \right) \sin(gz) \right] e^{-i \Delta k z / 2}, \quad (2.63)$$

$$A_3(z) = \left[A_3(0) \cos(gz) + \left(\frac{-i \Delta k}{2g} A_3(0) + \frac{\kappa_3}{g} A_1(0) \right) \sin(gz) \right] e^{i \Delta k z / 2}. \quad (2.64)$$

To get some feeling for this result, let us consider the case in which $A_3(0) = 0$, for which:

$$A_3(z) = \frac{\kappa_3}{g} A_1(0) \sin(gz) e^{i\Delta kz/2}, \quad (2.65)$$

and the intensity is proportional to

$$I_3 \sim |A_3(z)|^2 = \frac{|\kappa_3|^2}{g^2} |A_1(0)|^2 \sin^2(gz). \quad (2.66)$$

As the phase mismatch Δk increases - also the rate g increases which leads to faster oscillations in I_3 at a lower amplitude. This can be seen in Fig.2.11.

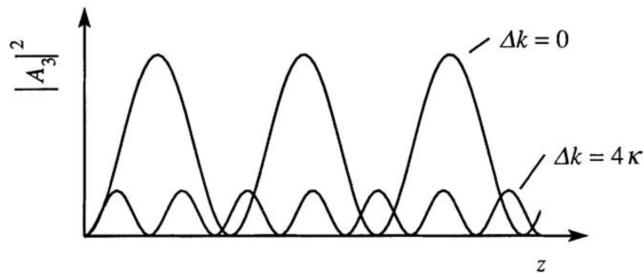


Fig. 2.11

Intensity of A_3 for different phase mismatch values for undepleted pump, weak signal conditions.

2.6 Second Harmonic Generation

A special case of SFG is SHG, which can be viewed as the prototypical optical frequency conversion process.

Let us go back to the coupled wave equations for three wave mixing describing the exchange of energy relating to the condition $\omega_3 = \omega_1 + \omega_2$:

$$\frac{dA_1}{dz} = \frac{2id_{eff}\omega_1^2}{k_1c^2} A_3 A_2^* e^{-i\Delta kz} \quad (2.67)$$

$$\frac{dA_2}{dz} = \frac{2id_{eff}\omega_2^2}{k_2c^2} A_3 A_1^* e^{-i\Delta kz} \quad (2.68)$$

$$\frac{dA_3}{dz} = \frac{2id_{eff}\omega_3^2}{k_3c^2} A_1 A_2 e^{i\Delta kz} \quad (2.69)$$

And let us consider the degenerate case in which $\omega_1 = \omega_2 \rightarrow \omega_1$ and $\omega_3 = 2\omega \rightarrow \omega_2$. For this we set $A_1 = A_2 \rightarrow A_1(\omega_1)$ and $A_3(\omega_3) \rightarrow A_2(\omega_2)$. Additionally we note that the degeneracy in which we have two photons for one of the fields in

the SHG process $\omega_2 = 2\omega_1$ breaks the symmetry in the numerical pre-factor of the right-hand-side of the equations. This is due to the fact that if we were to start from the wave equation, the form of the polarizations for each frequency component we should have used are $P_1(\omega_1, z) = 4\varepsilon_0 d_{eff} E_2 E_1^*$ and $P_2(\omega_2, z) = 2\varepsilon_0 d_{eff} E_1^2$ (see section 1.3.6 and the form of the polarizations given after Eq.2.2).

All of these give use the following form for the two coupled wave equations for SHG:

$$\frac{dA_1}{dz} = \frac{2id_{eff}\omega_1^2}{k_1 c^2} A_2 A_1^* e^{-i\Delta kz}, \quad (2.70)$$

$$\frac{dA_2}{dz} = \frac{id_{eff}\omega_2^2}{k_2 c^2} A_1^2 e^{i\Delta kz}, \quad (2.71)$$

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

We would solve the most general case of SHG (albeit, under the SVEA). For this purpose we separate the amplitudes with:

$$A_1 = \sqrt{\frac{I}{2n_1\varepsilon_0 c}} u_1 e^{i\phi_1}, \quad (2.72)$$

$$A_2 = \sqrt{\frac{I}{2n_2\varepsilon_0 c}} u_2 e^{i\phi_2}, \quad (2.73)$$

where $I = I_1 + I_2$ is the total intensity of the two waves with $I_i = 2n_i\varepsilon_0 c|A_i|^2$. We remember from the Manley-Rowe relations (section 2.2) that I is a constant of propagation. So:

$$\text{const} = I = 2n_i\varepsilon_0 c|A_1|^2 + 2n_i\varepsilon_0 c|A_2|^2 = I(u_1^2 + u_2^2). \quad (2.74)$$

Hence $u_1^2 + u_2^2 = 1$.

To proceed we define a dimensionless distance parameter:

$$\zeta = z/l, \quad (2.75)$$

with

$$l = \sqrt{\frac{2n_1^2 n_2}{\varepsilon_0 c I}} \frac{c}{2\omega_1 d_{eff}}. \quad (2.76)$$

Next we define the relative phase between the fields:

$$\theta = 2\phi_1 - \phi_2 + \Delta kz, \quad (2.77)$$

and a dimensionless phase mismatch parameter:

$$\Delta s = \Delta kl. \quad (2.78)$$

The new variables $u_i, \phi_i, \zeta, \Delta s$ can all be incorporated into the coupled wave equations Eq.2.70-2.71 to give us (after a lot of algebra) an alternative system of coupled equations but for the real quantities u_1, u_2, θ :

$$\frac{du_1}{d\zeta} = u_1 u_2 \sin(\theta), \quad (2.79)$$

$$\frac{du_2}{d\zeta} = -u_1^2 \sin(\theta), \quad (2.80)$$

$$\frac{d\theta}{d\zeta} = \Delta s + \frac{\cos(\theta)}{\sin(\theta)} \frac{d}{d\zeta} (\ln(u_1^2 u_2)). \quad (2.81)$$

For the phase matched case $\Delta k = \Delta s = 0$ Eq.2.81 can be cast in the form:

$$\frac{d}{d\zeta} \ln(u_1^2 u_2 \cos(\theta)) = 0. \quad (2.82)$$

As can be easily checked by differentiating Eq.2.82.

This naturally means that we have found another constant of propagation (in addition to $u_1^2 + u_2^2$):

$$u_1^2 u_2 \cos(\theta) = \text{const} = \Gamma. \quad (2.83)$$

In principle we can now work with these results to decouple Eq.2.79-2.81 for any boundary conditions.

We would examine here the simplest case in which at the start of the interaction one of the fields is zero, or the two fields are initially in a relative phase such that $\cos \theta = 0$. In this case: $\Gamma = 0$. As Eq.2.83 must be satisfied for all ζ and we assume that u_1 and u_2 would exchange energy between them, this forces

$$\cos(\theta) = 0 \rightarrow \sin(\theta) = \pm 1, \quad (2.84)$$

regardless of the value of ζ .

Using this result in Eq.2.79-2.80 we get:

$$\frac{du_1}{d\zeta} = \pm u_1 u_2, \quad (2.85)$$

$$\frac{du_2}{d\zeta} = \mp u_1^2. \quad (2.86)$$

Using $u_1^2 + u_2^2 = 1$ in the last equation we get:

$$\frac{du_2}{d\zeta} = \mp(1 - u_2^2). \quad (2.87)$$

The solution to this equations is:

$$u_2(\zeta) = \mp \tanh(\zeta + \zeta_0), \quad (2.88)$$

with ζ_0 a constant of integration (and $\tanh(x) = (e^{2x} - 1)/(e^{2x} + 1)$).

Let us assume that at the boundary there is no second harmonic at all: $u_1(0) = 1$, $u_2(0) = 0$. This means that $\tanh(\zeta_0) = 0$ hence $\zeta_0 = 0$. Using $u_1^2 + u_2^2 = 1$ again we find that

$$u_1(\zeta) = \operatorname{sech}(\zeta), \quad (2.89)$$

with $\operatorname{sech}(x) = 2e^x/(e^{2x} + 1)$.

The trend is: $u_1(\zeta \rightarrow \infty) = 0$ and $|u_2(\zeta \rightarrow \infty)| = 1$ - the two fields monotonously exchange their energy. This can be seen in Fig.2.12 for $u_2 = +\tanh(\zeta)$. Note that even if u_2 is not zero initially, that is, ζ_0 is not zero, still the $\tanh(\zeta + \zeta_0)$ behavior would ensure that all the radiation of the fundamental would be converted to second harmonic.

Another remark - looking at Fig.2.12 we see that the exchange of energy takes place over the length scale of l (defined in Eq.2.76).

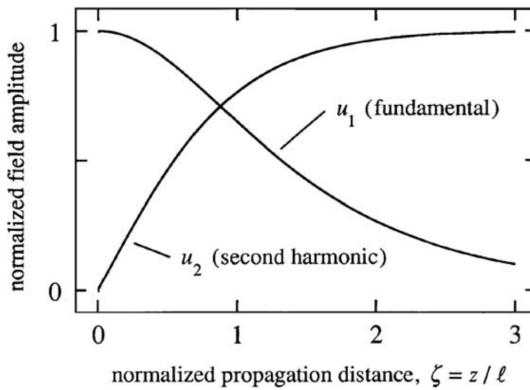


Fig. 2.12

Evolution for the fundamental and second harmonic for depleted, phase matched SHG, where initially there is no second harmonic at all.

More complicated cases of SHG can be solved numerically or analytically using Eq.2.79-2.81. We would not do so here, but we would note the general behavior for two cases. First, when $\Delta k = \Delta s = 0$ but with $\Gamma \neq 0$ (see Eq.2.83) the exchange of energy would not be monotonous but oscillatory as can be seen in Fig.2.13.

Finally, when $\Delta k \neq 0$ the overall effect is to lower the conversion efficiency as can be seen in Fig.2.14.

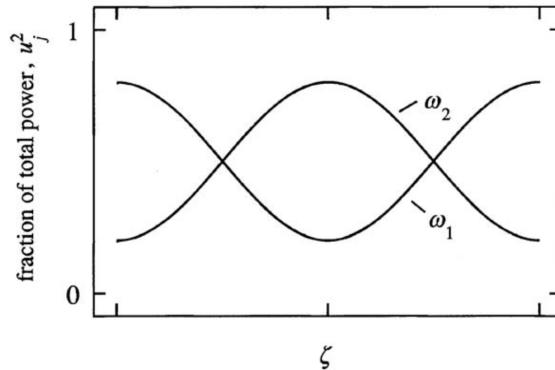


Fig. 2.13

Evolution for the fundamental and second harmonic for depleted, phase matched SHG, where initially both fields and $\cos \theta$ are not zero.

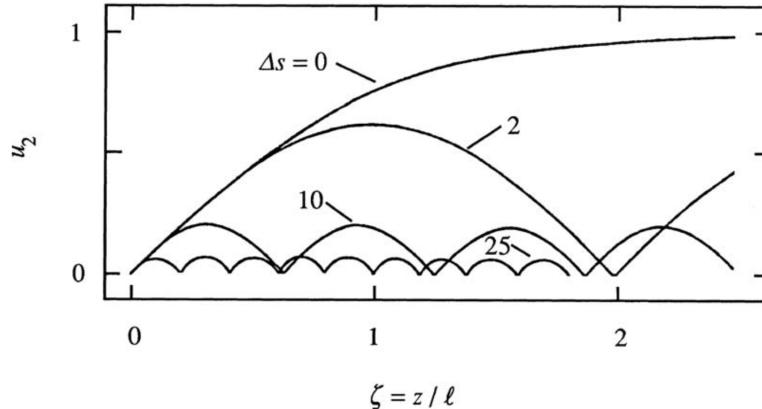


Fig. 2.14

Evolution of the second harmonic for depleted, SHG with different values for the phase mismatch and initially no second harmonic.

2.7 Difference-Frequency Generation, strong pump, weak signal approximation

Once again we return to the three coupled wave equations of three-wave-mixing, but this time we consider a case in which the wave at frequency ω_3 is strong (undepleted) and we analyze the process of DFG $\omega_2 = \omega_3 - \omega_1$ with the waves at ω_1 and ω_2 much weaker. Schematically this case is shown in Fig. 2.15.

Because now A_3 is assumed to be constant we are left with two equations:

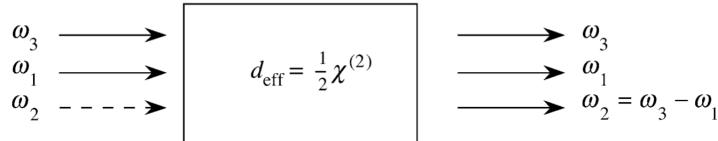


Fig. 2.15 DFG with a strong pump and a weak signal.

$$\frac{dA_1}{dz} = \kappa_1 A_2^* e^{i\Delta kz} \quad (2.90)$$

$$\frac{dA_2}{dz} = \kappa_2 A_1^* e^{i\Delta kz} \quad (2.91)$$

with $\Delta k = k_3 - k_1 - k_2$ (which is $(-\Delta k)$ from section 2.5) and with $\kappa_1 = \frac{2id_{\text{eff}}\omega_1^2}{k_1 c^2} A_3$ and $\kappa_2 = \frac{2id_{\text{eff}}\omega_2^2}{k_2 c^2} A_3$ (notice we again use our convention of putting all the constants into κ_i but they are different than the ones we used in section 2.5).

Again we start with the simple case of perfect phase matching: $\Delta k = 0$. In this case we differentiate Eq.2.91 and substitute into it $\frac{dA_1^*}{dz}$ from the conjugate of Eq.2.90 to get:

$$\frac{d^2 A_2}{dz^2} = \kappa^2 A_2, \quad (2.92)$$

with $\kappa^2 = \kappa_1 \kappa_2^*$ (which is a real number). The solution is:

$$A_2(z) = C \sinh(\kappa z) + D \cosh(\kappa z). \quad (2.93)$$

Now, we set the following boundary conditions: $A_1(z = 0) = A_1(0)$ and $A_2(z = 0) = 0$, for which the solution for the amplitudes $A_1(z)$ and $A_2(z)$ is:

$$A_1(z) = A_1(0) \cosh(\kappa z), \quad (2.94)$$

$$A_2(z) = i \sqrt{\frac{n_1 \omega_2}{n_2 \omega_1}} \frac{A_3}{|A_3|} A_1^*(0) \sinh(\kappa z). \quad (2.95)$$

These solutions are shown in Fig.2.16.

We see that both waves grows monotonically. One way to understand this results is to think on each ω_1 photon as stimulating the emission of an ω_2 photon and vice versa. This can be appreciated from Fig.2.17.

Since the ω_1 field is amplified by the parametric process of difference-frequency generation, this process is also known as *parametric amplification*.

In this language one says that the signal wave (the ω_1 wave) is amplified by the nonlinear mixing process, and an idler wave (at $\omega_2 = \omega_3 - \omega_1$) is generated by the

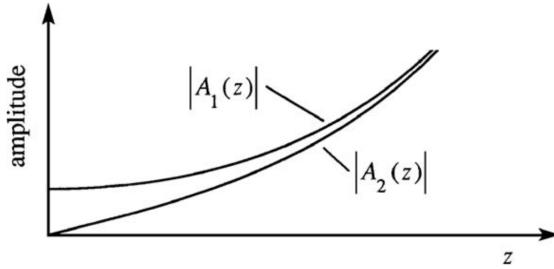


Fig. 2.16 Amplitudes for DFG with a strong pump and a weak signal.

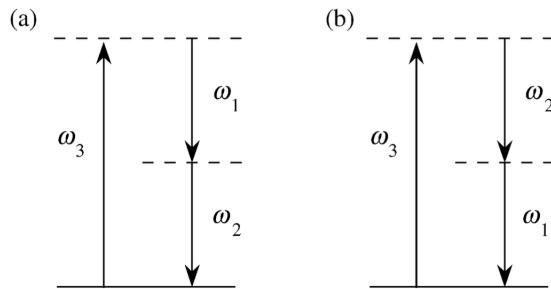


Fig. 2.17 Energy diagram for DFG. (a) ω_1 photon stimulates the emission of an ω_2 photon (b) ω_2 photon stimulates the emission of an ω_1 photon

process. We note that usually the "signal" frequency relates to the highest of the two frequencies ω_1, ω_2 .

When the phase mismatch Δk is non zero, it can be shown, using similar derivation done in section 2.5 that the solution is:

$$A_1(z) = \left[A_1(0) \left(\cosh(gz) - \frac{i\Delta k}{2g} \sinh(gz) \right) + \frac{\kappa_1}{g} A_2^*(0) \sinh(gz) \right] e^{i\Delta kz/2}, \quad (2.96)$$

$$A_2(z) = \left[A_2(0) \left(\cosh(gz) - \frac{i\Delta k}{2g} \sinh(gz) \right) + \frac{\kappa_2}{g} A_1^*(0) \sinh(gz) \right] e^{i\Delta kz/2}, \quad (2.97)$$

with:

$$g = \sqrt{\kappa^2 - \frac{1}{4}\Delta k^2}. \quad (2.98)$$

2.8 Optical Parametric Oscillators

We have seen that the process of DFG is also a process of Optical Parametric Amplification (OPA). There is gain in the process for the signal and idler waves. If there is a mechanism for gain we can construct a kind of a laser when we enclose the gain within a resonator. Indeed such a device is known as an *Optical Parametric Oscillator* (OPO). The nomenclature used for the waves in an OPO is the following: the high frequency pumping of the process is known simply as the pump with frequency ω_p (called ω_3 in the language of DFG). The two lower frequency waves which are amplified are called the signal ($\omega_s = \omega_1$) and idler ($\omega_i = \omega_2$) waves where usually $\omega_s > \omega_i$. The three frequencies obey the relation $\omega_i = \omega_p - \omega_s$ ($\omega_2 = \omega_3 - \omega_1$). The OPO is constructed when the nonlinear material is inside a cavity for either or both $\omega_i = \omega_2$ and $\omega_s = \omega_1$. The reflectivities of the mirrors at ω_1 and ω_2 are given with R_1 and R_2 respectively. A summary of these concepts is shown in Fig.2.18.

If the mirrors are highly reflective for both ω_s and ω_i the OPO is known as a *doubly resonant OPO* (DRO). If the mirrors are highly reflective for just one of these frequencies the OPO is known as a *singly resonant OPO* (SRO).

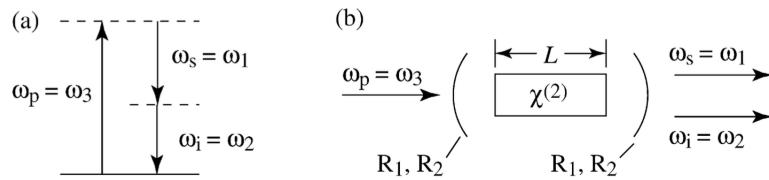


Fig. 2.18

(a) Nomenclature relations between DFG and an OPO. (b) Schematic structure for an OPO.

As with a laser - the OPO achieves "oscillation" operation (build-up of the radiation) when the gain is larger than the losses in the system. To analyze when an oscillation is achieved (also known as threshold condition) we consider first the phase matched operation ($\Delta k = 0$) of DFG, for the general case of any boundary conditions (see section 2.7):

$$A_1(z) = A_1(0) \cosh(gz) + \frac{\kappa_1}{g} A_2^*(0) \sinh(gz), \quad (2.99)$$

$$A_2(z) = A_2(0) \cosh(gz) + \frac{\kappa_2}{g} A_1^*(0) \sinh(gz), \quad (2.100)$$

while we remind that $g = \sqrt{\kappa^2 - \frac{1}{4}\Delta k^2}$ (which for the phase matched case is simply $|\kappa|$). Notice that - asymptotically ($z \rightarrow \infty$): $\cosh(gz), \sinh(gz) \rightarrow \frac{1}{2}e^{gz}$ and so g is simply the exponential gain of the OPO.

2.8.1 Self consistency condition for the OPO

Now, to find the threshold we demand *self-consistency* of the waves being amplified - that is - we would like that after a round trip in the resonator - both the signal and idler fields would be exactly the same. The self-consistency conditions *for the fields* can be written as:

$$A_1(0) = \left[A_1(0) \cosh(gL) + \frac{\kappa_1}{g} A_2^*(0) \sinh(gL) \right] R_1^2 e^{ik_1 2L}. \quad (2.101)$$

$$A_2(0) = \left[A_2(0) \cosh(gL) + \frac{\kappa_2}{g} A_1^*(0) \sinh(gL) \right] R_2^2 e^{ik_2 2L}. \quad (2.102)$$

A few important points regarding these expressions:

- It is important to notice that the pump does not circulate in the resonator, and that efficient (phase-matched) build up of the idler and signal is possible only along a single direction (from left to right in Fig.2.18.b). For the opposite direction there would be a huge phase mismatch and we can neglect the interaction for this part of the round trip. This is the reason why we have $\cosh(gL), \sinh(gL)$ and not $\cosh(2gL), \sinh(2gL)$ in the expression above.
- The R_1^2 and R_2^2 factors are the reflectivity applied twice - at each mirror, for each field.
- We would use in the following the definition for the loss per roundtrip: $l_1 = 1 - R_1^2$ and $l_2 = 1 - R_2^2$.
- The last terms $e^{ik_1 2L}$ and $e^{ik_2 2L}$ is simply the phase acquired by the fields - and we require coherent addition of each field after a round trip of length $2L$.

We can first require that:

$$k_1 2L = 2\pi p; \quad k_2 2L = 2\pi q, \quad (2.103)$$

with p, q integers - this would satisfy the requirement that the fields would be coherently added after each round trip. Then we can substitute Eq.2.102 in Eq.2.101 and after some algebra we would find the following threshold condition:

$$\cosh(gL) = 1 + \frac{l_1 l_2}{2 - l_1 - l_2} \quad (2.104)$$

Assuming the factor $gL \ll 1$ we can approximate $\cosh(gL) \simeq 1 + \frac{1}{2}(gL)^2$. Assuming also that the losses from the mirrors are small $l_1, l_2 \ll 1 \rightarrow 2 - l_1 - l_2 \simeq 2$ we get the condition:

$$(gL)^2 = l_1 l_2. \quad (2.105)$$

This is in principle the threshold condition for the DRO as we required conditions on both waves. For the SRO - we assume that the idler frequency just passes

through the mirrors (the loss within the resonator is infinite - as the wave completely escapes). This means that we can take $l_2 = 1$. Going back to Eq.2.104 we get that

$$(gL)^2 = 2l_1. \quad (2.106)$$

We would like to note two things:

- First notice that:

$$\frac{(gL)_{SRO}^2}{(gL)_{DRO}^2} = \frac{2}{l_2} \gg 1. \quad (2.107)$$

This mean that it is much easier to get to threshold with the DRO - mainly this means that the pump A_3 can be smaller. Still - most OPOs are of the SRO type because they are mush more stable as we explain below.

- Second, the threshold conditions when there is some phase mismatch (eq.2.105-2.106) is modified by replacing g^2 with $g^2 \text{sinc}^2(\Delta kL/2)$ (to see this one needs to develop the self consistency condition for non-phased matched conditions).

2.8.2 Wavelength tuning and stability of the OPO

Generally, we can choose the idler and signal frequencies to be generated by tuning the wavelengths for which $\Delta k = 0$. This can be achieved by using angle or temperature tuning as explained in section 2.4.1.

Now, the gain curve of the amplification process always has some bandwidth which typically can contain many resonator modes, as illustrated in Fig.2.19. The amplification bandwidth is estimated by assuming modes can be amplified up to phase mismatch values of $\Delta kL = \pi$. Cavity modes spacing is dependent on the length of the cavity, while the width of each mode depends on its life-time (which in turn depends on the losses in the cavity).

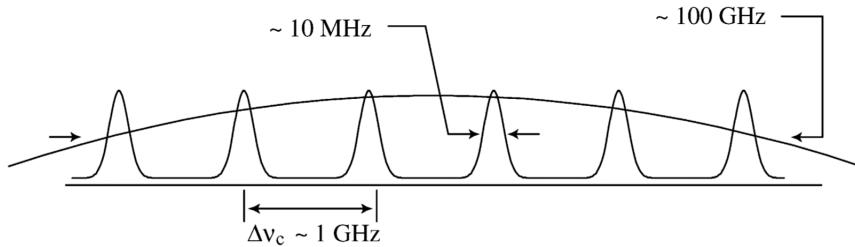


Fig. 2.19

(a) Schematic representation of an OPO amplification gain curve.

Now, we might anticipate that all the modes under the gain curve would radiate, but in reality for the SRO - only the mode with smallest phase mismatch (the modes closest to the top of the gain curve) would oscillate, as depicted in Fig.2.20(a). The reason for this is that once this mode starts to oscillate the pump power is depleted

- lowering the gain value to the threshold condition (gain would equal loss) and the gain for all other modes would be below their loss.

For the DRO the situation is different - as the consistency condition needs to be satisfied simultaneously for both the idler and signal - the respective modes need to coincide (essentially this is restatement of Eq.2.103 which is more important than minimizing the phase mismatch). This is depicted in Fig.2.20(b). This behavior prevents smooth tuning - because moving the phase matching curve can cause the modes that would oscillate to jump over several modes to a location where they are again situated most closely on the energy axis. The SRO in contrast can be tuned smoothly - from mode to adjacent mode. Additionally, some fluctuations - mechanical or temperature in the OPO - can lead to instabilities in the DRO - as its emitted spectrum can "jump" - that is - vary over a relatively large frequency range.

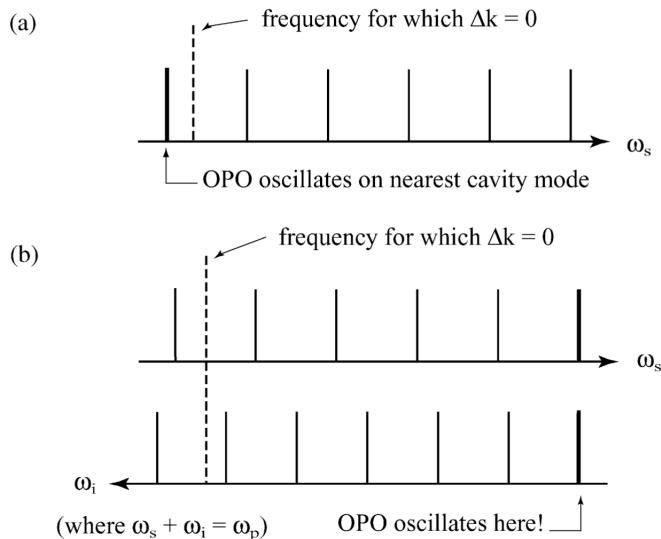


Fig. 2.20

(a) Mode structure for the SRO. (b) Mode structure for the DRO. Notice that the energy axes are situated so that $\omega_s + \omega_i = \text{const} = \omega_p$ to satisfy energy conservation.

2.9 Nonlinear optics with focused beams

Until now we assumed that the interactions were all conducted with plane waves. In this section we consider the more natural case in which the pump beam is a Gaussian. For this purpose let us first go back to the wave equation for a specific frequency mode (Eq.1.17):

$$\nabla^2 \mathbf{E}_n(\mathbf{r}) + \frac{\omega_n^2}{c^2} \boldsymbol{\varepsilon}^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) = -\frac{\omega_n^2}{\epsilon_0 c^2} \mathbf{P}_n^{NL}(\mathbf{r}). \quad (2.108)$$

We assume the modes are monochromatic and that they can be represented with:

$$\mathbf{E}_n(\mathbf{r}, t) = \mathbf{A}_n(\mathbf{r}) e^{i(k_n z - \omega_n t)} + c.c., \quad (2.109)$$

$$\mathbf{P}_n(\mathbf{r}, t) = \mathbf{p}_n(\mathbf{r}) e^{i(k'_n z - \omega_n t)} + c.c. \quad (2.110)$$

The dependence on \mathbf{r} allows the amplitudes to be position dependent in more than a single coordinate. Also note that k'_n and k_n can be different - allowing for phase mismatch.

Next, it is useful to separate the Laplacian operator into a longitudinal part dependent on z and a transverse part dependent on the other coordinates: $\nabla^2 = \frac{\partial^2}{\partial z^2} + \nabla_T^2$ where in cartesian coordinates $\nabla_T^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ and in cylindrical coordinates: $\nabla_T^2 = \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \phi^2}$ with $r^2 = x^2 + y^2$ in cylindrical coordinates.

We now substitute Eq.2.109-2.110 in Eq.2.108 and evoke the SVEA (see section 2.1) to get:

$$2ik_n \frac{\partial \mathbf{A}_n}{\partial z} + \nabla_T^2 \mathbf{A}_n = -\frac{\omega_n^2}{\epsilon_0 c^2} \mathbf{p}_n e^{i\Delta kz}, \quad (2.111)$$

with $\Delta k = k'_n - k_n$. This result is known is the *paraxial wave equation* where neglecting the second derivative $\partial^2 \mathbf{A}_n / \partial z^2$ is equivalent to cases for which the angle between the wave vector \mathbf{k}_n and the z axis is small. To understand this notice that if there is a large angle the amplitude would contain an exponent with a large spatial frequency \tilde{k} along z such that $\tilde{k} + k_n$ would equal the total spatial component along z . For small angles $\tilde{k} \simeq 0$.

2.9.1 Gaussian beams

A Gaussian beam is represented in scalar form (not writing the field polarization direction) with:

$$A(r, z) = \tilde{A} \frac{w_0}{w(z)} e^{-r^2/w^2(z)} e^{ikr^2/2R(z)} e^{i\Phi(z)}, \quad (2.112)$$

where

$$w(z) = w_0 \left[1 + \left(\frac{\lambda z}{\pi w_0^2} \right)^2 \right]^{1/2}, \quad (2.113)$$

represents the width of the beam, with w_0 being the waist of the beam (its minimum width),

$$R(z) = z \left[1 + \frac{\pi w_0^2}{\lambda z^2} \right], \quad (2.114)$$

is the radius of curvature of the beam (of its wavefront), and finally, the *Gouy phase*:

$$\Phi(z) = -\text{atan} \left(\frac{\lambda z}{\pi w_0^2} \right) = -\text{atan} \left(\frac{z}{z_R} \right) \quad (2.115)$$

is the variation in the phase of the beam compared to a plane wave. This phase has important consequences when we deal below with phase matching a frequency conversion process pumped with a Gaussian beam. Notice that $\text{atan}(x \rightarrow \pm\infty) = \pm\pi/2$ (see Fig. 2.21), so the Gouy phase sweeps over a total value of π as the beam propagates from minus infinity to plus infinity. From $z = -z_R$ to $z = z_R$ the phase sweep is $\pi/2$ (because $\text{atan}(\pm 1) = \pm\pi/4$).

z_R is known as the *Rayleigh range*:

$$z_R = \frac{\pi w_0^2}{\lambda}, \quad (2.116)$$

which is half the *confocal parameter* defined as:

$$b = \frac{2\pi w_0^2}{\lambda} = kw_0^2. \quad (2.117)$$

This parameter describes the extent of the focal area of the beam - the area of the beam whose width is smaller than $2\sqrt{2}w_0$.

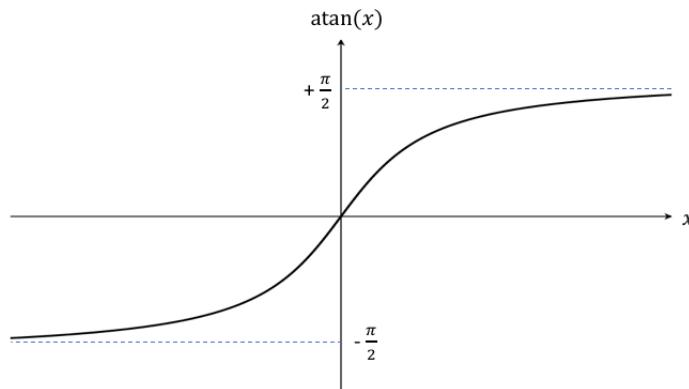


Fig. 2.21 $\text{atan}(x)$ function.

The angle of divergence of the Gaussian beam is given with

$$\Theta = \lim_{z \rightarrow \infty} \frac{w(z)}{z} = \frac{\lambda}{\pi w_0}. \quad (2.118)$$

All of these characteristics of Gaussian beams are shown graphically in Fig.2.22.

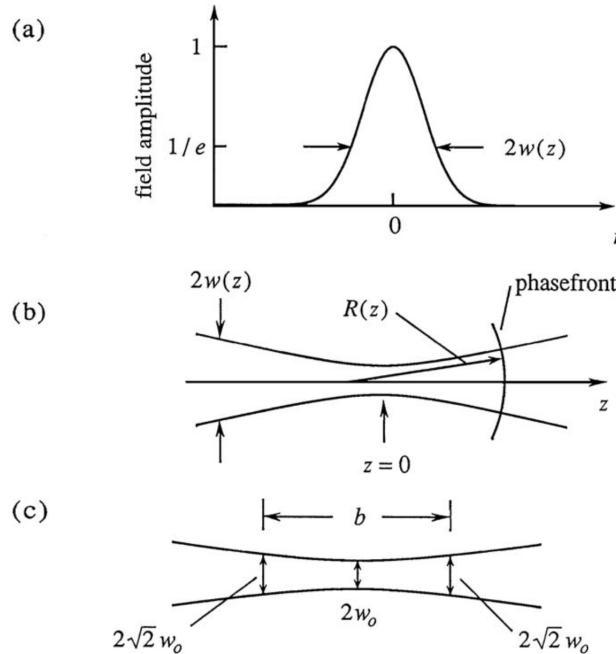


Fig. 2.22

Characteristics of a Gaussian beam. (a) Field amplitude. (b) Width $w(z)$ and curvature $R(z)$ (c) The confocal parameter $b = 2z_R$.

An equivalent, simpler mathematical representation of Gaussian beams is given with:

$$A(r, z) = \frac{\tilde{A}}{1 + i\xi} e^{-r^2/w_0^2(1+i\xi)}, \quad (2.119)$$

where $\xi = 2z/b$.

2.9.2 Optical frequency conversion using Gaussian beams in the undepleted pump approximation

For simplicity we assume that all the fields are in the same polarization so we can use a scalar form of the paraxial equation Eq.2.111. Additionally we'll use the notation $A_n = A_q$ denoting the q-th harmonic with frequency $\omega_q = q\omega$. The wave equation for the q-th harmonic generation in this case would be:

$$2ik_q \frac{\partial A_q}{\partial z} + \nabla_T^2 A_q = -\frac{\omega_q^2}{\varepsilon_0 c^2} p_q e^{i\Delta kz}, \quad (2.120)$$

where $\Delta k = qk_1 - k_q$.

We set the nonlinear polarization term to be: $p_q = \varepsilon_0 \chi^{(q)} A_1^q$ with $\chi^{(q)}$ the nonlinear susceptibility for the q -th harmonic generation and A_1 has the form of a Gaussian beam's amplitude as given in Eq.2.119 where A and \tilde{A} are replaced with A_1 and \tilde{A}_1 . We would assume non-depletion approximation for the pump beam. We also assume a trial solution where the harmonic field is in the form of a modified Gaussian beam:

$$A_q(r, z) = \frac{\tilde{A}_q(z)}{1 + i\xi} e^{-qr^2/w_0^2(1+i\xi)}. \quad (2.121)$$

Notice the modification $\tilde{A}_q = \tilde{A}_q(z)$, and also that this trial solution assumes that both beams - the pump and harmonic share the same confocal parameter (they have the same $\xi = 2z/b$). Additionally note that the waist of the q -th harmonic is \sqrt{q} times smaller than that of the pump w_0 while using Eq.2.118 we see that the divergence in the far field is of the q -th harmonic is also \sqrt{q} times smaller.

To proceed we substitute the trial solution into the wave equation Eq.2.120 but under the approximation that the variation of $\tilde{A}_q(z)$ in z is much faster than the variation in z of the exponential term $e^{-qr^2/w_0^2(1+i\xi)}$ and also much faster than the transverse variation, allowing us to approximate:

$$\begin{aligned} 2ik_q \frac{\partial A_q}{\partial z} + \nabla_T^2 A_q &\sim 2ik_q \frac{\partial A_q}{\partial z} \\ &= 2ik_q \left[\frac{\partial}{\partial z} \left(\frac{\tilde{A}_q(z)}{1 + i\xi} \right) e^{-qr^2/w_0^2(1+i\xi)} + \frac{\tilde{A}_q(z)}{1 + i\xi} \frac{\partial}{\partial z} \left(e^{-qr^2/w_0^2(1+i\xi)} \right) \right] \\ &\sim 2ik_q \frac{\partial}{\partial z} \left(\frac{\tilde{A}_q(z)}{1 + i\xi} \right) e^{-qr^2/w_0^2(1+i\xi)}. \end{aligned} \quad (2.122)$$

If we further assume that the variation in $\tilde{A}_q(z)$ are much larger than $\tilde{A}_q(z)$ itself:

$$2ik_q \frac{\partial}{\partial z} \left(\frac{\tilde{A}_q(z)}{1 + i\xi} \right) e^{-qr^2/w_0^2(1+i\xi)} \sim 2ik_q \frac{\frac{\partial \tilde{A}_q}{\partial z}(1 + i\xi)}{(1 + i\xi)^2} e^{-qr^2/w_0^2(1+i\xi)} \quad (2.123)$$

Finally using this result in Eq.2.120 we'll get:

$$\frac{d\tilde{A}_q(z)}{dz} = \frac{iq\omega}{2n_q c} \chi^{(q)} \tilde{A}_1^q \frac{e^{i\Delta kz}}{(1 + i\xi)^{q-1}}, \quad (2.124)$$

with $n_q = n(\omega_q)$ and we have used $k_q = \frac{q\omega}{c} n_q$. Integrating the above equation we get:

$$\tilde{A}_q(z) = \frac{i q \omega}{2 n_q c} \chi^{(q)} \tilde{A}_1^q J_q(\Delta k, z_0, z), \quad (2.125)$$

where

$$J_q(\Delta k, z_0, z) = \int_{z_0}^z \frac{e^{i \Delta k z'} dz'}{(1 + 2iz'/b)^{q-1}}, \quad (2.126)$$

with z_0 being the coordinate at which the nonlinear interaction starts. This integral is easy to solve for the two limits of very shallow focusing or very strong focusing. For the first case - the beam is almost a plane wave where: $b \gg |z_0|, |z|$ so:

$$J_q(\Delta k, z_0, z) = \int_{z_0}^z e^{i \Delta k z'} dz' = \frac{e^{i \Delta k z} - e^{i \Delta k z_0}}{i \Delta k}, \quad (2.127)$$

so:

$$|J_q(\Delta k, z_0, z)|^2 = L^2 \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right), \quad (2.128)$$

with $L = z - z_0$ being the total interaction length.

For very strong focusing $b \ll |z_0|, |z|$ we can extend the integration limits to $\pm\infty$ for which case the integral can be solved using contour integration in the complex plane. The result is:

$$J_q(\Delta k, z_0, z) = \begin{cases} 0 & ; \Delta k \leq 0 \\ \frac{b}{2} \frac{2\pi}{(q-2)!} \left(\frac{b \Delta k}{2}\right)^{q-2} e^{-b \Delta k / 2} & ; \Delta k > 0 \end{cases} \quad (2.129)$$

The form of this expression (normalized by the confocal parameter) for third-harmonic-generation ($q = 3$) can be seen in Fig.2.23.

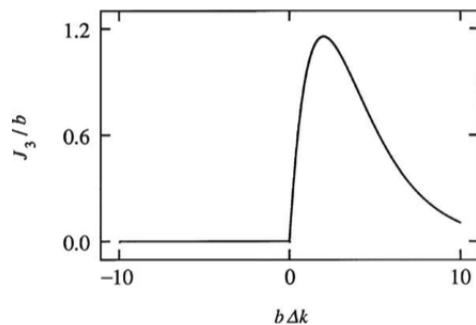


Fig. 2.23

The amplitude factor J_3 for third-harmonic-generation with a Gaussian beam as a function of the phase mismatch.

It is somewhat surprising at first glance that at exactly the phase matching condition $\Delta k = 0$ the efficiency of frequency conversion would be zero. However this is due to the nature of the spatial spectral content of a Gaussian beam which is quite different from that of a plane wave. One consequence of the nature of a Gaussian beam is the Gouy phase shift (Eq.2.115) which as we saw experience a π phase shift as the beam transverse the focus (long before the focus to long after the focus, but most of the change happens close to the focus within the Rayleigh range). The nonlinear polarization which is proportional to A_1^q experience a $q\pi$ phase shift. As a result we actually need a phase mismatch to compensate for this added accumulated phase. This is the reason that the maximum in Fig.2.23 is at a phase mismatch value which is different than zero. The reason that this value is actually positive can be appreciated using the illustration shown in Fig.2.24 showing that only positive phase mismatch can be compensated for by utilizing off-axis wave vectors of the Gaussian beam. A more careful analysis (Boyd, G.D., Kleinman, D.A., 1968. J. Appl. Phys. 39, 3597) shows that the best conditions for SHG using a Gaussian beam are when the focus is in the middle of the nonlinear crystal, for $L/b = 2.84$ and for $\Delta k \cdot L = 3.2$.

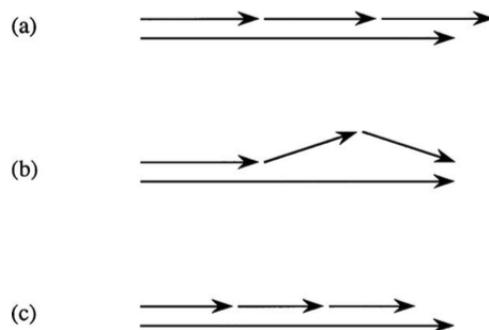


Fig. 2.24

Illustrating phase matching a frequency conversion process (third-harmonic-generation in this case) with a Gaussian beam. (a) With positive phase mismatch $\Delta k > 0$ The on axis wave pump wave vector is phase-mismatched to on-axis generation of the third harmonic. (b) However, the same process can be phase mismatched for off-axis pump wave vectors (to on-axis and off-axis wave vectors of the third harmonic) (c) For negative phase mismatch $\Delta k < 0$ there are no wave vectors in the spatial spectrum of the pump beam that can be phased matched to the third harmonic.

2.10 The electrooptic effect

The electrooptic effect is the change in an optical property of a material due to the presence of a static (DC) or very low frequency electric field. Here we consider one

type of such an effect, known as the *Pockels effect* or the *linear electrooptic effect* in which the presence of the DC electric field changes the index of refraction linearly in the field. This linear electrooptic effect is basically a three-wave-mixing process whose polarization is given with a second order nonlinear susceptibility:

$$P_i(\omega) = 2\epsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(\omega = \omega + 0) E_j(\omega) E_k(0). \quad (2.130)$$

This is a SFG process (see Eq.1.59) where one of the fields is a DC field.

As the Pockels effect is a three wave mixing process, it is restricted to non-centrosymmetric materials.

We mention that there is also a non-linear electrooptics effect (The Kerr electrooptic effect) in which the index of refraction changes quadratically in the DC field, owing to a polarization of the form:

$$P_i(\omega) = 3\epsilon_0 \sum_{jkl} \chi_{ijkl}^{(2)}(\omega = \omega + 0 + 0) E_j(\omega) E_k(0) E_l(0). \quad (2.131)$$

2.10.1 Mathematical formalism for the linear Electrooptic effect

Getting back to the Pockels effect we analyze it through its operation on the index of refraction, or dielectric permittivity which relates an electric field to the displacement field $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$ (see Section 1.1) in a general anisotropic material through:

$$D_i = \epsilon_0 \sum_j \epsilon_{ij} E_j. \quad (2.132)$$

In matrix form these three equations can be written together as:

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \epsilon_0 \begin{bmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}. \quad (2.133)$$

For lossless materials which also lack a magnetic response the dielectric tensor would be real and symmetric (see Section 2.4.1). With a magnetic response the tensor would be Hermitian. Using matrix theory, in both cases, the dielectric tensor can be diagonalized in some coordinate system X, Y, Z , called the *principal-axis system*, such that:

$$\begin{bmatrix} D_X \\ D_Y \\ D_Z \end{bmatrix} = \epsilon_0 \begin{bmatrix} \epsilon_{XX} & 0 & 0 \\ 0 & \epsilon_{YY} & 0 \\ 0 & 0 & \epsilon_{ZZ} \end{bmatrix} \begin{bmatrix} E_X \\ E_Y \\ E_Z \end{bmatrix}. \quad (2.134)$$

In the X, Y, Z coordinate system we can define a surface equation in the form of an ellipsoid, known as the *index ellipsoid* (also known as the *optical indicatrix*):

$$\frac{X^2}{\varepsilon_{XX}} + \frac{Y^2}{\varepsilon_{YY}} + \frac{Z^2}{\varepsilon_{ZZ}} = 1 \quad (2.135)$$

In any other coordinate system the ellipsoid can be transformed to an equivalent form (we assume no magnetic response, and no loss and gain, such that all coefficients are real):

$$\left(\frac{1}{n^2}\right)_1 x^2 + \left(\frac{1}{n^2}\right)_2 y^2 + \left(\frac{1}{n^2}\right)_3 z^2 + 2\left(\frac{1}{n^2}\right)_4 yz + 2\left(\frac{1}{n^2}\right)_5 xz + 2\left(\frac{1}{n^2}\right)_6 xy = 1. \quad (2.136)$$

The $\left(\frac{1}{n^2}\right)_i$ coefficients can be described in terms of the $\varepsilon_{XX}, \varepsilon_{YY}, \varepsilon_{ZZ}$ coefficients. The exact form is not very important to our discussion.

The importance of the index ellipsoid is that it can be used to describe the index of refraction experienced by a plane wave propagating with any direction and any linear polarization within the material (see Saleh and Teich, Fundamentals of Photonics.) The procedure to find these indices is as follows (see Fig.2.25):

- For a given direction of propagation $\hat{\mathbf{u}}$ we construct a plane which is perpendicular to the propagation vector and passes through the center of the index ellipsoid.
- The intersection of the plane and the ellipsoid is an ellipse.
- The semi-major and semi-minor axes of the ellipse give the values of the refractive index for the given direction of propagation. The orientation of the axes corresponds to two orthogonal polarization directions of the displacement vector \mathbf{D} vector.

We now consider how the index ellipsoid is modified due to the Pockels effect. For this it is first convenient to introduce the *impermeability tensor* whose components η_{ij} relate the displacement vector to the electric field vector:

$$E_i = \frac{1}{\varepsilon_0} \sum_j \eta_{ij} D_j. \quad (2.137)$$

This equation is the inverse of the relation given in Eq..2.132, and so $\eta_{ij} = (\varepsilon^{-1})_{ij}$

It can be shown that the index ellipsoid given in Eq.2.136 can be described using the impermeability tensor by:

$$\eta_{11}x^2 + \eta_{22}y^2 + \eta_{33}z^2 + 2\eta_{12}xy + 2\eta_{23}yz + 2\eta_{13}xz = 1, \quad (2.138)$$

where

$$\begin{aligned} \eta_{11} &= \left(\frac{1}{n^2}\right)_1, \eta_{22} = \left(\frac{1}{n^2}\right)_2, \eta_{33} = \left(\frac{1}{n^2}\right)_3, \\ \eta_{23} &= \eta_{32} = \left(\frac{1}{n^2}\right)_4, \eta_{13} = \eta_{31} = \left(\frac{1}{n^2}\right)_5, \eta_{12} = \eta_{21} = \left(\frac{1}{n^2}\right)_6. \end{aligned} \quad (2.139)$$

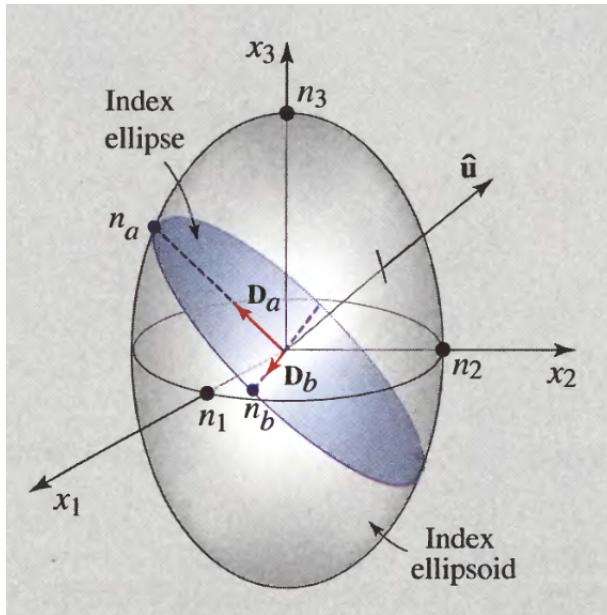


Fig. 2.25 The index ellipsoid.

We now assume that due to the electrooptic effect, the η_{ij} components can be written as a power series on the components of the electric field as:

$$\eta_{ij} = \eta_{ij}^{(0)} + \sum_k r_{ijk} E_k + \sum_{kl} s_{ijkl} E_k E_l + \dots \quad (2.140)$$

Here r_{ijk} are the components of the second order tensor describing the linear (Pockels) electrooptic effect, while s_{ijkl} are the components of the third order tensor describing the quadratic (Kerr) electrooptic effect.

Assuming we are dealing with materials with negligible absorption (or gain) and lacking magnetic response - the dielectric tensor ε_{ij} is real and symmetric ($\varepsilon_{ij} = \varepsilon_{ji}$). As a consequence also its inverse η_{ij} must be real and symmetric. This dictates that the electrooptic tensor r_{ijk} must be invariant to exchange of its first two indices. For this reason we can represent the r_{ijk} tensor using a contracted notation with only two indices r_{hk} according to:

$$\begin{array}{lllllll} ij : & 11 & 22 & 33 & 23, 32 & 31, 13 & 12, 21 \\ h : & 1 & 2 & 3 & 4 & 5 & 6 \end{array} \quad (2.141)$$

This is very similar to the contracted notation used in describing the second order nonlinear optical tensor (see section 1.3.3). Anyway, with this notation we can use Eq.2.139-2.140 to describe the modification in the $(1/n^2)_i$ terms as:

$$\Delta \left(\frac{1}{n^2} \right)_i = \sum_j r_{ij} E_j. \quad (2.142)$$

In matrix form:

$$\begin{bmatrix} \Delta \left(\frac{1}{n^2} \right)_1 \\ \Delta \left(\frac{1}{n^2} \right)_2 \\ \Delta \left(\frac{1}{n^2} \right)_3 \\ \Delta \left(\frac{1}{n^2} \right)_4 \\ \Delta \left(\frac{1}{n^2} \right)_5 \\ \Delta \left(\frac{1}{n^2} \right)_6 \end{bmatrix} = \begin{bmatrix} r_{11} & r_{12} & r_{13} \\ r_{21} & r_{22} & r_{23} \\ r_{31} & r_{32} & r_{33} \\ r_{41} & r_{42} & r_{43} \\ r_{51} & r_{52} & r_{53} \\ r_{61} & r_{62} & r_{63} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}. \quad (2.143)$$

The r_{ij} coefficients are known as the *electrooptic coefficients*.

2.10.2 Electrooptic modulators

Here we consider an actual application of the electrooptic effect - the electrooptic modulator, which can be used to modulate the amplitude, phase or the polarization of the field, depending on how it is integrated with other components and on its mode of operation.

We start with a uniaxial crystal (see Section 2.4.1) whose index ellipsoid (Eq.2.136) in its principal axis system is given with:

$$\frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2} = 1 \quad (2.144)$$

with $\varepsilon_{XX} = \varepsilon_{YY} = n_o^2$ and $\varepsilon_{ZZ} = n_e^2$.

If an electric field is applied to the crystal then the electrooptic effect dictates that the index ellipsoid would change to the form of Eq.2.138 according to Eq.2.143.

Let us chose our medium to be made of KDP, for which the electrooptic coefficients matrix is:

$$\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \quad (2.145)$$

With this matrix we get a modified index ellipsoid when an electric field is applied:

$$\frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2} + 2r_{41}E_XYZ + 2r_{41}E_YXZ + 2r_{63}EZXY = 1. \quad (2.146)$$

Let us assume that the applied field has only a Z component:

$$\frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2} + 2r_{63}E_ZXY = 1. \quad (2.147)$$

In this case we can transform the coordinate system to a new principal-axis system by using:

$$X = \frac{x - y}{\sqrt{2}}, \quad Y = \frac{x + y}{\sqrt{2}}, \quad Z = z \quad (2.148)$$

with which we get:

$$\left(\frac{1}{n_o^2} + r_{63}E_z \right) x^2 + \left(\frac{1}{n_o^2} - r_{63}E_z \right) y^2 + \frac{z^2}{n_e^2} = 1 \quad (2.149)$$

This ellipsoid can also be written as:

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_e^2} = 1 \quad (2.150)$$

Where for the realistic case of $r_{63}E_z \ll 1$:

$$n_x \simeq n_o - \frac{1}{2}n_o^3r_{63}E_z, \quad (2.151)$$

$$n_y \simeq n_o + \frac{1}{2}n_o^3r_{63}E_z. \quad (2.152)$$

And this is the effect that is being used in modulators - for light propagating in the z direction, we can induce birefringence in the xy plane by applying a voltage in the z direction. See Fig.2.26 for an illustration of this process.

The spatial phase $kz = \frac{\omega}{c}n(\omega)z$ accumulated by propagating through this device would be polarization and applied-voltage dependent. The phase difference accumulated by propagating through a length L would be:

$$\Delta\phi = (n_y - n_x)\frac{\omega}{c}L = \frac{n_o^3r_{63}E_z\omega L}{c} = \frac{n_o^3r_{63}\omega V}{c} \quad (2.153)$$

where we used Eq.2.151-2.152 and $E_z = V/L$. If we define the voltage $V_{\lambda/2}$ as the voltage required to accomplish a π phase difference (equivalent to a retardation by a half wave-length) we can write:

$$\Delta\phi = \pi\frac{V}{V_{\lambda/2}}, \quad (2.154)$$

with $V_{\lambda/2} = \pi c/(\omega n_o^3 r_{63})$. We note that typical values for $V_{\lambda/2}$ are of the order of 10kV for visible light.

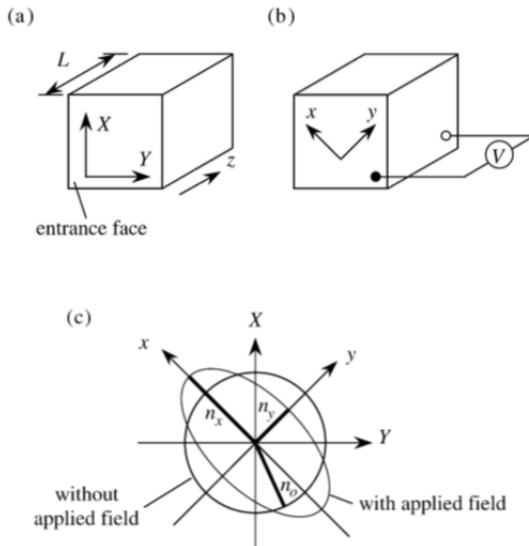


Fig. 2.26

The electrooptic effect in the uniaxial crystal KDP. (a) Principal axes when no electric field is applied. (b) The new principal axes after the application of a field in the z direction. (c) Intersection of the index ellipsoid with the plane $z = 0$ for finding the indices of refraction for light propagating in the z direction (see also Fig. 2.25).

2.10.3 Phase modulator

If the input light is linearly polarized along either the x or y axes - the field accumulates a phase which is dependent on the applied voltage, while its amplitude does not change:

$$E \rightarrow E e^{\pm i\Delta\phi/2} e^{in_o \omega L/c}, \quad (2.155)$$

Where we have the plus sign if the polarization is in the y direction.

2.10.4 Polarization modulator

To create a polarization modulator all we need is to use light initially linearly polarized in the X direction (45° with respect to the x and y directions). In this case the field can be written as:

$$\mathbf{E}_{in} = \frac{E_{in}}{\sqrt{2}} (\hat{x} + \hat{y}) e^{-i\omega t} + c.c. \quad (2.156)$$

Then applying different $\Delta\phi$ values can change the polarization state of the light, because now the field would be

$$\mathbf{E}_{out} = \frac{E_{in}}{\sqrt{2}}(\hat{x} + e^{i\Delta\phi}\hat{y})e^{-i\omega t} + c.c. \quad (2.157)$$

When $\Delta\phi = \pm\pi/2 = \pi/2, 3\pi/2$ we would have a right and left circularly polarization $\hat{x} \pm i\hat{y}$. When $\Delta\phi = \pi$ we have again linear polarization $\hat{x} - \hat{y}$ but perpendicular to the input polarization. In all other cases we have elliptical polarization.

We note that when $\Delta\phi = \pm\pi/2$ the device acts as a $\lambda/4$ wave-plate, and when $\Delta\phi = \pi$ the device acts as a $\lambda/2$ wave-plate.

2.10.5 Amplitude (intensity) modulator

We examine here a particular simple way for constructing an amplitude modulator as shown in Fig. 2.27. Here light is first set to be in the X polarization, while a perpendicular polarizer is set after the crystal. It is possible to add an optional quarter wave plate before the final polarizer. This wave plate is set with its axes matching the principal axes of the crystal. This way its operation is equal to the operation of the crystal for $\Delta\phi = \Delta\phi_B = \pi/2$ (where 'B' stands for 'bias'). If we add the phase retardance of both elements we have:

$$\Delta\phi = \pi \frac{V}{V_{\lambda/2}} + \frac{\pi}{2}. \quad (2.158)$$

This way the quarter wave plate gives us a $\Delta\phi_B = \pi/2$ bias for the overall applied retardation.

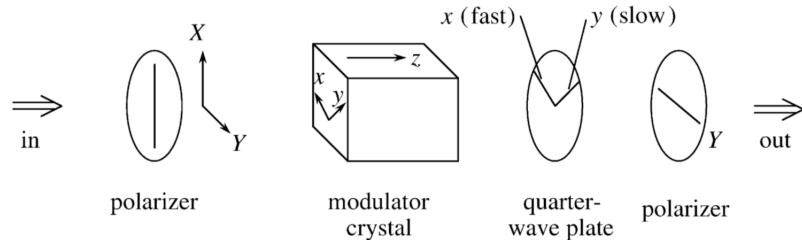


Fig. 2.27

Setup for an amplitude modulator using the electrooptic effect.

To analyze the operation of the modulator we again assume a field of the form:

$$\mathbf{E}_{in} = E_{in}\hat{\mathbf{X}}e^{-i\omega t} + c.c. = \frac{E_{in}}{\sqrt{2}}(\hat{x} + \hat{y})e^{-i\omega t} + c.c. \quad (2.159)$$

After the crystal and wave-plate the field would again be:

$$\mathbf{E} = \frac{E_{in}}{\sqrt{2}}(\hat{x} + e^{i\Delta\phi}\hat{y})e^{-i\omega t} + c.c. \quad (2.160)$$

Now, after the final polarizer we are left with the projection of the field along $\hat{\mathbf{Y}} = (-\hat{\mathbf{x}} + \hat{\mathbf{y}})/\sqrt{2}$ to give us:

$$\mathbf{E}_{out} = (\mathbf{E} \cdot \hat{\mathbf{Y}}) \hat{\mathbf{Y}} = \frac{E_{in}}{2} (-1 + e^{i\Delta\phi}) \hat{\mathbf{Y}} e^{-i\omega t} + c.c. \quad (2.161)$$

We define the transmission of the amplitude modulator as the ratio of the intensities of the spatial phasor components. For this we write the fields as $\mathbf{E}_{in} = \tilde{\mathbf{E}}_{in} e^{-i\omega t} + c.c$ and $\mathbf{E}_{out} = \tilde{\mathbf{E}}_{out} e^{-i\omega t} + c.c$ so:

$$T = \left| \frac{\tilde{\mathbf{E}}_{out}}{\tilde{\mathbf{E}}_{in}} \right|^2 = \sin^2 \Delta\phi/2 = \sin^2 \left(\frac{\pi V}{2V_{\lambda/2}} + \frac{\pi}{4} \right). \quad (2.162)$$

The form of the modulation of the transmission as a function of the retardance $\Delta\phi$ and the bias $\Delta\phi_B$ is shown in Fig. 2.28.

We see that the bias is set such that when no voltage is applied $\Delta\phi = \Delta\phi_B$ and the transmission is exactly 1/2. If the depth of modulation is being kept small by keeping the voltage small, the modulation would be approximately linear in voltage.

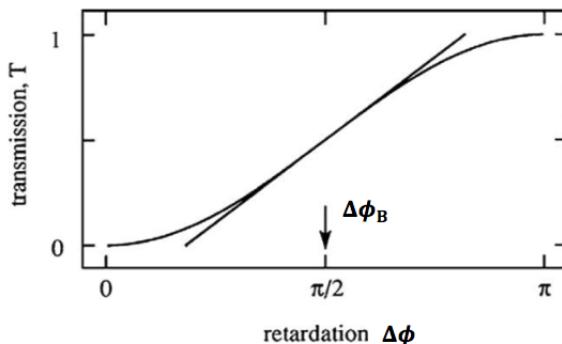


Fig. 2.28

Transmission of an electrooptic amplitude modulator.

The intensity dependent refractive index - basics

3.1 The optical Kerr effect - Introduction

In its essence, the optical Kerr effect is a Four-Wave Mixing (FWM) process in which the index of refraction of a material is a linear function of the beam intensity:

$$n = n_0 + n_2 I = n_0 + 2\bar{n}_2 |E|^2, \quad (3.1)$$

where $I = 2n_0\epsilon_0 c|E|^2$ is the beam intensity, n_0 being the index of refraction in the limit of weak fields, and n_2 (or \bar{n}_2) is the rate of change of the index of refraction in the applied intensity. The definition of \bar{n}_2 is related to n through:

$$n_2 = \frac{\bar{n}_2}{n_0\epsilon_0 c}. \quad (3.2)$$

We mention that a similar process is given with the application of a separate DC field - a process known as the Kerr electrooptic effect (see Eq.2.131). To see how the nonlinear polarization causes the change in the index of refraction we first write the nonlinear polarization term for Four-Wave Mixing:

$$P_i(\omega_o + \omega_n + \omega_m) = \epsilon_0 \sum_{jkl} \sum_{(onm)} \chi_{ijkl}^{(3)}(\omega_o + \omega_n + \omega_m; \omega_o, \omega_n, \omega_m) E_j(\omega_o) E_k(\omega_n) E_l(\omega_m). \quad (3.3)$$

The Kerr effect is related to the process $\omega = \omega + \omega - \omega$. So on the right hand side of the expression for the polarization we have the three frequencies $+\omega$, $+\omega$ and $-\omega$. Thus the number of (nm) combinations is 3 - just choosing which of the frequencies is the $-\omega$ one. For this reason we can write:

$$P_i(\omega) = 3\epsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)}(\omega = \omega + \omega - \omega) E_j(\omega) E_k(\omega) E_l(-\omega). \quad (3.4)$$

For simplicity we would consider a scalar process - all the fields are linearly polarized along the same direction. Overall, we get:

$$P^{NL}(\omega) = 3\epsilon_0 \chi^{(3)}(\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega). \quad (3.5)$$

Including the linear response of the material, the total polarization is:

$$P^{TOT}(\omega) = \varepsilon_0 \chi^{(1)} E(\omega) + 3\varepsilon_0 \chi^{(3)} (\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega) \equiv \varepsilon_0 \chi_{eff} E(\omega), \quad (3.6)$$

where the effective susceptibility is:

$$\chi_{eff} = \chi^{(1)} + 3\chi^{(3)} |E(\omega)|^2. \quad (3.7)$$

We now use the usual relation between the index of refraction and the nonlinear susceptibility but with χ_{eff} replacing the linear susceptibility when there is no nonlinearity:

$$n^2 = 1 + \chi_{eff} \quad (3.8)$$

Using Eq.3.1 and Eq.3.7 in the last expression we get:

$$n_0^2 + 4n_0 \bar{n}_2 |E(\omega)|^2 + 4\bar{n}_2^2 |E(\omega)|^4 = 1 + \chi^{(1)} + 3\chi^{(3)} |E(\omega)|^2. \quad (3.9)$$

Approximating the left hand side by neglecting the $4\bar{n}_2^2 |E(\omega)|^4$ term we find:

$$n_0 = \left(1 + \chi^{(1)}\right)^{1/2} \quad (3.10)$$

and

$$\bar{n}_2 = \frac{3\chi^{(3)}}{4n_0}. \quad (3.11)$$

In terms of n_2 we get using Eq.3.2:

$$n_2 = \frac{3}{4n_0^2 \varepsilon_0 c} \chi^{(3)}. \quad (3.12)$$

The effect we discussed here assumed that the Kerr effect is solely a self-action of a beam on itself. However, the Kerr effect can also be used in a configuration in which one beam (call it the pump beam) at frequency ω changes the index of refraction for another probe beam at frequency ω' (see Fig.3.1), also in a FWM process:

$$P^{NL}(\omega') = 6\varepsilon_0 \chi^{(3)} (\omega' = \omega' + \omega - \omega) |E(\omega)|^2 E(\omega'). \quad (3.13)$$

Notice that the degeneracy factor is now 6 as there are 6 different ways to assign the three different frequencies $+\omega$, $-\omega$ and ω' to the o, n and m components in Eq.3.15. In fact, even if $\omega = \omega'$ but there are two distinct beams such that $\mathbf{k}(\omega) \neq \mathbf{k}(\omega')$ the degeneracy factor would still be 6 (we can think about it by writing $\omega = \omega(\mathbf{k})$ and $\omega' = \omega(\mathbf{k}')$ that is - by tagging the wave frequency with different wave-vectors, so we still have the same number of permutations of ordering the different frequencies). In any case the higher degeneracy factor shows that a strong

pump beam affects the index of refraction for another (weak) beam twice as much as it does for itself:

$$\bar{n}_2^{(cross)} = 2 \cdot \frac{3\chi^{(3)}}{4n_0} = \frac{3\chi^{(3)}}{2n_0}. \quad (3.14)$$

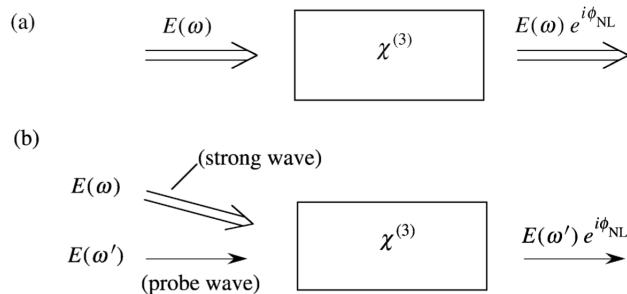


Fig. 3.1

Configuration for the optical Kerr effect. (a) Direct - where the beam changes its own index of refraction. (b) Indirect (cross) - where a strong pump beam changes the index of refraction for a weaker probe beam. In both cases ϕ_{NL} denotes the phase acquired by the Kerr effect.

3.2 Tensorial description of the susceptibility for 3rd order nonlinear response

We start with writing an explicit expression for the nonlinear polarization involving interaction of four waves - *Four-Wave Mixing* (FWM):

$$P_i(\omega_o + \omega_n + \omega_m) = \varepsilon_0 \sum_{jkl} \sum_{(onm)} \chi_{ijkl}^{(3)}(\omega_o + \omega_n + \omega_m; \omega_o, \omega_n, \omega_m) E_j(\omega_o) E_k(\omega_n) E_l(\omega_m). \quad (3.15)$$

The $\chi^{(3)}$ tensor is a fourth-rank tensor - connecting together groups of four components: three components, each of one input field, to a component of the output (generated) field. In total this tensor has $3^4 = 81$ components. Generally these components would not be independent due to material symmetries. We consider next materials with very high symmetry - isotropic materials.

3.2.1 The third-order susceptibility in isotropic materials

We consider here FWM in isotropic materials such as gases, liquids and glass.

An isotropic system posses a large degree of symmetry - that is - it is invariant

to many symmetry operations. With regard to the third order susceptibility we would see now that the isotropy dictates that for $\chi_{ijkl} \equiv \chi_{ijkl}^{(3)}$ with $i, j, k, l \in x = 1, y = 2, z = 3$ an index cannot repeat an odd number of times. That is, χ_{yxyy} for example must be zero - as it has two indices that repeat an odd number of times. To see this - we use the fact due to the isotropy the system must posses inversion symmetry for $x \rightarrow (-x)$, such that:

$$\chi_{yxyy} = \chi_{y(-x)yy}, \quad (3.16)$$

but this susceptibility term gives rise to polarization in the y direction which is not influenced by the inversion along x ¹:

$$\chi_{yxyy} E_x E_y^3 = \chi_{y(-x)yy} (-E_x) E_y^3 \quad (3.17)$$

Both equations can be satisfied only if $\chi_{yxyy} = \chi_{2122} = 0$. Applying this rule that an index cannot repeat an odd number of times leaves 21 non-zero elements out of the full 81 components.

Another symmetry that further reduces the number of independent components is that as all directions are equivalent - we can permute between the indices of any component of the tensor without changing it. We can also exchange indices while keeping orthogonality with the other indices. These can be written explicitly for the 21 non-zero components as:

$$\chi_{1111} = \chi_{2222} = \chi_{3333}, \quad (3.18)$$

$$\chi_{1122} = \chi_{1133} = \chi_{2211} = \chi_{2233} = \chi_{3311} = \chi_{3322}, \quad (3.19)$$

$$\chi_{1212} = \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232}, \quad (3.20)$$

$$\chi_{1221} = \chi_{1331} = \chi_{2112} = \chi_{2332} = \chi_{3113} = \chi_{3223}. \quad (3.21)$$

All of these components are not independent. To see this let us examine the polarization component:

$$P_x = \chi_{xxxx} E_x^3. \quad (3.22)$$

which is applicable when the input field is completely polarized in the x direction.

Now let us use a rotated coordinate system ($x', y', z' = z$) where x', y' are rotated by 45° with respect to x, y (see Fig. 3.2).

Additionally we can write: $E_{x'} = \frac{1}{\sqrt{2}}(E_x + E_y) = \frac{1}{\sqrt{2}}E_x$ and $E_{y'} = \frac{1}{\sqrt{2}}(-E_x + E_y) = -\frac{1}{\sqrt{2}}E_x$ when again the field is polarized only along x . Now, we can write:

$$\begin{aligned} P_{x'} &= \chi_{x'x'x'x'} E_{x'}^3 + \chi_{x'x'y'y'} E_{x'} E_{y'}^2 + \chi_{x'y'x'y'} E_{x'} E_{y'}^2 + \chi_{x'y'y'x'} E_{x'} E_{y'}^2 \\ &= \frac{1}{2^{3/2}} (\chi_{x'x'x'x'} + \chi_{x'x'y'y'} + \chi_{x'y'x'y'} + \chi_{x'y'y'x'}) E_x^3. \end{aligned} \quad (3.23)$$

¹ Notice that we invert the x coordinate and check the polarization along y - so we do not need to invert the polarization (it is not along x) as we did in Eq.1.41.

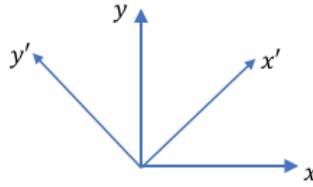


Fig. 3.2 Rotated coordinate axes.

where in the last step we used $E_{x'} = \frac{1}{\sqrt{2}}E_x$ and $E_{y'} = -\frac{1}{\sqrt{2}}E_x$.
Similarly we can write:

$$P_{y'} = \chi_{y'y'y'y'} E_{y'}^3 + \chi_{y'y'x'x'} E_{y'} E_{x'}^2 + \chi_{y'x'y'x'} E_{y'} E_{x'}^2 + \chi_{y'x'x'y'} E_{y'} E_{x'}^2 \quad (3.24)$$

$$= -\frac{1}{2^{3/2}} (\chi_{y'y'y'y'} + \chi_{y'y'x'x'} + \chi_{y'x'y'x'} + \chi_{y'x'x'y'}) E_x^3. \quad (3.25)$$

Using Eq.3.18-3.21 we notice that $P_{y'} = -P_{x'}$, so:

$$P_x = \frac{1}{\sqrt{2}}(P_{x'} - P_{y'}) = \frac{2}{\sqrt{2}}P_{x'}. \quad (3.26)$$

Now, substituting Eq.3.22 into the left hand side and Eq.3.23 into the right hand side we get:

$$\chi_{xxxx} E_x^3 = \frac{1}{2} (\chi_{xxxx} + \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}) E_x^3, \quad (3.27)$$

where we have replaced each "tag" coordinates with the regular coordinates. This is justified because the system is isotropic and so the susceptibility components are not dependent upon any specific orientations. That is: $\chi_{xxxx} = \chi_{x'x'x'x'}$ as an example, but it is true to all the components. The final result here is:

$$\chi_{xxxx} = \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}. \quad (3.28)$$

Or in terms of numerical coordinates:

$$\chi_{1111} = \chi_{1122} + \chi_{1212} + \chi_{1221}. \quad (3.29)$$

Taking this result together with Eq.3.18-3.21 we come to the conclusion that for the general case (in which all input frequencies are given arbitrary values) the susceptibility has only three independent components.

We can write all the information contained in Eq.3.18-3.21 and Eq.3.29 together in a compact form:

$$\chi_{ijkl} = \chi_{1122}\delta_{ij}\delta_{kl} + \chi_{1212}\delta_{ik}\delta_{jl} + \chi_{1221}\delta_{il}\delta_{jk} \quad (3.30)$$

Let us consider the consequence of Eq.3.29 on the optical Kerr effect - that is - on the nonlinear index of refraction. For this case we study $\chi_{ijkl}(\omega = \omega + \omega - \omega)$. Using intrinsic permutation symmetry (see section 1.3.2 in which we can permute the indices together with their associated frequencies we find that we have $\chi_{1122} = \chi_{1212}$. This is because the second and third indices are associated here with the same frequency) we can write:

$$\begin{aligned}\chi_{ijkl}(\omega = \omega + \omega - \omega) = \\ \chi_{1122}(\omega = \omega + \omega - \omega)(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) + \chi_{1221}(\omega = \omega + \omega - \omega)\delta_{il}\delta_{jk}\end{aligned}\quad (3.31)$$

We can use the last result in Eq.3.4 which we repeat here:

$$P_i(\omega) = 3\varepsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)}(\omega = \omega + \omega - \omega) E_j(\omega) E_k(\omega) E_l(-\omega), \quad (3.32)$$

to get (after some work) that:

$$P_i = 6\varepsilon_0 \chi_{1122} E_i(\mathbf{E} \cdot \mathbf{E}^*) + 3\varepsilon_0 \chi_{1221} E_i^*(\mathbf{E} \cdot \mathbf{E}), \quad (3.33)$$

where $\mathbf{E} \cdot \mathbf{E}^* = E_1 E_1^* + E_2 E_2^* + E_3 E_3^*$ and $\mathbf{E} \cdot \mathbf{E} = E_1^2 + E_2^2 + E_3^2$.

We can write Eq.3.33 together for all components in vector form:

$$\mathbf{P} = 6\varepsilon_0 \chi_{1122} (\mathbf{E} \cdot \mathbf{E}^*) \mathbf{E} + 3\varepsilon_0 \chi_{1221} (\mathbf{E} \cdot \mathbf{E}) \mathbf{E}^*. \quad (3.34)$$

If we set $A = 6\chi_{1122}$ and $B = 6\chi_{1221}$ we can write:

$$\mathbf{P} = \varepsilon_0 A (\mathbf{E} \cdot \mathbf{E}^*) \mathbf{E} + \frac{1}{2} \varepsilon_0 B (\mathbf{E} \cdot \mathbf{E}) \mathbf{E}^*. \quad (3.35)$$

The most important result here is that the nonlinear polarization vector has two contributions here - one proportional to the induced field vector \mathbf{E} and one to its conjugate \mathbf{E}^* leading to a response with opposite handedness. For example if the incoming field is right circularly polarized, we'll have two contributions - one with the same polarization and the other with left circular polarization. An analysis that we do not carry here can show that this form of the polarization causes an input field with elliptical polarization (but not linear or circular polarization) to rotate its polarization ellipse upon propagation.

3.3 The classical anharmonic oscillator - for the 3rd order nonlinear optical susceptibility

We now revisit the classical anharmonic oscillator model which we have encountered when we described the 2nd order nonlinear susceptibility in Section 1.4. Here we treat the case for a centrosymmetric media.

In a centrosymmetric media the restoring force contains a cubic nonlinearity:

$$F_{restoring} = -m\omega_0^2 x + mbx^3. \quad (3.36)$$

which is derived from the potential:

$$U(x) = - \int F_{restoring} dx = \frac{1}{2}m\omega_0^2 x^2 - \frac{1}{4}mbx^4, \quad (3.37)$$

where b sets the strength of the nonlinearity.

The form of the potential is shown in Fig.3.3. It is clear that the potential is symmetric: $U(-x) = U(x)$.

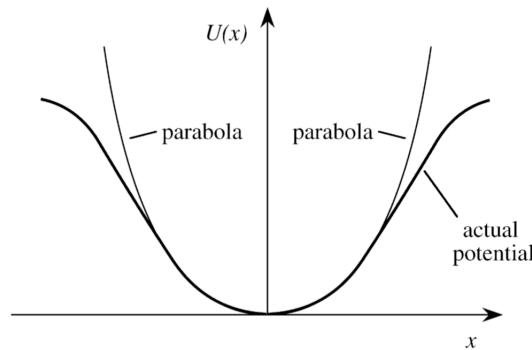


Fig. 3.3

The potential energy of a centrosymmetric medium. The parabola is the form of the potential energy for the harmonic oscillator.

In contrast with the previous case of non-centrosymmetric media, here the centrosymmetric media can be isotropic. This allows us to use a full tensorial model for the susceptibility (in the previous case a tensorial treatment would have been much more difficult as we would have needed further assumptions on the symmetry of the material, and because it cannot be isotropic, any symmetry we would have chosen would have been more complicated than the isotropic model we consider now). For this reason we extend Eq.3.36 to a vectorial force:

$$\mathbf{F}_{restoring} = -m\omega_0^2 \mathbf{r} + mb(\mathbf{r} \cdot \mathbf{r})\mathbf{r}. \quad (3.38)$$

The term $(\mathbf{r} \cdot \mathbf{r})\mathbf{r}$ is the only possible third-order displacement factor which is directed in the \mathbf{r} direction, which is required in an isotropic medium.

The electron equation of motion is:

$$\frac{d^2 \mathbf{r}}{dt^2} + 2\gamma \frac{d\mathbf{r}}{dt} + \omega_0^2 \mathbf{r} - b(\mathbf{r} \cdot \mathbf{r})\mathbf{r} = -\frac{e}{m} \mathbf{E}(t). \quad (3.39)$$

We assume the applied electric field is in the form:

$$\mathbf{E}(t) = \mathbf{E}_1 e^{-i\omega_1 t} + \mathbf{E}_2 e^{-i\omega_2 t} + \mathbf{E}_3 e^{-i\omega_3 t} + c.c. = \sum_n \mathbf{E}(\omega_n) e^{-i\omega_n t}. \quad (3.40)$$

Once again we solve the equation using perturbation theory:

$$\mathbf{r}(t) = \lambda \mathbf{r}^{(1)}(t) + \lambda^2 \mathbf{r}^{(2)}(t) + \lambda^3 \mathbf{r}^{(3)}(t) + \dots \quad (3.41)$$

We substitute this expression to the equation of motion and require that terms proportional to λ^n satisfy the equation separately. For $n = 1, 2, 3$ we have:

$$\lambda : \frac{d^2 \mathbf{r}^{(1)}}{dt^2} + 2\gamma \frac{d\mathbf{r}^{(1)}}{dt} + \omega_0^2 \mathbf{r}^{(1)} = -\frac{e}{m} \mathbf{E}(t). \quad (3.42)$$

$$\lambda^2 : \frac{d^2 \mathbf{r}^{(2)}}{dt^2} + 2\gamma \frac{d\mathbf{r}^{(2)}}{dt} + \omega_0^2 \mathbf{r}^{(2)} = 0, \quad (3.43)$$

$$\lambda^3 : \frac{d^2 \mathbf{r}^{(3)}}{dt^2} + 2\gamma \frac{d\mathbf{r}^{(3)}}{dt} + \omega_0^2 \mathbf{r}^{(3)} - b(\mathbf{r}^{(1)} \cdot \mathbf{r}^{(1)}) \mathbf{r}^{(1)} = 0 \quad (3.44)$$

The solution to the lowest order, linear equation (Eq.3.42) would be:

$$\mathbf{r}^{(1)} = \sum_n \mathbf{r}^{(1)}(\omega_n) e^{-i\omega_n t} \quad (3.45)$$

with, obviously, as in the previous sections:

$$\mathbf{r}^{(1)}(\omega_n) = \frac{-e \mathbf{E}(\omega_n)}{m D(\omega_n)}, \quad (3.46)$$

while, as usual, $D(\omega_n) = \omega_0^2 - \omega_n^2 - 2i\omega_n\gamma$. The only difference is that now the field appears as a vector.

The linear polarization term, in its vector form, is now:

$$\mathbf{P}^{(1)}(\omega_n) = -N e \mathbf{r}^{(1)}(\omega_n), \quad (3.47)$$

while, at the same time the different cartesian components of the polarization are described using:

$$P_i^{(1)} = \varepsilon_0 \sum_j \chi_{ij}^{(1)}(\omega_n) E_j(\omega_n), \quad (3.48)$$

That is - we use a tensorial description.

Because the medium is isotropic we demand that:

$$\chi_{ij}^{(1)}(\omega_n) = \chi^{(1)}(\omega_n) \delta_{ij}, \quad (3.49)$$

That is - a field in the x direction creates polarization solely in the x direction, and so on.

Using together the four last equations we get:

$$\chi^{(1)} = \frac{Ne^2}{\varepsilon_0 m D(\omega_n)}. \quad (3.50)$$

The second order response of the system - is the solution to Eq.3.43 - which is an undriven damped oscillator equation. It's solution is of the form:

$$\mathbf{r}^{(2)} = ae^{-\gamma t} \cos(\omega' t - \phi) \hat{\mathbf{r}} \quad (3.51)$$

The parameters $a, \omega', \phi, \hat{\mathbf{r}}$ are determined by the initial conditions. The important thing here is that the steady state solution is simply zero.

The third order response is derived by substituting the solution for $\mathbf{r}^{(1)}(t)$ (Eq.3.45) in the equation of motion for $\mathbf{r}^{(3)}(t)$ (Eq.3.44), which gives:

$$\frac{d^2 \mathbf{r}^{(3)}}{dt^2} + 2\gamma \frac{d\mathbf{r}^{(3)}}{dt} + \omega_0^2 \mathbf{r}^{(3)} = - \sum_{mnp} \frac{be^3 [\mathbf{E}(\omega_n) \cdot \mathbf{E}(\omega_m)] \mathbf{E}(\omega_p)}{m^3 D(\omega_m) D(\omega_n) D(\omega_p)} e^{-i(\omega_m + \omega_n + \omega_p)t}. \quad (3.52)$$

On the right hand side there are many combinations of the three frequencies that we denote with $\omega_q = \omega_m + \omega_n + \omega_p$. The solution to the equation is thus in the form:

$$\mathbf{r}^{(3)}(t) = \sum_q \mathbf{r}^{(3)}(\omega_q) e^{-i\omega_q t}. \quad (3.53)$$

Substituting back into Eq.3.52 we get a frequency domain equation:

$$(-\omega_q^2 - i\omega_q 2\gamma + \omega_0^2) \mathbf{r}^{(3)}(\omega_q) = - \sum_{(mnp)} \frac{be^3 [\mathbf{E}(\omega_n) \cdot \mathbf{E}(\omega_m)] \mathbf{E}(\omega_p)}{m^3 D(\omega_m) D(\omega_n) D(\omega_p)}. \quad (3.54)$$

The notation (mnp) indicates that now we must restrict the frequencies such that $\omega_q = \omega_m + \omega_n + \omega_p$. We can observe that on the left hand side $\mathbf{r}^{(3)}(\omega_q)$ is multiplied by $D(\omega_q)$, so we can write:

$$\mathbf{r}^{(3)}(\omega_q) = - \sum_{(mnp)} \frac{be^3 [\mathbf{E}(\omega_n) \cdot \mathbf{E}(\omega_m)] \mathbf{E}(\omega_p)}{m^3 D(\omega_q) D(\omega_m) D(\omega_n) D(\omega_p)}. \quad (3.55)$$

The third-order polarization vector is now given with:

$$\mathbf{P}^{(3)}(\omega_q) = -Ner^{(3)}(\omega_q). \quad (3.56)$$

We would like to connect the last expression with a general expression for the 3rd order nonlinear polarization containing explicitly the 3rd order susceptibility (see Eq.3.15):

$$P_i(\omega_q) = \varepsilon_0 \sum_{jkl} \sum_{(mnp)} \chi_{ijkl}^{(3)}(\omega_q = \omega_m + \omega_n + \omega_p; \omega_m, \omega_n, \omega_p) E_j(\omega_m) E_k(\omega_n) E_l(\omega_p). \quad (3.57)$$

Comparing Eq.3.55 and Eq.3.57 we can see that there are several constraints and symmetries operating here. First, the vector direction of $\mathbf{r}^{(3)}$ is the same as the direction of the field $\mathbf{E}(\omega_p)$, this indicates that the cartesian direction components i and l in Eq.3.57 must be the same - so we'll have a δ_{il} in the expression for the susceptibility. Next, the $E_j(\omega_m)E_k(\omega_n)$ component must correspond to $\mathbf{E}(\omega_n) \cdot \mathbf{E}(\omega_m)$. As we have here a dot product the two components must be in the same direction - otherwise we would get zero. So we must have a δ_{jk} as well, together giving us $\delta_{il}\delta_{jk}$. But actually the order of the j and k components does not matter because the dot product is insensitive to the order of the terms. So we actually should have $\delta_{il}(\delta_{jk} + \delta_{kj}) = 2\delta_{il}\delta_{jk}$. But now we can also use the intrinsic permutation symmetry - we chose to associate $E_l(\omega_p)$ with $\mathbf{E}(\omega_p)$, but actually we can switch indices as long as we carry the frequencies with them. This means we should also have the terms $2\delta_{ik}\delta_{jl}$ and $2\delta_{ij}\delta_{kl}$. Over all then we have them six components: $2\delta_{il}\delta_{jk} + 2\delta_{ik}\delta_{jl} + 2\delta_{ij}\delta_{kl}$. Because of that we define the susceptibility to be the sixth of the sum of six expressions giving us:

$$\chi_{ijkl}^{(3)}(\omega_q; \omega_m, \omega_n, \omega_p) = \frac{Nbe^4[\delta_{il}\delta_{jk} + \delta_{ik}\delta_{jl} + \delta_{ij}\delta_{kl}]}{3\varepsilon_0 m^3 D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_p)}. \quad (3.58)$$

This last expression can also be written as a product of linear susceptibilities using Eq.3.50:

$$\begin{aligned} \chi_{ijkl}^{(3)}(\omega_q; \omega_m, \omega_n, \omega_p) &= \frac{bm\varepsilon_0^3}{3N^3 e^4} [\chi^{(1)}(\omega_q)\chi^{(1)}(\omega_m)\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_p)] \\ &\cdot [\delta_{il}\delta_{jk} + \delta_{ik}\delta_{jl} + \delta_{ij}\delta_{kl}]. \end{aligned} \quad (3.59)$$

To estimate the strength of the nonlinearity b we assume that the linear and nonlinear contributions to the restoring force (Eq.3.36) would become about the same when the displacement x would become comparable to the separation of the atoms in the crystal which we denote with d , that is when:

$$m\omega_0^2 d = mbd^3 \rightarrow b = \frac{\omega_0^2}{d^2} \quad (3.60)$$

Estimating $d = 3\text{\AA}$, $\omega_0 = 7 \cdot 10^{15} \text{ rad/s}$ and approximating $N \simeq 1/d^3$ and $D(\omega) \simeq \omega_0^2$ we get:

$$\chi^{(3)} \simeq \frac{Nbe^4}{\varepsilon_0 m^3 \omega_0^8} = \frac{e^4}{\varepsilon_0 m^3 \omega_0^6 d^5} \simeq 344 pm^2/V^2 \quad (3.61)$$

4.1 Self-focusing and self-trapping

With self-focusing the light changes the properties of the material such it causes itself to focus. This is a process driven by the intensity-dependent refractive index. Specifically we assume that the nonlinear index n_2 (see Eq.3.1) is positive. We also assume that the beam entering the nonlinear medium has a Gaussian intensity profile - so at the middle the change in the index of refraction is stronger and it drops radially from the center. The overall effect is of a focusing lens.

A simple model of self-focusing assumes a beam with waist w_0 and on-axis intensity I_0 entering a nonlinear crystal with a positive n_2 (see Fig.4.1). In this model we neglect diffraction and use the Fermat's principle to determine the non-linear induced self-focusing distance z_{sf} from the input face of the crystal. Fermat's principle states that the optical path length $\int n(\mathbf{r})dl$ of all rays from the wavefront at the entrance to the crystal to the focus must be equal. So we equate the on-axis optical path with the optical path for the marginal ray of the beam:

$$(n_0 + n_2 I)z_{sf} = n_0 \frac{z_{sf}}{\cos \theta_{sf}}, \quad (4.1)$$

where θ_{sf} is the half angle between the focus and the beam waist at the entrance to the crystal.

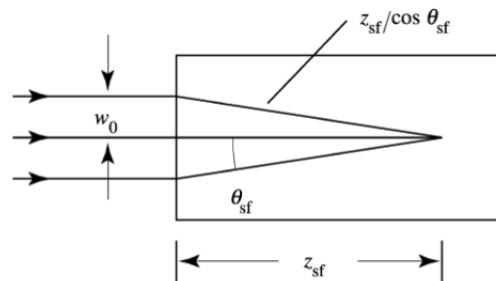


Fig. 4.1

A simple model for self-focusing.

Using a small angle approximation: $[\cos \theta_{sf}]^{-1} = 1 + \frac{1}{2}\theta_{sf}^2$ in the above equation we find the *self-focusing angle*:

$$\theta_{sf} = \sqrt{\frac{2n_2 I}{n_0}}. \quad (4.2)$$

Now we can also find that:

$$z_{sf} = \frac{w_0}{\theta_{sf}} = w_0 \sqrt{\frac{n_0}{2n_2 I}}. \quad (4.3)$$

If we also consider the effects of diffraction we first remind that the diffraction angle of a Gaussian beam with waist w_0 is given with (see Eq.2.118)

$$\theta_{diff} = \frac{\lambda_0}{\pi n_0 w_0}, \quad (4.4)$$

where we have modified the medium to have an index of refraction n_0 when there is no nonlinearity.

Now we can ask the following question - at what intensity would the self focusing would exactly balance diffraction? That is - when does the intensity satisfies the condition:

$$\theta_{diff} = \theta_{sf}. \quad (4.5)$$

Substituting the expressions for the angles we get:

$$I_{st} = \frac{\lambda_0^2}{2\pi^2 n_0 n_2 w_0^2}. \quad (4.6)$$

Here the "st" subscript stands for self-trapping.

The power of such a beam is the intensity of the beam times its area which we estimate as πw_0^2 , so:

$$P_{st} = \frac{\lambda_0^2}{2\pi n_0 n_2}. \quad (4.7)$$

The "self trapping power" P_{st} is also known as the "critical power" P_{cr} . When the beam power P obeys $P < P_{cr}$ the beam diffracts, when $P > P_{cr}$ the beam would self-focus and when $P = P_{cr}$ the beam would self-trap. The beam in the self trapping case is also known as a spatial *soliton*.

4.1.1 Mathematical description of beam self-action

A mathematical description of spatial self-action uses the paraxial wave equation (see Eq.2.111):

$$2ik_n \frac{\partial \mathbf{A}_n}{\partial z} + \nabla_T^2 \mathbf{A}_n = -\frac{\omega_n^2}{\varepsilon_0 c^2} \mathbf{p}_n e^{i\Delta kz}, \quad (4.8)$$

In the scalar case and where we have only a single beam self-acting on itself we replace \mathbf{A}_n with A , k_n with k and ω_n with ω . As all frequencies involved in the process are ω we set $\Delta k = 0$. For the nonlinear polarization term, we first remind that (see section 2.9):

$$\mathbf{P}_n(\mathbf{r}, t) = \mathbf{p}_n(\mathbf{r}) e^{i(k_n z - \omega_n t)} + c.c. \quad (4.9)$$

Using its scalar form together with (see Eq.3.5)¹:

$$P_n = P^{NL}(\omega) = 3\epsilon_0 \chi^{(3)} (\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega), \quad (4.10)$$

we can write:

$$p_{NL} = 3\epsilon_0 \chi^{(3)} |A|^2 A. \quad (4.11)$$

Notice that we assume CW for all fields so there is no dependency on time, or on ω for that matter.

Let us treat the case where the beam is confined in a planar waveguide so the beam dynamics occur only along two directions, say x and z such that:

$$2ik \frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3 \frac{\omega^2}{c^2} \chi^{(3)} |A|^2 A. \quad (4.12)$$

It can be verified by direct substitution that this equation has the following solution:

$$A(x, z) = A_0 \operatorname{sech}(x/x_0) e^{i\gamma z}, \quad (4.13)$$

where the width of the beam is given with:

$$x_0 = \frac{1}{k} \sqrt{n_0 / (2\bar{n}_2 |A_0|^2)}, \quad (4.14)$$

and the rate of change of the nonlinear phase is:

$$\gamma = k\bar{n}_2 |A_0|^2 / n_0. \quad (4.15)$$

We remind that (see Eq.3.11) $\bar{n}_2 = 3\chi^{(3)}/4n_0$.

This solution in the form of the sech function is the spatial soliton. It is characterized by having an invariant profile $|A(x, z)| = |A(x, z = z_0)|$. The soliton in two dimensions is usually marked as "1+1" dimensions - as one dimension corresponds to the transverse profile (x here) and one for evolution (z). These solitons are stable: it does not matter what is the power or intensity of the beam, they would keep their form. In contrast, in full three dimensions (2+1 dimensions) and in the regular Kerr media the solitons need to have the critical power P_{cr} in order to stay invariant, in

¹ Also note that in Eq.3.5 we used k' instead of k to allow for phase mismatch, which is absent here as all frequencies in the process are the same.

other words - they are unstable. However if the Kerr media itself is saturable (once the intensity reaches a certain threshold - it does not change the index of refraction any further) it can lead to stable solitons even in (2+1) D. We also mention that for the (2+1) D problem in regular Kerr media there are no known exact analytical soliton solutions.

4.2 Optical phase conjugation by four wave mixing

4.2.1 Optical phase conjugation

Optical phase conjugation is a nonlinear optical process which generates from a given field another field whose phasor components are the complex conjugate of the phasor components of the original field. Mathematically, if an original monochromatic field \mathbf{E}_s is written as:

$$\begin{aligned}\tilde{\mathbf{E}}_s(\mathbf{r}, t) &= \mathbf{E}_s(\mathbf{r})e^{i\omega t} + c.c. \\ &= \mathbf{E}_s(\mathbf{r})e^{i\omega t} + \mathbf{E}_s^*(\mathbf{r})e^{-i\omega t}.\end{aligned}\quad (4.16)$$

then, the phase conjugate field is given with:

$$\begin{aligned}\tilde{\mathbf{E}}_c(\mathbf{r}, t) &= r\mathbf{E}_s^*(\mathbf{r})e^{i\omega t} + c.c. \\ &= r\mathbf{E}_s^*(\mathbf{r})e^{i\omega t} + r\mathbf{E}_s(\mathbf{r})e^{-i\omega t}. \\ &= r\mathbf{E}_s(\mathbf{r})e^{-i\omega t} + r\mathbf{E}_s^*(\mathbf{r})e^{i\omega t}. \\ &= r\tilde{\mathbf{E}}_s(\mathbf{r}, -t),\end{aligned}\quad (4.17)$$

where r is a "reflection coefficient" relating to the phase conjugate field generated from a "phase conjugate mirror". The last result $\tilde{\mathbf{E}}_c(\mathbf{r}, t) = r\tilde{\mathbf{E}}_s(\mathbf{r}, -t)$ indicates that phase conjugation creates a time-reversed wave-front.

Let us further examine phase-conjugation by describing the amplitude $\mathbf{E}_s(\mathbf{r})$ with:

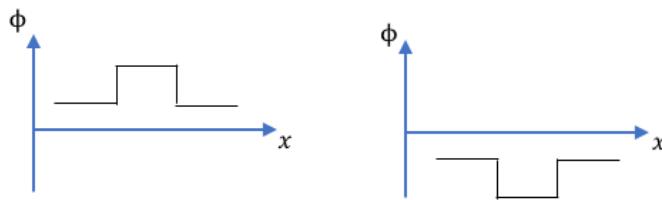
$$\mathbf{E}_s(\mathbf{r}) = \hat{\epsilon}_s A_s(\mathbf{r}) e^{i\mathbf{k}_s \cdot \mathbf{r}}, \quad (4.18)$$

where $\hat{\epsilon}_s$ denotes the polarization state of the field (the polarization unit vector), $A_s(\mathbf{r})$ is the slowly varying envelope of the field and \mathbf{k}_s is the mean wave-vector of the field. The complex conjugate of Eq.4.18 is given with:

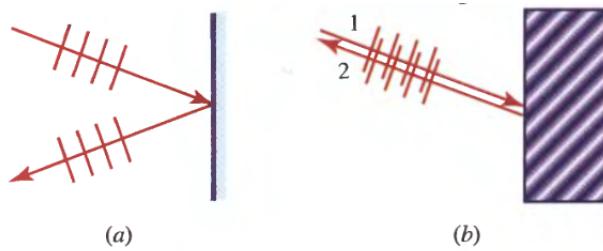
$$\mathbf{E}_s^*(\mathbf{r}) = \hat{\epsilon}_s^* A_s^*(\mathbf{r}) e^{-i\mathbf{k}_s \cdot \mathbf{r}}. \quad (4.19)$$

We can thus now state three effects related to phase-conjugation:

1. The polarization unit vector is replaced by its conjugate. This does not happen in regular reflection. Remember that in regular reflection the handedness (relevant for circular polarization) is switched - because the polarization vector stays the same but the wave vector component perpendicular to reflecting surface changes sign, and the handedness is defined as the rotation around the propagation axis. Here the switch in the direction of the wave vector comes with a switch in the rotation direction of the optical field - so overall the handedness does not change.
2. The wavefront described by $A_s(\mathbf{r}) = |A_s(\mathbf{r})|e^{i\phi(\mathbf{r})}$ is replaced by $A_s^*(\mathbf{r}) = |A_s(\mathbf{r})|e^{-i\phi(\mathbf{r})}$, implying wave-front reversal. See Fig.4.2 for an illustration.
3. The wave-vector \mathbf{k}_s is replaced with $-\mathbf{k}_s$. Not only the component perpendicular to the reflecting surface is reversed as in regular reflection - but also the component parallel to it. This means that a beam is reflected exactly back to itself. This is demonstrated in Fig.4.3 for a reflection from a regular mirror and from a Phase-Conjugate-Mirror (PCM) and in Fig.4.4 for a similar reflection of a spherical wave (which can be seen as a superposition of many plane waves).

**Fig. 4.2**

Effect of phase conjugations on the phase fronts of the field for a simple case of normal incidence where x is the coordinate perpendicular to the propagation direction. left: $\phi(x)$ right: $\phi(-x)$.

**Fig. 4.3**

Reflection of a plane wave from (a) a regular mirror (b) a PCM mirror

Looking at Figures 4.3-4.4, it is quite obvious that the reflection of a PCM is akin to simply turning back time and following all ray directions.

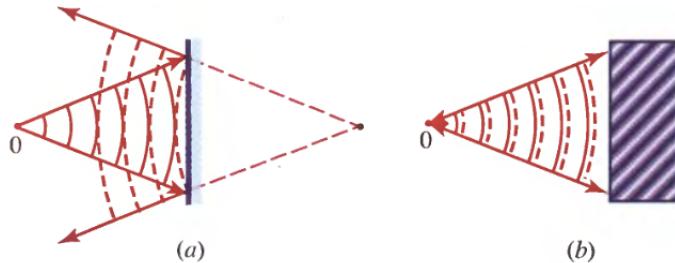


Fig. 4.4

Reflection of a spherical wave from (a) a regular mirror (b) a PCM mirror

4.2.2 Aberration correction using phase conjugation

With phase conjugation it is possible to remove aberrations acquired by a field propagating through some medium. This is done by using a PCM - then, the reflected wavefronts would retrace the wavefronts of the incident wave so that after the medium we get again an undistorted field. Conceptually this situation is depicted in Fig.4.5.

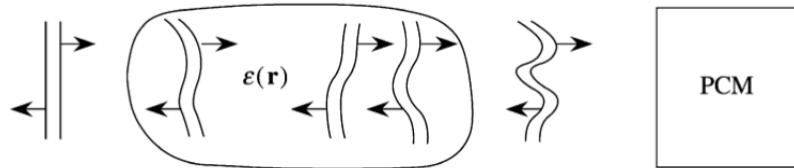


Fig. 4.5

An incident wave and its reflected conjugate propagating in some distorting medium.

To prove this claim we derive again a scalar version of the paraxial wave equation (see Eq.2.111) but for a linear propagation equation (no nonlinear polarization term) and also for a field of the form $E(\mathbf{r}, t) = A(\mathbf{r})e^{i(kz-\omega t)}$ where we choose $k = \frac{\omega}{c}$ and not $\frac{\omega}{c}n$ (it does not include the index of refraction). Under these assumptions we get:

$$2ik \frac{\partial A}{\partial z} + \nabla_T^2 A + \left[\frac{\omega^2 \epsilon(\mathbf{r})}{c^2} - k^2 \right] A = 0. \quad (4.20)$$

Notice that if we were to choose $k = \frac{\omega}{c}n = \frac{\omega}{c}\sqrt{\epsilon}$ the last term in square parentheses would have been zero (which is a form also often used in the literature).

Now - this equation would still be true if we complex conjugate it:

$$-2ik \frac{\partial A^*}{\partial z} + \nabla_T^2 A^* + \left[\frac{\omega^2 \epsilon(\mathbf{r})}{c^2} - k^2 \right] A^* = 0. \quad (4.21)$$

But this last equation describes the field:

$$E_c = A^*(\mathbf{r})e^{i(-kz-\omega t)}, \quad (4.22)$$

which is exactly a wave propagating in the negative z direction with an amplitude which is the complex conjugate of the forward propagating wave *everywhere*. In particular if the ingoing wave was a simple plane wave with no aberrations , before entering the medium, then the return wave would also be a plane wave (after going back through the medium) with no aberrations.

4.2.3 Phase Conjugation by Degenerate Four-Wave Mixing

Lets us consider the scheme presented in Fig.4.6. We have four interacting waves in a medium with a $\chi^{(3)}$ nonlinearity. The four waves are given with:

$$E_i(\mathbf{r}, t) = E_i(\mathbf{r})e^{-i\omega t} + c.c. = A_i(\mathbf{r})e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + c.c., \quad (4.23)$$

where $i = 1, 2, 3, 4$. The term "degeneracy" stands for the fact that all frequencies are the same for all fields.

Using Eq.3.4 in the scalar case (all field polarized in the same direction), we would have a FWM nonlinear polarization in the form of:

$$P_{NL} = 3\varepsilon_0 E^2 E^*, \quad (4.24)$$

Where $E(\mathbf{r}) = E_1(\mathbf{r}) + E_2(\mathbf{r}) + E_3(\mathbf{r}) + E_4(\mathbf{r})$.

If we expand the last expression we would find many terms. However, we are interested just in the following specific term:

$$P^{NL} = 6\varepsilon_0 \chi^{(3)} E_1(\mathbf{r}) E_2(\mathbf{r}) E_3^*(\mathbf{r}) = 6\varepsilon_0 \chi^{(3)} A_1 A_2 A_3^* e^{i(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3) \cdot \mathbf{r}}. \quad (4.25)$$

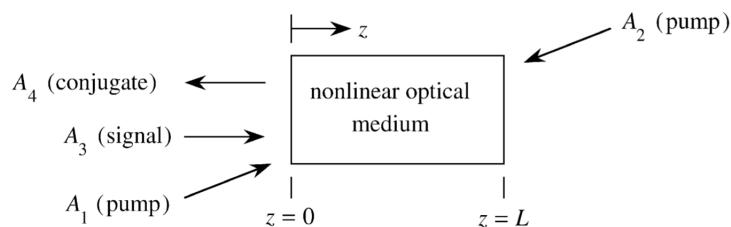


Fig. 4.6

Scheme for phase conjugation using degenerate Fourer-Wave-Mixing.

Now, we assume that the waves E_1 and E_2 are counter-propagating: $\mathbf{k}_1 + \mathbf{k}_2 = 0$, so we can write:

$$P^{NL} = 6\varepsilon_0 \chi^{(3)} A_1 A_2 A_3^* e^{-i\mathbf{k}_3 \cdot \mathbf{r}}. \quad (4.26)$$

This term would be the phase-matched polarization for the field E_4 if $\mathbf{k}_4 = -\mathbf{k}_3$. Further, the field E_4 would be the phase conjugate of E_3 if the term $A_1 A_2$ has at most a constant phase. This is easily satisfied if A_1 and A_2 are the amplitudes of plane waves (so they are constants) or if they are phase conjugate of each other so $A_1 A_2 = A_1 A_1^* = |A_1|^2$.

An illuminating interpretation for the scheme in Fig.4.6 is the following: The signal wave with amplitude A_3 interferes with one of the pump waves, say the one with amplitude A_1 to create an intensity standing wave. Through the $\chi^{(3)}$ driven Kerr effect - this creates a modulation in the index of refraction in the form of a volume diffraction grating. The second pump wave with amplitude A_2 diffracts from this grating to create the wave with amplitude A_4 . Notice that the volume grating would have small wiggling due to the phase of the signal wave (if it is not flat) - and these "aberrations" are the factor that generates the specific amplitude giving back the phase conjugate wave (and not just diffraction with no change in form of the amplitude).

4.3 Optical bistability

Optical bistability describes a system that can produce two different stable output intensities for a given input intensity. Optical bistability can be used as an all-optical switch.

A model system that exhibit bistability is a Fabry-Perot interferometer containing a nonlinear medium inside it (see Fig.4.7). Either or both the real part of the index of refraction (which we usually call the *index of refraction*) or its imaginary part (which we usually term the *absorption coefficient*) can be dependent on the light intensity.

We define the reflectance and transmittance amplitudes with ρ and τ such that the various amplitudes are connected through:

$$A'_2 = \rho A_2 e^{2ikl - \alpha l}, \quad (4.27)$$

and

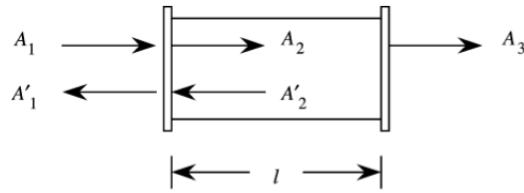
$$A_2 = \tau A_1 + \rho A'_2. \quad (4.28)$$

We assume that the wave-vector $k = n_2 \omega / c$ (with n_2 denoting the linear index of refraction within the cavity, denoted with the index 2) and the absorption coefficient α are real and that they can depend in a nonlinear way on the intensity of light within the cavity.

Also we assume the system is lossless so that $|\rho|^2 + |\tau|^2 = R + T = 1$.

We can eliminate A'_2 from the above equations to get:

$$A_2 = \frac{\tau A_1}{1 - \rho^2 e^{2ikl - \alpha l}}. \quad (4.29)$$

**Fig. 4.7**

Bistable optical device. A Fabry-Perot interferometer containing a nonlinear medium.

We now examine the case in which the cavity is in resonance so that $kl = \pi \cdot m$ with m being some integer, so $e^{i2kl} = 1$ and we can set $\rho^2 e^{i2kl} = R$. We also assume that $\alpha l \ll 1$ so $e^{-\alpha l} \simeq 1 - \alpha l$. Overall we get:

$$A_2 = \frac{\tau A_1}{1 - R(1 - \alpha l)}. \quad (4.30)$$

Notice that we neglect here any nonlinear effect on the index of refraction, but allow for a nonlinear effect on the absorption. In effect we analyze here *Absorptive Bistability*.

We can write the above relation again using intensities with the conversion $I_i = 2n_i \varepsilon_0 |A_i|^2$:

$$I_2 = \frac{\left(\frac{n_2}{n_1}\right) T I_1}{[1 - R(1 - \alpha l)]^2}. \quad (4.31)$$

We introduce the *cooperation number*:

$$C = \frac{R \alpha l}{1 - R}, \quad (4.32)$$

which is used to get:

$$I_2 = \frac{n_1}{n_2 T} \frac{I_1}{(1 + C)^2}. \quad (4.33)$$

Now we explicitly introduce nonlinearity in the absorption coefficient in the form that describes a *saturable absorber*:

$$\alpha = \frac{\alpha_0}{1 + I/I_s}, \quad (4.34)$$

where I is the total intensity and I_s is the saturation intensity. We see that $\alpha \xrightarrow{I \rightarrow 0} \alpha_0$ and that $\alpha \xrightarrow{I \rightarrow \infty} 0$.

We note that in our analysis we ignored the fact that in our interferometer the intensity is actually a standing wave due to the interference of the waves propagating to both directions in the cavity. So we did an approximate analysis. Actually if we were to take a ring-resonator (or traveling-wave interferometer) we could have excited modes inside the cavity which are propagating solely to one direction.

Anyway, with our current cavity we can approximate the total intensity within the cavity with $I_2 + I'_2 \simeq 2I_2$.

We can now express the cooperation number (using $I = 2I_2$) with:

$$C = \frac{C_0}{1 + 2I_2/I_s}, \quad (4.35)$$

with $C_0 = R\alpha_0 l / (1 - R)$

Using the last expression in Eq.4.33 we get:

$$I_1 = \frac{n_2}{n_1} TI_2 \left(1 + \frac{C_0}{1 + 2I_2/I_s} \right)^2. \quad (4.36)$$

The output intensity is related to I_2 through $I_3 = TI_2$ so overall the relation between the output to input intensities is given with:

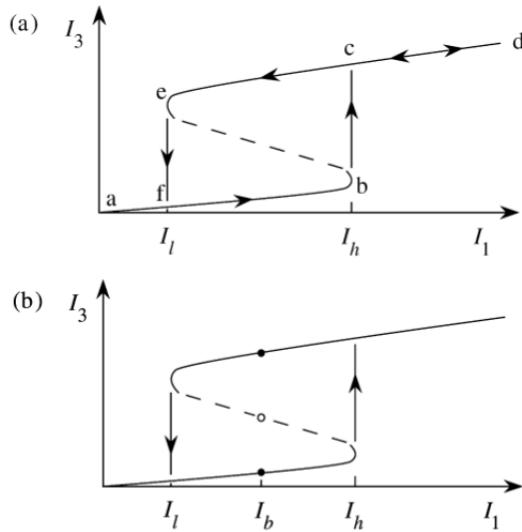
$$I_1 = \frac{n_2}{n_1} I_3 \left(1 + \frac{TC_0}{T + 2I_3/I_s} \right)^2. \quad (4.37)$$

This relation can have two or three output intensity solutions for a given input value. This is depicted in Fig.4.8.

The continuous line in this graph corresponds to cases in which the change in the output intensity is the same as in the input intensity: they both increase or decrease together. The dashed line correspond to a case in which the output and input intensities change in opposite manner. Using stability analysis (which we do not perform here) it can be shown that the dashed line corresponds to unstable states. So if the system is on that line - it would jump to a stable state on the continuous line. So in general, changing the input intensity would cause the output intensity to move on the continuous line. This also explains the hysteresis that we get (see Fig.4.8.(a)) if we gradually increase the input intensity from zero to over the value I_h and then decrease it gradually down below I_h .

The use of a bistable optical device as an all-optical switch is depicted in Fig.4.8.(b). The input intensity is held at the value I_b . The two stable value in this case are marked with filled dots. Now, if the system is in the lower state, it can be forced to make a transition to the upper state if momentarily an input pulse is applied - so that the total intensity rises above I_h . Similarly, if the system is at the upper state, blocking momentarily the whole beam would bring it back again to the lower state.

We would like to note that optical bistability can also be achieved with a nonlinear index of refraction (not only with a nonlinear absorption) leading to *Refractive Bistability*. We do no analyze this case here.

**Fig. 4.8**

Input-output intensity relations for a bistable optical device. (a) Hysteresis (b) switching.

4.4 Two beam coupling

Tow beam coupling is a nonlinear process in which the intensity of each beam modifies the index of refraction for the other beam. In such a process efficient transfer of energy can occur between the beams. Such a process can be driven by different mechanisms (e.g. stimulated scattering, in photorefractive materials, in a two-level system). The analysis we give here is general, without considering a specific mechanism.

We assume the existence of two beams oscillating at possibly different frequencies and propagating in possibly different directions:

$$E(\mathbf{r}, t) = A_1(\mathbf{r})e^{i(\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)} + A_2(\mathbf{r})e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)} + c.c.. \quad (4.38)$$

We assume here a model where the explicit value of the wave-vectors is given with $k_i = n_0\omega/c$; $i = 1, 2$, as if there is a single index of refraction n_0 for the whole field. The differences between the actual indices of refraction would be expressed in the phases of the amplitudes A_1 and A_2 .

The two beams interfere to create an averaged intensity:

$$I = n_0\varepsilon_0c\langle E^2 \rangle. \quad (4.39)$$

where $\langle \rangle$ denote time average of an interval of many optical periods. This aver-

aging can still leave out slowly varying time-dependent terms. Notice also that the use of a single index of refraction n_0 comes handy here.

Substituting Eq.4.38 in Eq.4.39 gives:

$$I = 2n_0\epsilon_0c \left\{ |A_1|^2 + |A_2|^2 + \left[A_1 A_2^* e^{i(\mathbf{q} \cdot \mathbf{r} - \delta t)} + c.c. \right] \right\}, \quad (4.40)$$

where

$$\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2 \quad (4.41)$$

is the grating wave-vector (which is the wave-vector difference), and

$$\delta = \omega_1 - \omega_2, \quad (4.42)$$

is the frequency difference which is assumed to obey $|\delta| \ll \omega_1, \omega_2$. The interference pattern given by the intensity Eq.4.40 is shown in Fig.4.9. The interference pattern moves with phase velocity $\delta/|\mathbf{q}|$ in the direction of \mathbf{q} if $\delta > 0$, it moves in the other direction if $\delta < 0$ and it is stationary for $\delta = 0$.

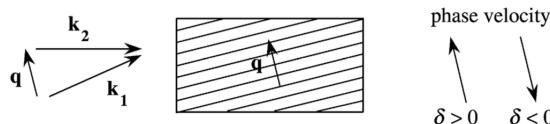


Fig. 4.9

Interference pattern for the superposition of two beams.

We assume here that this modulation in the intensity translates to a modulation in the index of refraction, and so each beam is scattered from this induced grating. This scattering has two effects - first due to the spatial variations, each wave scatters energy to a mode propagating in another direction and oscillating with the same frequency. Actually the wave with direction \mathbf{k}_1 can be scattered to a mode propagating in direction \mathbf{k}_2 and vice-versa, as the grating vector \mathbf{q} momentum match these two momenta. Now, the temporal variations might also lead to an exchange of energy between the modes. However, we shall see below, that this is only possible if the nonlinearity is not instantaneous - that is, if it takes the medium some time to change the index of refraction following the changes in the intensity (temporal changes in the interference grating).

To analyze the possibility of energy transfer we assume that the nonlinear part of the index of refraction obeys a *Debye relaxation equation*:

$$\tau \frac{dn_{NL}}{dt} + n_{NL} = n_2 I. \quad (4.43)$$

This equation is known to be applicable to Kerr media and to a two-level atom far from resonance. τ is the typical time interval dynamics for the nonlinear index of refraction.

It is obvious that in steady state $n_{NL} = n_2 I$ in accordance with Eq.3.1.

It can be shown that the solution to the Eq.4.43 is given with:

$$n_{NL} = \frac{n_2}{\tau} \int_{-\infty}^t I(t') e^{(t'-t)/\tau} dt'. \quad (4.44)$$

Substituting the intensity (Eq.4.40) into Eq.4.44 gives:

$$n_{NL} = 2n_0 n_2 \varepsilon_0 c \left[|A_1|^2 + |A_2|^2 + \frac{A_1 A_2^* e^{i(\mathbf{q} \cdot \mathbf{r} - \delta t)}}{1 - i\delta\tau} + \frac{A_1^* A_2 e^{-i(\mathbf{q} \cdot \mathbf{r} - \delta t)}}{1 + i\delta\tau} \right]. \quad (4.45)$$

Let us note that when $\delta\tau = 0$ then n_{NL} is real, and when $\delta\tau \neq 0$ then n_{NL} becomes complex - it gains a phase. In particular we can think about this as some kind of lag (delay) between the nonlinear index of refraction and the intensity pattern generating it.

We now use the scalar version of the wave equation for isotropic, dispersion-less media (see Eq.1.8), but with no sources, where the nonlinearity is acting back on the index of refraction:

$$\nabla^2 E - \frac{n^2}{c^2} \frac{\partial^2}{\partial t^2} E = 0, \quad (4.46)$$

with

$$n = n_0 + n_{NL}. \quad (4.47)$$

we assume that $|n_{NL}| \ll n_0$ so we can write:

$$n^2 = n_0^2 + 2n_0 n_{NL}. \quad (4.48)$$

If we now substitute the field (Eq.4.38) into the wave equation Eq.4.46 and compare all terms multiplying $e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)}$ we find:

$$\begin{aligned} & \frac{d^2 A_2}{dz^2} + 2ik_2 \frac{dA_2}{dz} - k_2^2 A_2 + \frac{n_0^2 \omega_2^2}{c^2} A_2 \\ &= -\frac{4n_0^2 n_2 \omega_2^2 \varepsilon_0}{c} (|A_1|^2 + |A_2|^2) A_2 - \frac{4n_0^2 n_2 \omega_1^2 \varepsilon_0}{c} \frac{|A_1|^2 A_2}{1 + i\delta\tau}. \end{aligned} \quad (4.49)$$

The last term on the right is the result of scattering the field $A_1 e^{i(\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)}$ from the last term on the right of the nonlinear time-varying index of refraction (Eq.4.45), which is time-dependent.

The first term on the right is the result of scattering of the field $A_2 e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)}$ from the first component on the right of Eq.4.45, which is a stationary component.

Let us now make use of the SVEA by dropping the first term on the left of Eq.4.49. We also note that the third and fourth terms on the left cancel each other. If we further assume $\omega_1 \simeq \omega_2$ and we replace both with ω we are left with:

$$\frac{dA_2}{dz} = 2in_0n_2\varepsilon_0\omega \left[(|A_1|^2 + |A_2|^2) A_2 + \frac{|A_1|^2 A_2}{1+i\delta\tau} \right]. \quad (4.50)$$

We would like to investigate the spatial rate of change of the intensity to find out how energy can be exchanged between the two fields. For this we first write:

$$\frac{dI_2}{dz} = \frac{d}{dz}(2n_0\varepsilon_0cA_2A_2^*) = 2n_0\varepsilon_0c \left(A_2^* \frac{dA_2}{dz} + A_2 \frac{dA_2^*}{dz} \right). \quad (4.51)$$

Using Eq.4.50-4.51 together with $I_1 = 2n_0\varepsilon_0cA_1A_1^*$ we find that:

$$\frac{dI_2}{dz} = \frac{2n_2\omega}{c} \frac{\delta\tau}{1+\delta^2\tau^2} I_1 I_2. \quad (4.52)$$

The dependency of the intensity (or energy) exchange on the detuning δ and relaxation time of the nonlinear index of refraction τ is shown in Fig.4.10.

This result is quite interesting - we see that for an energy exchange we must have some finite relaxation time. For an instantaneous response $\tau \rightarrow 0$ there would be no energy exchange (but also not for $\tau \rightarrow \infty$). Returning back to Eq.4.45, we see that for an energy exchange we must have some delay or temporal phase shift between the intensity grating and the change in the index of refraction induced by it. Finally, we would like to note, that energy exchange can also occur in another setting of two-beam coupling (in photorefractive crystals) where $\delta = 0$ but another mechanism allows for a *spatial* phase shift instead of the temporal phase shift observed here.

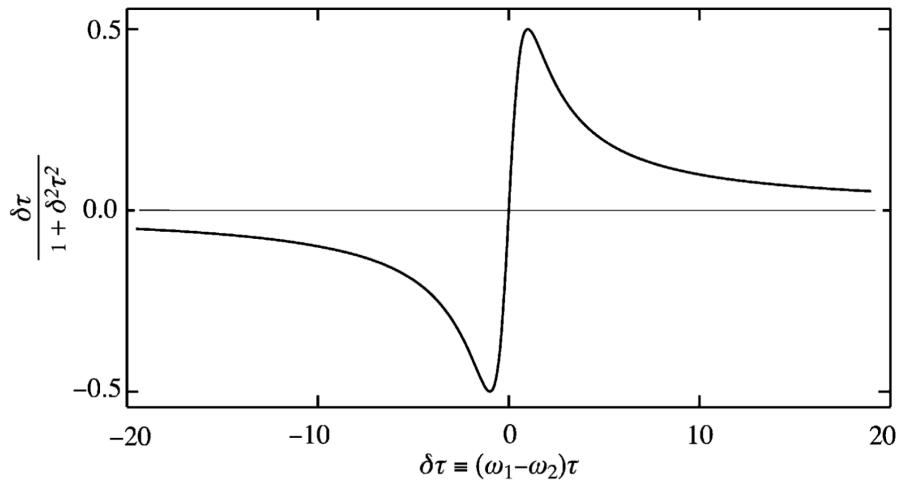


Fig. 4.10 Gain in two-beam-coupling as a function of frequency detuning δ and relaxation time τ .

5.1 Characterization of optical pulses

We consider here pulses in one spatial dimension:

$$E(z, t) = A(z, t)e^{i(k_0 z - \omega_0 t)} + c.c., \quad (5.1)$$

where ω_0 and k_0 are the carrier frequencies in the time and spatial domains respectively.

The spectral representation of the pulse is given using the Fourier transform:

$$E(z, \omega) = \int_{-\infty}^{\infty} E(z, t)e^{i\omega t} dt, \quad (5.2)$$

and its inverse:

$$E(z, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(z, \omega)e^{-i\omega t} d\omega. \quad (5.3)$$

We can define a Fourier transform for the envelope $A(z, t)$ as well:

$$A(z, \omega) = \int_{-\infty}^{\infty} A(z, t)e^{i\omega t} dt, \quad (5.4)$$

If we substitute Eq.5.1 into Eq.5.2 and make use of Eq.5.4 we get:

$$E(z, \omega) = A(z, \omega - \omega_0)e^{ik_0 z} + A^*(z, \omega + \omega_0)e^{-ik_0 z} \quad (5.5)$$

Of special interest for us would be the phase of the envelope both in the time domain and in the spectral domain.

We denote:

$$A(z, t) = |A(z, t)|e^{i\phi(t)}, \quad (5.6)$$

and

$$A(z, \omega - \omega_0) = |A(z, \omega - \omega_0)|e^{i\phi(\omega - \omega_0)}. \quad (5.7)$$

For signals which have a bandwidth which is smaller compared to the central frequency, we can expand the spectral phase in a Taylor expansion and keep only a finite number of elements:

$$\begin{aligned}\phi(\omega - \omega_0) &= \phi_0 + \frac{d\phi}{d\omega} \Big|_{\omega=\omega_0} (\omega - \omega_0) + \frac{1}{2} \frac{d^2\phi}{d\omega^2} \Big|_{\omega=\omega_0} (\omega - \omega_0)^2 + \dots \\ &= \phi_0 + \phi_1 \cdot (\omega - \omega_0) + \frac{1}{2} \phi_2 \cdot (\omega - \omega_0)^2 + \dots\end{aligned}\quad (5.8)$$

The term ϕ_0 is known as the *carrier-envelope-phase* or absolute phase. In the time domain it just adds the same phase value which act as a constant phase shift between the carrier and the envelope.

The linear spectral phase term translates to a delay in the time domain:

$$A(z, \omega - \omega_0) \cdot e^{i\phi_1 \cdot (\omega - \omega_0)} \xrightarrow{\mathfrak{F}} A(z, t - \phi_1) \quad (5.9)$$

where \mathfrak{F} denotes the Fourier transform. For this reason $\phi_1 = \frac{d\phi}{d\omega} \Big|_{\omega=\omega_0} = \tau_g$ is also known as the *group delay* and it has units of time.

The $\phi_2 = \frac{d^2\phi}{d\omega^2} \Big|_{\omega=\omega_0}$ term describes how the group delay is changing with frequency, for this reason it is called the *group-delay dispersion* (GDD).

The various spectral phase terms can be acquired by a given pulse propagating through some medium. The phase response of the medium can be described through the dependency of the wave-vector on the frequency $k = k(\omega)$. That is - the field in the spectral domain is transformed through:

$$A(z, \omega - \omega_0) \rightarrow A(z, \omega - \omega_0) e^{ik(\omega)z}. \quad (5.10)$$

If the only effect the pulse experiences is dispersion.

Expanding the wave-vector using:

$$\begin{aligned}k(\omega) &= k_0 + \frac{dk}{d\omega} \Big|_{\omega=\omega_0} (\omega - \omega_0) + \frac{1}{2} \frac{d^2k}{d\omega^2} \Big|_{\omega=\omega_0} (\omega - \omega_0)^2 + \dots \\ &= k_0 + k_1 \cdot (\omega - \omega_0) + \frac{1}{2} k_2 \cdot (\omega - \omega_0)^2 + \dots\end{aligned}\quad (5.11)$$

The linear term is:

$$k_1 = \frac{dk}{d\omega} \Big|_{\omega=\omega_0} = \frac{1}{v_g} \Big|_{\omega=\omega_0}, \quad (5.12)$$

with v_g known as the *group velocity*.

The second order term:

$$k_2 = \frac{d^2k}{d\omega^2} \Big|_{\omega=\omega_0} = -\frac{1}{v_g^2} \frac{dv_g}{d\omega} \Big|_{\omega=\omega_0} \quad (5.13)$$

describes how the (inverse of the) group velocity is changing with frequency, for this reason it is also known as the *group velocity dispersion* (GVD). If the GVD is negative $k_2 < 0$ it means that the group velocity is larger for higher frequencies. So

during propagation the higher (bluish) frequency components of the pulse would precede the lower (reddish) frequency components, and vice-versa. This can be seen in Fig.5.1.

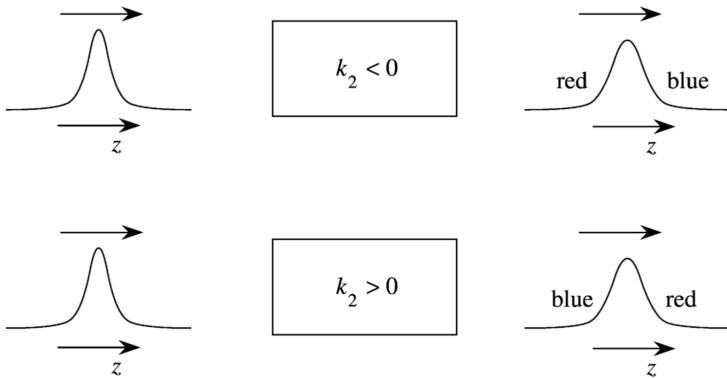


Fig. 5.1

Pulse spreading due to GVD.

In some textbooks an alternative notation is used:

$$\begin{aligned} k_1 &= \beta_1 \\ k_2 &= \beta_2 \end{aligned} \quad (5.14)$$

Now, equating $k(\omega)z = \phi(\omega)$ with the understanding that the spectral phase is acquired by propagation through some dispersive medium of length z we can see that:

$$\tau_g = \frac{z}{v_g}, \quad (5.15)$$

and

$$GDD = GVD \cdot z. \quad (5.16)$$

As an example for the action of the group velocity and GVD on a pulse, let us assume that we have a Gaussian pulse:

$$A(z = 0, t) = A e^{-\frac{1}{2}\left(\frac{t}{\tau_0}\right)^2}, \quad (5.17)$$

where τ_0 corresponds to the width of the Gaussian envelope.

The spectral envelope is found using standard Gaussian Fourier pair as:

$$A(z = 0, \omega - \omega_0) = A(0, \omega - \omega_0) = \frac{A\tau_0}{\sqrt{2\pi}} \exp\left\{-\frac{\tau_0^2}{2}(\omega - \omega_0)^2\right\}. \quad (5.18)$$

The spectral amplitude is transformed through propagation to:

$$A(z, \omega - \omega_0) = A(0, \omega - \omega_0) \cdot e^{i[k_0 + k_1 \cdot (\omega - \omega_0) + \frac{1}{2}k_2 \cdot (\omega - \omega_0)^2]z} \quad (5.19)$$

Substituting Eq.5.18 into Eq.5.19 and inverse Fourier transforming result (after some math) with:

$$A(z, t) = \frac{A}{\sqrt{1 - i\xi}} \exp\{ik_0 z\} \exp\left\{-\frac{(t - z/v_g)^2}{2\tau_0^2(1 + \xi^2)}\right\} \exp\left\{-\frac{i\xi(t - z/v_g)^2}{2\tau_0^2(1 + \xi^2)}\right\}, \quad (5.20)$$

where $\xi = k_2 z / \tau_0^2$.

There are several effects we can observe here.

1. First with no GVD (so here $\xi = 0$) the pulse just travels with a constant velocity v_g .
2. With GVD - the pulse becomes wider $\tau_0 \rightarrow \tau_0 \sqrt{1 + \xi^2}$. The amplitude drops because $|A| \rightarrow A / \sqrt{1 + \xi^2}$ (using the fact that for a complex number $|C|^2 = |C^2|$). We note that the energy in the pulse is constant: $\int_{-\infty}^{\infty} |A(z, t)|^2 dt$ is invariant of coordinate.
3. Regarding the phase we notice that the temporal phase of the whole field is now (excluding the minus sign) $\omega_0(t - z/v_g) + \frac{\xi(t - z/v_g)^2}{2\tau_0^2(1 + \xi^2)}$. The (minus of the) time derivative of this expression gives the instantaneous frequency (also known as the local frequency) which is now a linear function of time: $\omega_0 \rightarrow \omega_0 + \frac{\xi}{\tau_0^2(1 + \xi^2)}(t - z/v_g)$. Such a change in the local frequency is known as *chirp*, and we see that the source for this chirp is the GVD in the medium.

In the following we would consider nonlinear terms acting on the pulse, in particular, a Kerr term which would induce a change on the zeroth order wave-vector:

$$k_0 \rightarrow k_0 + \Delta k_{NL}, \quad (5.21)$$

with

$$\Delta k_{NL} = \Delta n_{NL} \frac{\omega_0}{c} = n_2 I \frac{\omega_0}{c}. \quad (5.22)$$

5.2 Propagation of optical pulses

We first start with the wave equation (see Eq.1.4):

$$\nabla^2 \mathbf{E} - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2}{\partial t^2} \mathbf{D}^{(1)} = 0, \quad (5.23)$$

with no nonlinear terms. We also would change $\mathbf{D}^{(1)}$ to \mathbf{D} to indicate that Kerr

nonlinearity would be incorporated into \mathbf{D} so the (1) symbol indicating linear response is no longer appropriate.

Next we simplify the problem to a single spatial dimension:

$$\frac{\partial^2}{\partial z^2} E - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2}{\partial t^2} D = 0. \quad (5.24)$$

We also use the Fourier transform Eq.5.2 $E(z, \omega)$ and we relate it to the Fourier transform of the displacement $D(z, \omega)$ through a constitutive relation:

$$D(z, \omega) = \varepsilon_0 \varepsilon(\omega) E(z, \omega), \quad (5.25)$$

where the effective dielectric constant $\varepsilon(\omega)$ is related to the wave-vector through:

$$k(\omega) = \sqrt{\varepsilon(\omega)} \frac{\omega}{c}. \quad (5.26)$$

We can now Fourier-transform the wave equation Eq.5.24 to get:

$$\frac{\partial^2}{\partial z^2} E(z, \omega) + \varepsilon(\omega) \frac{\omega^2}{c^2} E(\omega) = 0. \quad (5.27)$$

We now use Eq.5.5 defining the field amplitude in the frequency domain:

$$E(z, \omega) = A(z, \omega - \omega_0) e^{ik_0 z} + A^*(z, \omega + \omega_0) e^{-ik_0 z} \quad (5.28)$$

and substitute it into the wave equation Eq.5.27. Then we equate terms multiplied with $e^{ik_0 z}$ and make the SVEA approximation by dropping the second order derivative in z to get:

$$2ik_0 \frac{\partial A(z, \omega - \omega_0)}{\partial z} + (k^2(\omega) - k_0^2) A(z, \omega - \omega_0) = 0, \quad (5.29)$$

where we also made use of Eq.5.26.

We now assume that the values of $k(\omega)$ are not that different from k_0 so we can approximate:

$$k^2(\omega) - k_0^2 = (k(\omega) + k_0)(k(\omega) - k_0) \simeq 2k_0(k(\omega) - k_0), \quad (5.30)$$

to get:

$$\frac{\partial A(z, \omega - \omega_0)}{\partial z} - i(k(\omega) - k_0) A(z, \omega - \omega_0) = 0. \quad (5.31)$$

Using the Taylor expansion of $k(\omega)$ (see Eq.5.11) up to the second order terms and adding also the nonlinear term Eq.5.22 we can write:

$$\frac{\partial A(z, \omega - \omega_0)}{\partial z} - i\Delta k_{NL} A(z, \omega - \omega_0) - ik_1(\omega - \omega_0)A(z, \omega - \omega_0) - \frac{1}{2}ik_2(\omega - \omega_0)^2 A(z, \omega - \omega_0) = 0. \quad (5.32)$$

We note that if there is no nonlinearity $\Delta k_{NL} = 0$, then the solution for this equation is (see Eq.5.19):

$$A(z, \omega - \omega_0) = A(0, \omega - \omega_0) \cdot e^{i[k_0 + k_1 \cdot (\omega - \omega_0) + \frac{1}{2}k_2 \cdot (\omega - \omega_0)^2]z} \quad (5.33)$$

Next we operate on Eq.5.32 with the integral $\frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i(\omega - \omega_0)t} d(\omega - \omega_0)$ which inverse Fourier transform all terms:

$$\frac{\partial A(z, t)}{\partial z} + k_1 \frac{\partial A(z, t)}{\partial t} + \frac{1}{2}ik_2 \frac{\partial^2 A(z, t)}{\partial t^2} - i\Delta k_{NL} A(z, t) = 0. \quad (5.34)$$

A very useful transformation of the above equation is to switch to a coordinate system moving at the group velocity v_g :

$$\begin{aligned} z &\rightarrow \xi = z \\ t &\rightarrow \tau = t - k_1 z = t - z/v_g. \end{aligned} \quad (5.35)$$

With this transformation we can replace all the derivatives in the wave equation:

$$\begin{aligned} \frac{\partial}{\partial z} &= \frac{\partial}{\partial \xi} \frac{\partial \xi}{\partial z} + \frac{\partial}{\partial \tau} \frac{\partial \tau}{\partial z} = \frac{\partial}{\partial \xi} - \frac{1}{v_g} \frac{\partial}{\partial \tau}, \\ \frac{\partial}{\partial t} &= \frac{\partial}{\partial \xi} \frac{\partial \xi}{\partial t} + \frac{\partial}{\partial \tau} \frac{\partial \tau}{\partial t} = \frac{\partial}{\partial \tau}, \\ \frac{\partial^2}{\partial t^2} &= \frac{\partial}{\partial t} \left(\frac{\partial}{\partial t} \right) = \frac{\partial^2}{\partial \tau^2}. \end{aligned} \quad (5.36)$$

With these the wave equation is transformed to:

$$\frac{\partial A(\xi, \tau)}{\partial \xi} + \frac{1}{2}ik_2 \frac{\partial^2 A(\xi, \tau)}{\partial \tau^2} - i\Delta k_{NL} A(\xi, \tau) = 0. \quad (5.37)$$

We can further explicitly write (see also Eq.5.22):

$$\Delta k_{NL} = n_2 I \frac{\omega_0}{c} = 2n_0 \varepsilon_0 n_2 \omega_0 |A|^2 = \gamma |A|^2, \quad (5.38)$$

to get:

$$\frac{\partial A(\xi, \tau)}{\partial \xi} + \frac{1}{2}ik_2 \frac{\partial^2 A(\xi, \tau)}{\partial \tau^2} = i\gamma |A(\xi, \tau)|^2 A(\xi, \tau). \quad (5.39)$$

This last equation is also known as the *Nonlinear Schrodinger equation* (NLSE).

5.2.1 Self-phase modulation

Let us consider a very simple scenario in which we can neglect dispersion and also assume that the Kerr nonlinearity would only change the phase of A and not its magnitude $|A|$. In this case we can write the wave equation:

$$\frac{\partial A(\xi, \tau)}{\partial \xi} = i n_2 I(\tau) \frac{\omega_0}{c} A(\xi, \tau). \quad (5.40)$$

The reason we "hide" A within the intensity I is that for first order we decouple the intensity from the amplitude and treat it as a parameter acting only on its phase. In other words, we can write the solution to the above equations as:

$$A(\xi, \tau) = A(\xi = 0, \tau) e^{i n_2 I(\tau) \omega_0 \xi / c} = A(\xi = 0, \tau) e^{i \phi_{NL}(\tau)}. \quad (5.41)$$

The phase acquired is of course time-dependent $\phi_{NL}(\tau, \xi) = n_2 I(\tau) \omega_0 \xi / c$. Again - notice that the intensity in this model does not depend on ξ , as only the phase of the field is changing here as a function of ξ .

The time derivative of this nonlinear phase is acting as a frequency shift:

$$\delta\omega(\tau) = -\frac{d}{d\tau} \phi_{NL}(\tau, \xi) = -n_2 \frac{\omega_0 \xi}{c} \frac{dI(\tau)}{d\tau}. \quad (5.42)$$

We should remember that the whole instantaneous frequency of the field is of course $\omega_0 + \delta\omega(\tau)$.

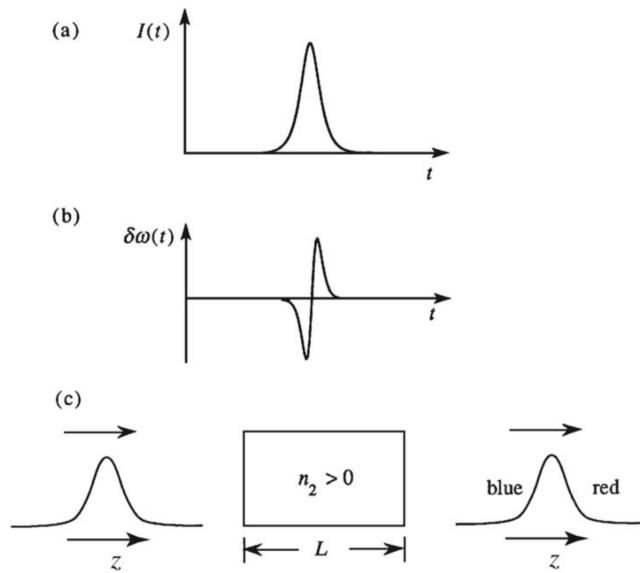
This phenomenon of a field modulating its own phase and so leading to a change in its frequency is known as *self-phase modulation* (SPM).

We note that this modulation increases the spectral bandwidth of the field as it generates new frequency components. In Fig.5.2 we can see the effect a symmetrical intensity pulse like a Gaussian or a Sech has on its own phase - by inducing an asymmetric instantaneous frequency which changes in time. In other words, SPM creates new frequencies and chirp.

5.2.2 Temporal solitons

Going back to the NLSE equation (Eq.5.39) we can now appreciate that there are two terms over there that can induce chirp - the linear dispersion term $\frac{1}{2} i k_2 \frac{\partial^2 A(\xi, \tau)}{\partial \tau^2}$ and the non-linear SPM term $i\gamma |A(\xi, \tau)|^2 A(\xi, \tau)$. There are actual solution to this equation in which the effects of SPM and dispersion completely balance. For this we need that for $n_2 > 0$ (which also sets $\gamma > 0$) which induces positive chirp the SPM would be accompanied with negative chirp induced by a negative $k_2 < 0$ (negative GVD). Also for $n_2 < 0$ we'll require $k_2 > 0$

An analytical solution to the NLSE of such a balanced solution which does not change its shape during propagation is known as a temporal soliton which is the temporal analogue to spatial solitons mentioned in section 4.1. We mention that for

**Fig. 5.2**

Self-phase modulation. (a) Pulse intensity. (b) Induced instantaneous frequency change through SPM .(c) With positive nonlinear coefficient $n_2 > 0$ SPM induces positive chirp - frequencies are swept from red to blue in the time domain, so the reddish part appears first.

spatial solitons beam diffraction was balanced with Kerr self-focusing. Here pulse dispersion is balanced with Kerr induced SPM.

One form of a soliton solution solving the NLSE is (this can be confirmed using direct substitution):

$$A(\xi, \tau) = A_0 \operatorname{sech}(\tau/\tau_0) e^{i\kappa\xi}, \quad (5.43)$$

where the amplitude A_0 and width τ_0 of the pulse are related through:

$$|A_0|^2 = \frac{-k_2}{\gamma\tau_0^2}, \quad (5.44)$$

and

$$\kappa = -\frac{k_2}{2\tau_0^2} = \frac{1}{2}\gamma|A_0|^2 \quad (5.45)$$

represents the phase accumulated during propagation.

This solution is known as a *fundamental soliton* while higher-order solitons are also known.

We note that the Fourier transform of the soliton apart from the spatial phase $\kappa\xi$

is invariant to propagation. This means that the spectral content of the soliton is also invariant to propagation. In a sense the nonlinear term and the dispersion term balance each other such there is no dispersion and no generation of new frequencies - no self-phase-modulation.

6.1 General features of light scattering

6.1.1 Nomenclature for light scattering

Light scattering is the coupling of incoming light modes to outgoing light modes in a medium through fluctuations in the optical properties of the medium. The input and output modes can differ in their momentum (direction of wave vector) and in their energy (frequency).

Light scattering can be characterized through various attributes.

First, scattering can be related to either *spontaneous scattering* or *stimulated scattering*. With spontaneous scattering the optical properties of the medium are unmodified by the incoming light, which is not the case with stimulated scattering, which occurs when light is intense enough. Stimulated scattering is formally a nonlinear phenomenon.

Second, scattering can be characterized by the shift (if any) of the frequency of the scattered light. A shift to lower frequency is known as *Stokes* components, while a shift to higher frequency is known as *anti-Stokes* components.

Third, light scattering can be characterized by the physical process underlying the scattering:

- Brillouin scattering: scattering of light from sound waves. That is - from propagating density waves quantized through acoustic phonons.
- Rayleigh scattering: scattering of light from non-propagating density fluctuations. Also known as Rayleigh-center scattering. This scattering is quasi-elastic as it induces no frequency shift.
- Rayleigh-wing scattering: scattering from fluctuations in the orientation of anisotropic molecules. As molecular orientation processes are very fast, this scattering component is broadband.
- Raman scattering: scattering of light from vibration modes of molecules, where these modes are quantized through optical phonons.

The spectrum of different types of light scattering processes is schematically shown in Fig.6.1(b).

Fourth, light scattering can be characterized by the way momentum is changing due to scattering. There are several options here:

- *Scattering from an homogenous medium.* In this case there is no change at all

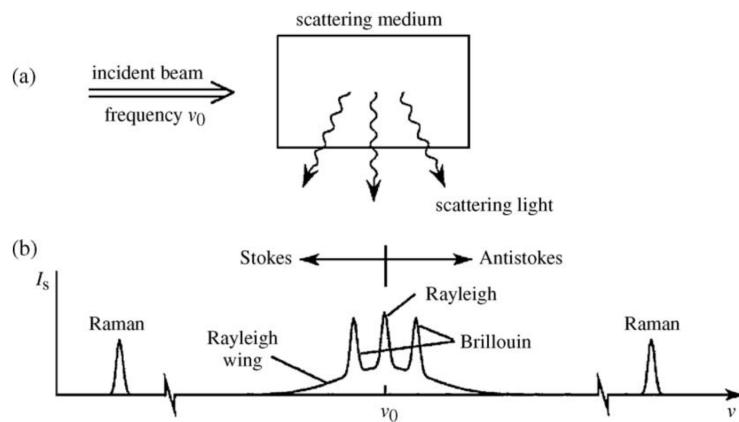


Fig. 6.1

Light scattering. (a) General setup for detecting light scattering. (b) Various types of light scattering processes.

for the momentum. Only forward scattering is possible. See also section 6.1.2 below.

- *Diffraction from an unordered medium.* In this case the scattering can be to all possible directions - momentum is not conserved at all.
- *Bragg diffraction.* Where the scattering is off an ordered medium in the form of a thick grating. For each reciprocal lattice vector (RLV) of the grating: a diffraction order different than forward scattering is possible provided that momentum matching is satisfied together with the RLV.
- *Raman-Nath diffraction.* Where the scattering is off an ordered medium in the form of a thin grating. For each reciprocal lattice vector (RLV) of the grating: several diffraction order different than forward scattering are possible provided that momentum matching is satisfied together with the RLV for the components of the momentum vectors along the RLV.

The last two cases are depicted in Fig.6.2). This difference between Bragg and Raman-Nath diffraction is a consequence of the position-momentum uncertainty relation. Because for a Bragg diffraction we have two equations (constraints) that needs to be fulfilled - momentum conservation along two perpendicular axes - there can be only a single solution for the diffraction equation - allowing for a single diffraction order. In the Raman-Nath case, there is only a single constraint, allowing for multiple diffraction orders. Put otherwise, with Bragg diffraction we need to conserve momentum in all axes, while with Raman-Nath - only along one of the axes. Now, notice that when the medium is static we also need to conserve the length of the momentum vectors. This then allows us for only two possible diffraction solutions for any RLV (In Fig.6.2 there is for the Raman-Nath case one K_g vector going down, giving the downward blue arrow, and one going up, giving the upward dashed green arrow).

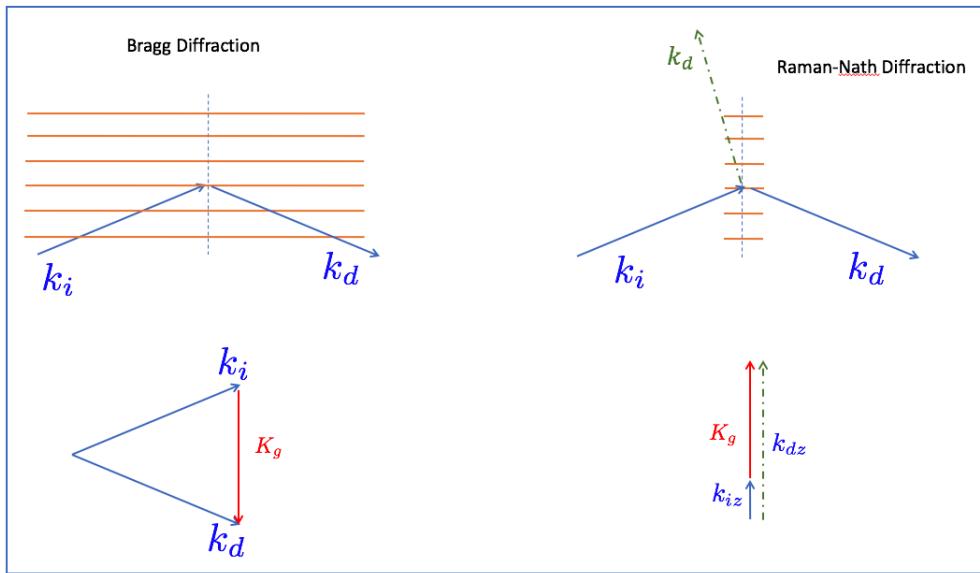


Fig. 6.2

Bragg and Raman-Nath diffraction (scattering). For Bragg diffraction momentum needs to be conserved exactly - along all directions. For Raman-Nath diffraction momentum is conserved along a single direction.

6.1.2 Fluctuations as the origin for light scattering

With no fluctuations light can only be scattered in the forward direction. Such scattering is known as *coherent forward scattering* and it is the origin for the index of refraction. To understand why a homogeneous medium (with no fluctuations) does not give rise to scattering other than forward scattering we can make use of Fig.6.3. In this figure we assume that a volume dV_1 scatters light at an angle θ (it actually emits light at all directions but we consider a particular direction which is not the forward direction). Because the medium is homogeneous we can always find a close by volume dV_2 whose scattering into the same direction is out of phase with the scattering from dV_1 . Thus destructive interference precludes any side scattering. Alternatively we can say that the dipole emissions from all the emitters in the medium can be built by constructive interference only in the forward direction. Notice that this explanation, which is a recount of Hugens' principle, does not account for the fact that there is no backward scattering as well. For this, a more elaborate explanation taking into account the full spatiotemporal behavior of the wave is needed.

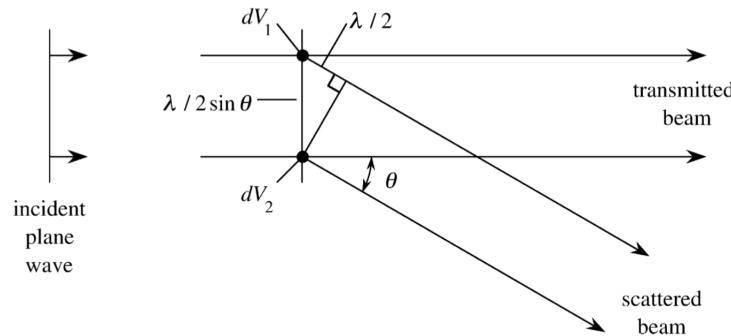


Fig. 6.3 Destructive interference for scattering in homogeneous medium.

6.1.3 Scattering coefficient R

The scattering coefficient R is used to describe the efficiency of spontaneous scattering according to the configuration shown in Fig.6.4. We assume that the scattered light intensity I_s of a volume V can be written as:

$$I_s = R \frac{I_0 V}{L^2}, \quad (6.1)$$

where I_0 is the incoming light intensity, L is the distance to the detector and R is the scattering coefficient. Notice, that according to the scheme in Fig.6.4, that R is dependent on the angle of scattering. Now, if the power hitting the detector of area dA is: $dP = I_s dA$ and the solid angle the detector subtend is $d\Omega = dA/L^2$ we can write:

$$\frac{dP}{d\Omega} = I_s L^2 = I_0 R V. \quad (6.2)$$

Either of the two equations given here are used to define the *scattering coefficient* R . And we again emphasize that R is dependent on scattering angle θ .

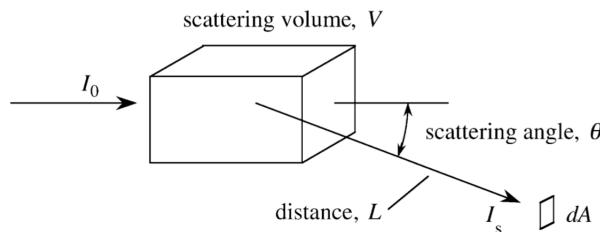


Fig. 6.4 Scheme for defining the scattering coefficient.

6.1.4 The scattering cross section σ

Another way to quantify the efficiency of scattering in the case of spontaneous scattering from a single molecule is to assume that the total power P scattered by the molecule is linear with respect to the incoming intensity I_0 :

$$P = \sigma I_0, \quad (6.3)$$

where σ is the *scattering cross section*, which has units of area. The cross section can be thought of as an effective area for removing light from the incoming beam. A related quantity is the *differential cross section* $d\sigma/d\Omega$ for solid angle $d\Omega$. This is the proportionality coefficient between the scattered power per solid angle and the incoming intensity:

$$\frac{dP}{d\Omega} = I_0 \frac{d\sigma}{d\Omega}. \quad (6.4)$$

If in a given volume V we have N molecules then the total power scattered per unit solid angle would be:

$$\frac{dP}{d\Omega} = I_0 N \frac{d\sigma}{d\Omega}. \quad (6.5)$$

If we compare this result with the expression for the scattering coefficient Eq.6.2 we get:

$$R = \frac{N}{V} \frac{d\sigma}{d\Omega}. \quad (6.6)$$

This result connects together the scattering coefficient with the scattering cross section, however - it is not always true, because it only works for a medium in which we can assume the scattering from N molecules is N times the scattering from a single molecule which in a sense is not applicable to a homogenous medium, as we saw that in such a medium there is only forward scattering. Thus this last relation can be applicable to dilute forms of matter (like a gas) but not to condensed matter.

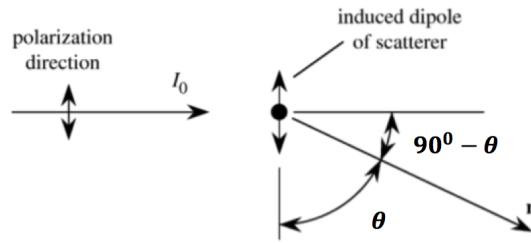
We next bring a model which describes R and σ .

6.1.5 Microscopic theory of spontaneous light scattering

A microscopic theory of scattering relates to the scattering from a single microscopic element whose size is much smaller than the wavelength of light. A single molecule satisfies this criterion. We consider the geometry depicted in Fig.6.5. As explained in the previous section, this treatment is of course relative only to a dilute collection of scatterers.

For an incoming field:

$$\mathbf{E} = \mathbf{E}_0 e^{-i\omega t} + c.c \quad (6.7)$$

**Fig. 6.5**

Light scattering from a single object much smaller than the wavelength of light. The dipole is assumed to be aligned along \hat{z} .

with intensity of $I_0 = (2nc\varepsilon_0)|E_0|^2$ we assume that the scatterer develops a dipole moment of the form:

$$\mathbf{p} = \varepsilon_0\alpha(\omega)\mathbf{E}_0 e^{-i\omega t} + c.c. = \varepsilon_0\alpha(\omega)E_0 e^{-i\omega t}\hat{z} + c.c., \quad (6.8)$$

where we assume that the field and the induced dipole moment are polarized along \hat{z} .

Here $\alpha(\omega)$ is the *polarizability* of the scatterer. Its precise form depends on the scatterer.

Using electromagnetic theory (see textbooks on electrodynamics) it can be shown that the time-varying dipole moment gives rise to a radiated (scattered) field, which in the far field can be approximated with:

$$\mathbf{E}_s = -\frac{\omega^2\mu_0\varepsilon_0\alpha(\omega)E_0}{4\pi} \sin\theta \frac{e^{i\omega(r/c-t)}}{r} \hat{\theta} + c.c. \quad (6.9)$$

The time-averaged intensity of this radiation in a location $r = L$ is:

$$I_s = 2n\varepsilon_0c\langle|\mathbf{E}_s|^2\rangle = \frac{n\omega^4\varepsilon_0|\alpha(\omega)|^2|\mathbf{E}_0|^2}{8\pi^2c^3L^2} \sin^2\theta. \quad (6.10)$$

where the time averaging is $\langle f(t) \rangle = \frac{1}{T} \int_0^T f(t)dt$ with $T = 2\pi/\omega$, and we used $c = 1/\sqrt{\varepsilon_0\mu_0}$.

We now use Eq.6.2, Eq.6.4 and Eq.6.10 to get:

$$\left(\frac{d\sigma}{d\Omega}\right)_p = \frac{1}{I_0} \frac{dP}{d\Omega} = \frac{I_s L^2}{I_0} = \frac{\omega^4}{16\pi^2c^4} |\alpha(\omega)|^2 \sin^2\theta. \quad (6.11)$$

The p subscript describes the fact that we chose to measure the angle θ in the plane of the polarization of the radiation, so we are dealing with p-type polarization. If instead we would have chosen to put the dipole perpendicular to the plane in Fig.6.5 (choosing s-type polarization) then the radiation is not dependent on θ in

the figure any more (now θ is equal to ϕ in the coordinates frame where the dipole is aligned along z), and in terms of the θ attached to the dipole coordinate frames we need to set $\theta = 90^\circ$. Overall:

$$\left(\frac{d\sigma}{d\Omega} \right)_s = \frac{\omega^4}{16\pi^2 c^4} |\alpha(\omega)|^2. \quad (6.12)$$

For unpolarized light we would take the average of the differential cross section for both polarizations:

$$\left(\frac{d\sigma}{d\Omega} \right)_{unpolarized} = \frac{\omega^4}{32\pi^2 c^4} |\alpha(\omega)|^2 (1 + \sin^2 \theta). \quad (6.13)$$

(Notice that our angle definition is different than in Boyd's textbook.)

We get the total cross section by integrating over all solid angles $d\Omega = \sin \theta d\theta d\phi$:

$$\sigma = \int_{4\pi} d\Omega \frac{d\sigma}{d\Omega} = \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi \frac{d\sigma}{d\Omega}. \quad (6.14)$$

For p-polarized light:

$$\sigma_p = \frac{\omega^4}{16\pi^2 c^4} |\alpha(\omega)|^2 \int_0^\pi d\theta \sin^3 \theta \int_0^{2\pi} d\phi = \frac{\omega^4}{16\pi^2 c^4} |\alpha(\omega)|^2 \cdot \frac{8\pi}{3} = \frac{\omega^4}{6\pi c^4} |\alpha(\omega)|^2. \quad (6.15)$$

Now, generally the polarizability is related to the susceptibility through the relation:

$$\chi(\omega) = N\alpha(\omega). \quad (6.16)$$

If we choose a Lorentz model of an harmonic oscillator for which (we repeat here Eq.6.18):

$$\chi^{(1)}(\omega) = \frac{Ne^2}{\varepsilon_0 m (\omega_0^2 - \omega^2 - 2i\gamma\omega)} = \frac{Ne^2}{\varepsilon_0 m D(\omega)}, \quad (6.17)$$

(where we remind that ω_0 is the resonance frequency and γ is the damping rate) then it is obvious that:

$$\alpha(\omega) = \frac{e^2}{\varepsilon_0 m (\omega_0^2 - \omega^2 - 2i\gamma\omega)}. \quad (6.18)$$

For p polarization, using Eq.6.15 we get:

$$\sigma = \frac{8\pi}{3} \left(\frac{e^2}{4\pi\varepsilon_0 mc^2} \right)^2 \frac{\omega^4}{(\omega_0^2 - \omega^2)^2 + 4\omega^2\gamma^2}. \quad (6.19)$$

This asymmetric line shape is shown in Fig.6.6.

Several limits of this expression are worth noting:

First for frequencies well below the resonance:

$$\omega \ll \omega_0 \rightarrow \sigma = \frac{8\pi}{3} \left(\frac{e^2}{4\pi\varepsilon_0 mc^2} \right)^2 \frac{\omega^4}{\omega_0^4} \quad (6.20)$$

This scattering regime is known as *Rayleigh scattering*. In this regime scattering is much more intense for shorter wavelengths. It explains why the sky is blue as the visible spectrum of the sun is below the resonance of dust particles in the atmosphere.

Close to the resonance:

$$\omega \simeq \omega_0 \rightarrow \sigma = \frac{8\pi}{3} \left(\frac{e^2}{4\pi\varepsilon_0 mc^2} \right)^2 \frac{\omega_0^2}{(\omega_0 - \omega)^2 + \gamma^2} \quad (6.21)$$

the scattering cross section spectrum is *Lorentzian*.

Finally, at frequencies much larger than the resonance:

$$\omega \gg \omega_0 \rightarrow \sigma = \frac{8\pi}{3} \left(\frac{e^2}{4\pi\varepsilon_0 mc^2} \right)^2 \quad (6.22)$$

the scattering is approximately constant as a function of frequency. This regime is known as *Thompson scattering*. Interestingly the scattering is close in value in this regime to the so-called "classical" electron radius $r_e = e^2/4\pi\varepsilon_0 mc^2 = 2.82 \cdot 10^{-15} m$.

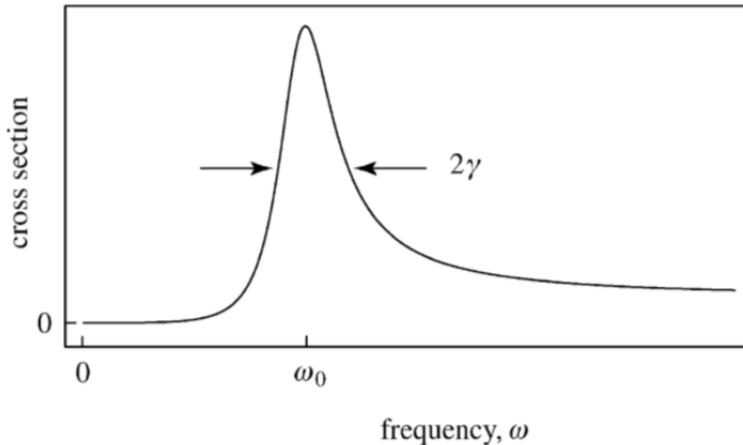


Fig. 6.6

Scattering cross section for the Lorentz model.

6.2 Brillouin Scattering

6.2.1 Spontaneous Brillouin scattering

We start with the equation describing the acoustic motion of a pressure wave Δp :

$$\frac{\partial^2 \Delta p}{\partial t^2} - \Gamma' \nabla^2 \frac{\partial \Delta p}{\partial t} - v^2 \nabla^2 \Delta p = 0. \quad (6.23)$$

Here Γ' is a damping parameter and v is the sound velocity given with ¹:

$$v^2 = \left(\frac{\partial p}{\partial \rho} \right), \quad (6.24)$$

with ρ being the medium density.

Let us consider a solution for a pressure wave of the form:

$$\Delta p = \Delta p_0 e^{i(qz - \Omega t)} + c.c. \quad (6.25)$$

Substituting this solution to the acoustic wave equation Eq.6.23 we can find the dispersion relation:

$$\Omega^2 = q^2(v^2 - i\Omega\Gamma') \rightarrow q^2 = \frac{\Omega^2/v^2}{1 - i\Omega\Gamma'/v^2} \simeq \frac{\Omega^2}{v^2} \left(1 + i \frac{\Omega\Gamma'}{v^2} \right), \quad (6.26)$$

where in the last step we assume that $\Omega\Gamma'/v^2 \ll 1$.

Now:

$$q \simeq \frac{\Omega}{v} \sqrt{1 + i \frac{\Omega\Gamma'}{v^2}} \simeq \frac{\Omega}{v} \left(1 + i \frac{\Omega\Gamma'}{2v^2} \right) = \frac{\Omega}{v} + i \frac{\Gamma}{2v}, \quad (6.27)$$

where $\Gamma = \Gamma' \left(\frac{\Omega}{v} \right)^2$ is the phonon decay rate. Assuming that the magnitude of q is approximately the magnitude of its real part, such that $|q| = \Omega/v$, we get:

$$\Gamma = \Gamma' |q|^2. \quad (6.28)$$

We can thus write the amplitude of the acoustic wave as:

$$\Delta p = \Delta p_0 e^{i\frac{\Omega}{v} z} e^{-\frac{\Gamma}{2v} z} e^{-i\Omega t} + c.c. \triangleq \Delta p(z) e^{-i\Omega t} + c.c. \quad (6.29)$$

so:

$$\Delta p(z) = \Delta p_0 e^{i\frac{\Omega}{v} z} e^{-\frac{\Gamma}{2v} z}. \quad (6.30)$$

The intensity of the acoustic wave varies with:

¹ Notice the units: $[P] = [F]/m^2$; $[F] = kg \cdot m/s^2$; $[\rho] = kg/m^3$.

$$|\Delta p(z)|^2 = |\Delta p_0|^2 e^{-\alpha_s z}, \quad (6.31)$$

where:

$$\alpha_s = \frac{\Gamma}{v} \quad (6.32)$$

is the *sound absorption coefficient*.

We define the phonon lifetime as the time it takes the phonon intensity to decay to $\exp(-1)$ of its original intensity:

$$\tau_p = \frac{1/\alpha_s}{v} = \frac{1}{\Gamma}. \quad (6.33)$$

The pressure wave can cause a change in the index of refraction (or dielectric constant) of the material as follows: Generally, the dielectric constant can change with either the temperature or the material density ρ . That is, we can represent a change $\Delta\varepsilon$ in the dielectric constant $\varepsilon = n^2$ with:

$$\Delta\varepsilon = \left(\frac{\partial\varepsilon}{\partial\rho} \right)_T \Delta\rho + \left(\frac{\partial\varepsilon}{\partial T} \right)_\rho \Delta T \quad (6.34)$$

The density itself can change due to changes in the pressure of the material, but also due to changes in the entropy S of the material (which by itself can change due to changes in the energy stored in the material or in its volume while keeping the pressure constant):

$$\Delta\rho = \left(\frac{\partial\rho}{\partial p} \right)_S \Delta p + \left(\frac{\partial\rho}{\partial S} \right)_p \Delta S. \quad (6.35)$$

Acoustic waves do not change the entropy and so for them we can write:

$$\Delta\rho = \left(\frac{\partial\rho}{\partial p} \right) \Delta p. \quad (6.36)$$

And we drop the S subscript. Together with Eq.6.34 while setting $\Delta T = 0$ as acoustic waves happen on time scales much faster than temperature fluctuations, we get :

$$\Delta\varepsilon = \left(\frac{\partial\varepsilon}{\partial\rho} \right) \left(\frac{\partial\rho}{\partial p} \right) \Delta p, \quad (6.37)$$

where we have dropped the T subscript.

We use the following definitions:

$$C_s = \frac{1}{\rho} \left(\frac{\partial\rho}{\partial p} \right)_S \quad (6.38)$$

for the *compressibility* at constant entropy, and:

$$\gamma_e = \left(\rho \frac{\partial \varepsilon}{\partial \rho} \right)_{\rho=\rho_0} \quad (6.39)$$

as the *electrostrictive constant*.

With these we can write either :

$$\Delta \varepsilon = \gamma_e \frac{\Delta \rho}{\rho_0}, \quad (6.40)$$

where we used Eq.6.36, or

$$\Delta \varepsilon = \gamma_e C_s \Delta p. \quad (6.41)$$

Now, as the dielectric constant is related to the susceptibility via $\varepsilon = 1 + \chi$, then changes (fluctuations included) in the dielectric constant are exactly changes in the susceptibility: $\Delta \chi = \Delta \varepsilon$. Assuming an instantaneous response of the optical properties of the material to the acoustic wave, the polarization developing in the medium due to this added modulation would be:

$$P = \varepsilon_0 \Delta \chi E = \varepsilon_0 \Delta \varepsilon E = \varepsilon_0 \gamma_e C_s \Delta p E_{in}. \quad (6.42)$$

Here E_{in} is the incident field. The pressure modulation component Δp that we take is a generalization of Eq.6.25 from a wave that propagates in the z direction to a wave propagating in a general direction in space:

$$\Delta p(\mathbf{r}, t) = \Delta p_0 e^{i(\mathbf{q} \cdot \mathbf{r} - \Omega t)} + c.c. \quad (6.43)$$

By substituting this pressure wave to the polarization Eq.6.42 using the standard wave equation Eq.1.1 and assuming the incident wave is a simple plane wave $E_{in} = E_0 e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + c.c$ we get:

$$\begin{aligned} \nabla^2 \mathbf{E} - \frac{n^2}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = & - \frac{\gamma_e C_s}{c^2} [(\omega - \Omega)^2 E_0 \Delta p^* e^{i(\mathbf{k} - \mathbf{q}) \cdot \mathbf{r} - i(\omega - \Omega)t} \\ & + (\omega + \Omega)^2 E_0 \Delta p e^{i(\mathbf{k} + \mathbf{q}) \cdot \mathbf{r} - i(\omega + \Omega)t} + c.c]. \end{aligned} \quad (6.44)$$

On the RHS we have two components - at frequency $\omega - \Omega$ which is the Stokes scattering component, and at frequency $\omega + \Omega$ which is the Anti-Stokes scattering component. So both processes leading to Stokes or anti-Stokes can happen at the same time. However, depending on phase matching conditions - over extended length scales - only one of them would be efficient.

We would like to note that the components of the acoustic wave can also be written using:

$$e^{i(\mathbf{q} \cdot \mathbf{r} - \Omega t)} = e^{i|\mathbf{q}|(\hat{\mathbf{q}} \cdot \mathbf{r} - \frac{\Omega}{|\mathbf{q}|} t)} = e^{i|\mathbf{q}| \hat{\mathbf{q}} \cdot (\mathbf{r} - \hat{\mathbf{q}} v t)}, \quad (6.45)$$

where $v = \frac{\Omega}{|\mathbf{q}|}$ is the velocity of the acoustic wave, directed in the $\hat{\mathbf{q}}$ direction.

Brillouin Stokes Scattering

We consider the first polarization term on the RHS of Eq.6.44 which leads to radiation having a wavevector:

$$\mathbf{k}' = \mathbf{k} - \mathbf{q}, \quad (6.46)$$

and having angular frequency of:

$$\omega' = \omega - \Omega. \quad (6.47)$$

These two relations are the phase-matching, or momentum and energy matching conditions of the scattering process. They must be fulfilled in order for the interaction to be efficient. Notice that Eq.6.46 is a Bragg scattering condition.

The momentum and energy of the incoming wave obeys the dispersion relation for the radiation in the medium:

$$|\mathbf{k}| = \frac{\omega}{c} n, \quad (6.48)$$

Similarly for the outgoing electromagnetic wave:

$$|\mathbf{k}'| = \frac{\omega'}{c} n, \quad (6.49)$$

while the acoustic wave obeys its own dispersion relation, which we can approximate (neglecting the imaginary part, see Eq.6.27):

$$|\mathbf{q}| = \frac{\Omega}{v}. \quad (6.50)$$

If we consider scattering at a specific angle θ , the momentum matching conditions is given graphically in Fig.6.7(b). Generally the acoustic frequency is much smaller than the optical frequency: $\Omega \ll \omega$ while of course $c \gg v$ which together leads to $|\mathbf{k}| \gg |\mathbf{q}|$ hence $|\mathbf{k}| \simeq |\mathbf{k}'|$. Because of that we can estimate safely (using Fig.6.7(b)) that:

$$|\mathbf{q}| = 2|\mathbf{k}| \sin(\theta/2) \quad (6.51)$$

Using the last result in the acoustic dispersion relation we get:

$$\Omega = 2|\mathbf{k}|v \sin(\theta/2) = 2n\omega \frac{v}{c} \sin(\theta/2) \quad (6.52)$$

For forward scattering $\theta = 0$ it seems that $\Omega = 0$. This actually means that there is no acoustic wave that couples an incoming wave to an outgoing wave at the same direction and frequency, put otherwise - "forward scattering" cannot be the result of scattering from an acoustic wave. For backward scattering we have the maximal possible value of the stokes shift:

$$\Omega_{max} = 2n \frac{v}{c} \omega. \quad (6.53)$$

Typical values for Ω_{max} are on the order of GHz.

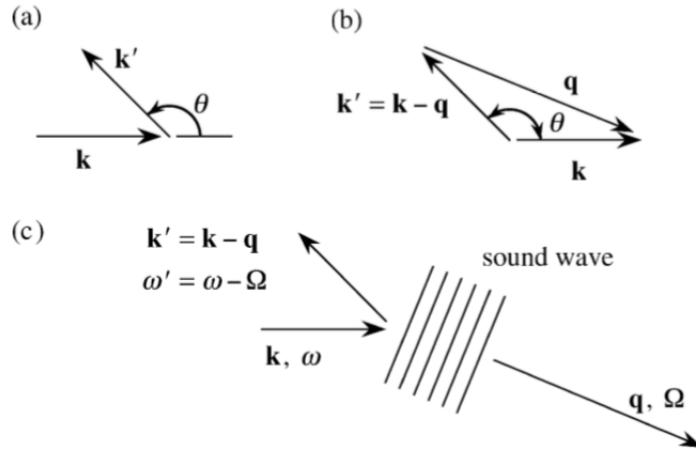


Fig. 6.7

Brillouin Stokes Scattering.

Brillouin Anti-Stokes Scattering

The difference from stokes scattering is that the energy of the scattered wave is blue-shifted (that is - shifted to higher frequencies). This entails the following phase matching relations:

$$\mathbf{k}' = \mathbf{k} + \mathbf{q}, \quad (6.54)$$

and

$$\omega' = \omega + \Omega. \quad (6.55)$$

The dispersion relations do not change, which again lead to the same expression for the frequency shift:

$$\Omega = 2|\mathbf{k}|v \sin(\theta/2) = 2n\omega \frac{v}{c} \sin(\theta/2) \quad (6.56)$$

The process is shown schematically in Fig.6.8.

At this point we would like to remind that actually the dispersion relation for the acoustic wave is a complex function (see Eq.6.27):

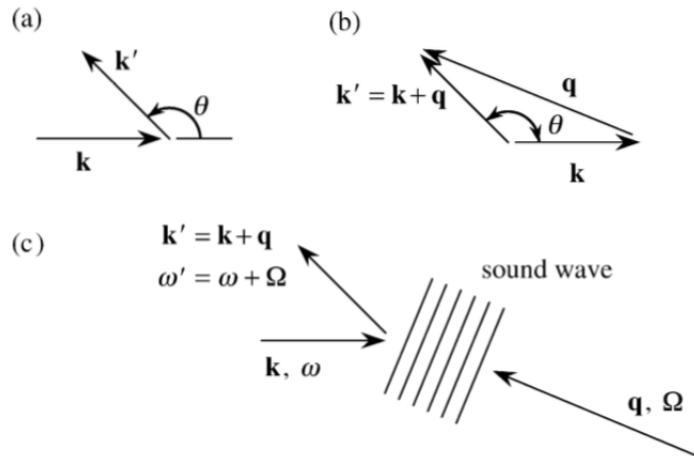


Fig. 6.8
Brillouin anti-Stokes Scattering.

$$q = \frac{\Omega}{v} + i \frac{\Gamma}{2v}, \quad (6.57)$$

for which the imaginary part is related to the decay of the acoustic wave characterized with life time (see Eq.6.33 and Eq.6.28):

$$\tau_p = \frac{1}{\Gamma} = \frac{1}{\Gamma' |q|^2}. \quad (6.58)$$

Using the time-energy uncertainty principle, this finite life time results in a finite spread in angular frequency for the acoustic wave:

$$\delta\omega = \frac{1}{\tau_p} = \Gamma' |q|^2. \quad (6.59)$$

Using Eq.6.51 we get:

$$\delta\omega = 4\Gamma' |\mathbf{k}|^2 \sin^2(\theta/2). \quad (6.60)$$

This frequency spread is usually on the order of $100MHz$.

The spectrum of both Stokes and anti-Stokes components of the Brillouin scattering, together with Rayleigh scattering (due to scattering from non-propagating density fluctuations) is shown schematically in Fig.6.9.

6.2.2 Acoustooptics devices

Scattering of light from an acoustic (sound) wave can be used to realize devices that control the intensity, frequency, or direction of propagation of light. Here the

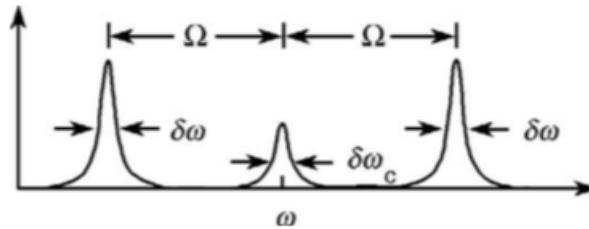


Fig. 6.9

Brillouin (Stokes and anti-Stokes) and Rayleigh Scattering.

scattering is from an ordered grating which is induced acoustically. Such a scattering is also known as diffraction.

Bragg scattering of light by sound waves

We now formulate Bragg scattering in more details, finding how the fields are coupled and exchange energy through the coupling acoustic wave. A scheme for a general acoustooptic modulator is shown in Fig.6.10, which shows the nomenclature we would be using in this section. In this figure the direction of the velocity of the acoustic wave, which is also the direction of the \mathbf{q} vector, is towards (against) the incident beam \mathbf{k}_1 which matches the case of anti-Stokes scattering as shown in Fig.6.8, that is:

$$\mathbf{k}_2 = \mathbf{k}_1 + \mathbf{q}; \quad \omega_2 = \omega_1 + \Omega. \quad (6.61)$$

If the direction of the acoustic wave velocity is switched, we are in the Stokes scattering case (Fig.6.7) for which:

$$\mathbf{k}_2 = \mathbf{k}_1 - \mathbf{q}; \quad \omega_2 = \omega_1 - \Omega. \quad (6.62)$$

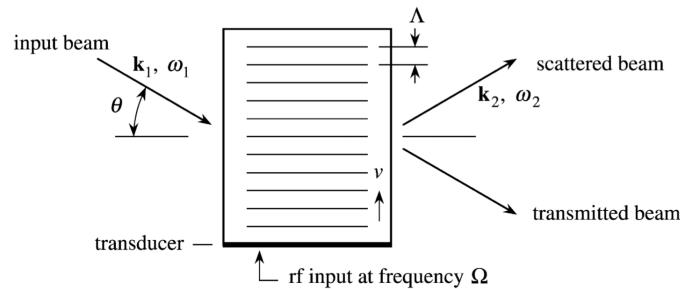


Fig. 6.10

An acoustooptic modulator. The transducer induced acoustic waves at RF frequencies.

We assume here a very simple case of a scalar equation in the field (all polarizations are the same and we would ignore the tensor nature of the dielectric constant) such that the total electric field $E = E_1 + E_2$ is comprised from the superposition of incident field:

$$E_1 = A_1 e^{i(\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)} + c.c. \quad (6.63)$$

with the diffracted field:

$$E_2 = A_2 e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)} + c.c. \quad (6.64)$$

We would assume anti-Stokes scattering $\omega_2 = \omega_1 + \Omega$ and that the interaction is nearly phase-matched:

$$\mathbf{k}_2 - \mathbf{k}_1 - \mathbf{q} = -\Delta \mathbf{k}, \quad (6.65)$$

where $\Delta \mathbf{k}$ is quite small, and the minus sign is a matter of convention. The acoustooptic modulation changes the dielectric constant as

$$\Delta \varepsilon = \Delta \varepsilon_0 e^{i(\mathbf{q} \cdot \mathbf{r} - \Omega t)} + c.c., \quad (6.66)$$

where (see Eq.6.40) $\Delta \varepsilon = \gamma_e \Delta \rho / \rho_0$.

The electric field $E = E_1 + E_2$ needs to satisfy the wave equation:

$$\nabla^2 E - \frac{n^2 + \Delta \varepsilon}{c^2} \frac{\partial^2 E}{\partial t^2} = 0, \quad (6.67)$$

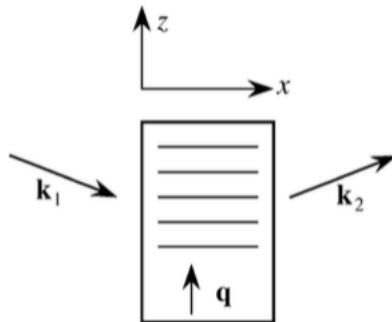


Fig. 6.11

Geometry chosen for describing the Bragg-type acoustooptic modulator.

where n is the index of refraction in the absence of the modulation induced by the acoustic wave. We assume that the interaction takes place within a plane (x, z) while $\mathbf{q} = q\hat{z}$ as shown in Fig.6.11. If we substitute the field into the last wave

equation, we find that all the terms oscillating at frequency ω_1 obey the following equation:

$$\frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial z^2} + 2ik_{1x} \frac{\partial A_1}{\partial x} + 2ik_{1z} \frac{\partial A_1}{\partial z} - (k_{1x}^2 + k_{1z}^2)A_1 + \frac{n^2\omega_1^2}{c^2}A_1 + \frac{\omega_2^2}{c^2}A_2\Delta\varepsilon_0^*e^{i(\mathbf{k}_2-\mathbf{k}_1-\mathbf{q})\cdot\mathbf{r}} = 0. \quad (6.68)$$

Now let us assume that the geometry is very long in the z direction and that all exchange of energy between the two fields takes place along x . This means that we neglect any variation in the z direction - so we can drop all the derivatives along z , and we require perfect phase matching along z as well, which leaves us with phase mismatch only along x :

$$(\mathbf{k}_2 - \mathbf{k}_1 - \mathbf{q}) \cdot \mathbf{r} \equiv -\Delta kx. \quad (6.69)$$

If we further employ the SVEA (see Eq.2.7) along the x direction - we can neglect the second order derivative in x . All of these leave us with a much simpler equation:

$$\frac{dA_1}{dx} = \frac{i\omega_2^2\Delta\varepsilon_0^*}{2k_{1x}c^2}A_2e^{-i\Delta kx} \quad (6.70)$$

where we have used $k_{1x}^2 + k_{1z}^2 = n^2\omega_1^2/c^2$.

A similar procedure would yield the coupled equation:

$$\frac{dA_2}{dx} = \frac{i\omega_1^2\Delta\varepsilon_0}{2k_{2x}c^2}A_1e^{i\Delta kx} \quad (6.71)$$

We can assume that the prefactor in the two coupled equations are about the same (for this we approximate in the prefactors $\omega_1 \simeq \omega_2 \equiv \omega$ and $k_{1x} \simeq k_{2x} \equiv k_x$). So with the coupling coefficient defined as :

$$\kappa = \frac{\omega^2\Delta\varepsilon^*}{2k_xc^2} \quad (6.72)$$

we finally get:

$$\frac{dA_1}{dx} = i\kappa A_2 e^{-i\Delta kx} \quad (6.73)$$

$$\frac{dA_2}{dx} = i\kappa^* A_1 e^{i\Delta kx} \quad (6.74)$$

We already studied a very similar set of coupled wave equations in Section 2.5 when we studied SFG in the strong pump and weak signal approximation (see Eq.2.44-2.45). So we can use the same methods to solve the equations given here.

We note that here the trivial case for which $\Delta k = 0$ the solution is:

$$A_1(x) = A_1(0) \cos(|\kappa|x), \quad (6.75)$$

$$A_2(x) = \frac{i\kappa^*}{|\kappa|} A_1(0) \sin(|\kappa|x), \quad (6.76)$$

which satisfy energy conservation:

$$|A_1(x)|^2 + |A_2(x)|^2 = |A_1(0)|^2 \quad (6.77)$$

We note that this conservation is only approximated as we assumed that $\Omega \ll \omega$.

6.2.3 Stimulated Brillouin scattering

When the intensity of incoming light gets high enough, nonlinear effects come into play which can cause stimulated scattering.

For *Stimulated Brillouin Scattering* (SBS) we can have the following scenario (depicted in Fig.6.12): Let us assume that the incoming light at frequency ω_L is propagating in the same direction of a sound wave with frequency Ω , so the scattering process would be a Stokes type and the scattered light would be down-shifted in frequency $\omega_s = \omega_L - \Omega$. Now - the interference of the two light beams beat at the difference frequency $\omega_L - \omega_s = \Omega$ which can be used as an additional driving term to the acoustic wave, which in turn strengthens the scattered Stokes wave and so on - we get a positive feedback loop which can lead to an exponential gain in the scattering.

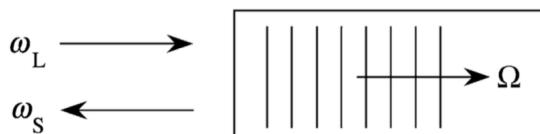


Fig. 6.12 Stokes component generation in Stimulated Brillouin Scattering.

Electrostriction

SBS can be mediated through electrostriction - the tendency of a material to become denser in places with high optical intensity. This mechanism acts as the coupling between optical and acoustic frequencies required for SBS.

The basic idea of electrostriction is that the potential energy *density* of a material is increasing with the square of the electric field:

$$u = \frac{1}{2} \varepsilon \varepsilon_0 E^2. \quad (6.78)$$

This means that if we move for example a piece of material towards the gap of a parallel plate capacitor as shown in Fig.6.13(a) then the energy of the material is increasing. Naturally, when we have a case in which the energy of a given body is changing as a function of coordinate - we have a force acting on that body.

Similarly for a single molecule with polarization $\mathbf{p} = \epsilon_0 \alpha \mathbf{E}$ with α being the molecular polarizability, the potential energy associated with an applied field is the one associated with building the polarization:

$$U = - \int_0^{\mathbf{E}} \mathbf{p} \cdot d\mathbf{E}' = - \frac{1}{2} \epsilon_0 \alpha \mathbf{E} \cdot \mathbf{E} = - \frac{1}{2} \epsilon_0 \alpha E^2. \quad (6.79)$$

And the force is simply the gradient of the potential energy:

$$\mathbf{F} = -\nabla U = \frac{1}{2} \epsilon_0 \alpha \nabla(E^2). \quad (6.80)$$

So - the molecule is attracted to regions with an increased electric field.

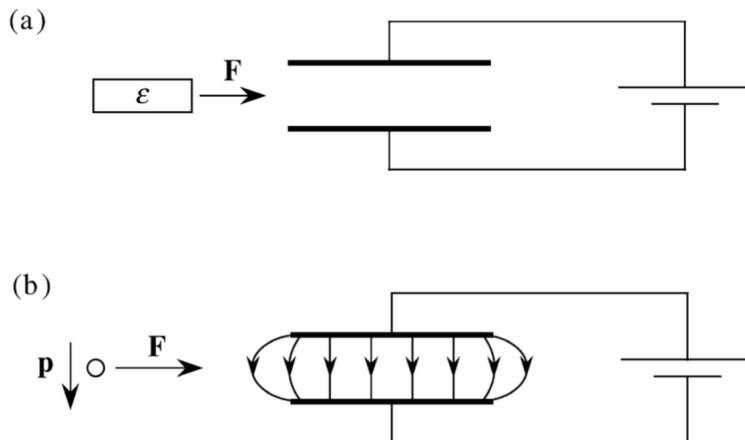


Fig. 6.13

Electrostriction. (a) A material with a given dielectric constant ϵ experience force \mathbf{F} towards regions in space with a higher value of the electric field (b) The same is applicable for a single molecule with a given polarization \mathbf{p} .

More generally, for a given material when it is radiated with an inhomogeneous electric field, the molecules (or atoms) would be attracted to regions with high intensity, increasing the density of the material at those regions by an amount $\Delta\rho$. This would then increase the dielectric constant $\epsilon \rightarrow \epsilon + \Delta\epsilon$ with (see Eq.6.34):

$$\Delta\epsilon = \left(\frac{\partial\epsilon}{\partial\rho} \right) \Delta\rho. \quad (6.81)$$

This changes the field energy density:

$$\Delta u = \frac{1}{2} \varepsilon_0 E^2 \Delta \varepsilon = \frac{1}{2} \varepsilon_0 E^2 \left(\frac{\partial \varepsilon}{\partial \rho} \right) \Delta \rho. \quad (6.82)$$

From Thermodynamics we know that the change in energy when a material is compressed is equal to work (density) done:

$$\Delta w = -p_{st} \frac{\Delta V}{V} = -p_{st} \frac{\Delta \rho}{\rho}, \quad (6.83)$$

where p_{st} is called the *strictive pressure*, which is the pressure due to the action of the electric field.

(Notice above that we consider work density - so we have work $-p_{st}\Delta V$ divided by the volume V .)

From the last two equations we get:

$$p_{st} = -\frac{1}{2} \varepsilon_0 \rho \left(\frac{\partial \varepsilon}{\partial \rho} \right) E^2 = -\frac{1}{2} \varepsilon_0 \gamma_e E^2, \quad (6.84)$$

where $\gamma_e = \rho(\partial \varepsilon / \partial \rho)$ is known as the *electrostrictive constant*. As p_{st} is negative, the pressure drops in regions with a high field strength.

Now if we would like to calculate the change in density $\Delta \rho$ by using:

$$\Delta \rho = -\frac{\partial \rho}{\partial p} \Delta p \quad (6.85)$$

We assume that the change in pressure Δp is solely due to the electrostrictive pressure: $\Delta p = p_{st}$. If we further use the compressibility (see Eq.6.38) $C = \rho^{-1}(\partial \rho / \partial p)$ we get that:

$$\Delta \rho = \frac{1}{2} \varepsilon_0 \rho C \gamma_e E^2. \quad (6.86)$$

In the last equation the field E is some constant. However, for the application of an optical field $\mathbf{E}(t)$ we need to replace E^2 above with the average $\langle \mathbf{E} \cdot \mathbf{E} \rangle$:

$$\Delta \rho = \frac{1}{2} \varepsilon_0 \rho C \gamma_e \langle \mathbf{E} \cdot \mathbf{E} \rangle. \quad (6.87)$$

The change in the susceptibility is:

$$\Delta \chi = \Delta \varepsilon = \frac{\partial \varepsilon}{\partial \rho} \Delta \rho = \frac{1}{2} \varepsilon_0 C \gamma_e^2 \langle \mathbf{E} \cdot \mathbf{E} \rangle. \quad (6.88)$$

If the field is a CW:

$$\mathbf{E}(t) = \mathbf{E}_0 e^{-i\omega t} + c.c. \quad (6.89)$$

then it can be easily shown that $\langle \mathbf{E} \cdot \mathbf{E} \rangle = 2|\mathbf{E}_0|^2$. The nonlinear polarization component due to $\Delta\chi$, oscillating at the frequency ω , is: $\mathbf{P} = \Delta\chi\mathbf{E}_0$:

$$\mathbf{P} = \varepsilon_0 C \gamma_e^2 |\mathbf{E}_0|^2 \mathbf{E}_0 \quad (6.90)$$

which can also be written as:

$$\mathbf{P} = 3\varepsilon_0 \chi^{(3)}(\omega = \omega + \omega - \omega) |\mathbf{E}_0|^2 \mathbf{E}_0, \quad (6.91)$$

with $\chi^{(3)}(\omega = \omega + \omega - \omega) = \frac{1}{3}\varepsilon_0 C \gamma_e^2$. Notice that the factor of 3 is required to agree with the form of the 3rd order nonlinear susceptibility given in Eq.3.5.

This shows that the electrostriction effect is a Kerr-type 3rd order process.

Electrostriction-induced Stimulated Brillouin Scattering - SBS Amplifier

Here we consider how the light is used as a source term in the acoustic wave equation, while at the same time the acoustic wave is being used as a source term in the optical wave equation. The coupling between the two equations is provided by electrostriction.

We start with the acoustic wave equation (see Eq.6.23) together with the relation between the density ρ and the pressure p (Eq.6.36) to formulate an acoustic wave equation for the density:²

$$\frac{\partial^2 \rho}{\partial t^2} - \Gamma' \nabla^2 \frac{\partial \rho}{\partial t} - v^2 \nabla^2 \rho = \nabla \cdot \mathbf{f}. \quad (6.92)$$

\mathbf{f} appearing in the source term on the RHS is the force per unit volume which is also the gradient of the strictive pressure p_{st} (see Eq.6.83):

$$\mathbf{f} = \nabla p_{st}, \quad (6.93)$$

with (see Eq.6.84 and remark before Eq.6.87):

$$p_{st} = -\frac{1}{2} \varepsilon_0 \gamma_e \langle \mathbf{E} \cdot \mathbf{E} \rangle. \quad (6.94)$$

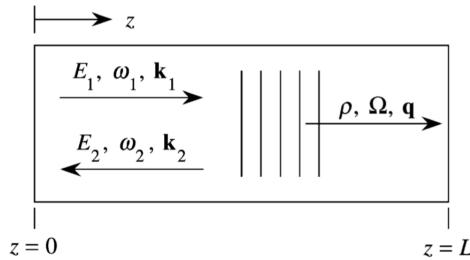
To make things simple we would assume a 1D problem - all waves, optical and acoustic, are propagating along z . In addition we assume a Stokes-type interaction for which the optical beams are counter-propagating and the acoustic wave as depicted in Fig.6.14 (see also Fig.6.12).

The phase matching conditions in this case are (see also Fig.6.7):

$$\omega_2 = \omega_1 - \Omega \quad (6.95)$$

and

² we replace also $\Delta\rho$ with ρ and Δp with p .

**Fig. 6.14**

Stokes Stimulated Brillouin Scattering.

$$k_2 = k_1 - q. \quad (6.96)$$

As $\Omega \ll \omega_1, \omega_2$ we would assume that $|k_2| \simeq |k_1|$, using the fact that in such interactions they are also counter propagating $k_1 = -k_2$ we get that $q = 2k_1$.

We remind that the acoustic wave obeys the dispersion relation:

$$\Omega = qv. \quad (6.97)$$

while the light obeys the dispersion relation:

$$|k_i| = \frac{n\omega_i}{c} \quad (6.98)$$

where we assume that $n_1 = n_2 = n$.

We can use the last four equations to show that the acoustic frequency, for the case of perfect phase matching (in momentum and energy) can be described in terms of the frequency ω_1 :

$$\begin{aligned} \Omega|_{Perfect\ Phase\ Matching} &\triangleq \Omega_B = \frac{v}{c/n}(\omega_1 + \omega_2) = \\ &= \frac{\frac{2v}{c/n}\omega_1}{1 + \frac{v}{c/n}} = \frac{2v}{c/n}\omega_1. \end{aligned} \quad (6.99)$$

where at the last step we approximated $v/c \ll 1$ (we also used Eq.6.95 with $\Omega = \Omega_B$ to eliminate ω_2 from the expression).

We would consider now a case in which the ω_2 is imposed externally as a seed signal - forcing an acoustic wave Ω which might be different than Ω_B .

In this case we assume that the acoustic wave which is the solution to Eq.6.92 is of the form:

$$\rho(z, t) = \rho_0 + [\rho_A(z, t)e^{i(qz - \Omega t)} + c.c], \quad (6.100)$$

where ρ_0 is the medium mean density, and ρ_A is the amplitude of the perturbation of the acoustic wave.

The total optical field in the medium is the superposition of two terms:

$$\begin{aligned} E(z, t) &= E_1(z, t) + E_2(z, t) \\ &= A_1(z, t)e^{i(k_1 z - \omega_1 t)} + A_2(z, t)e^{i(-k_2 z - \omega_2 t)} + c.c. \end{aligned} \quad (6.101)$$

which are coupled by the acoustic wave.

These two fields, used with Eq.6.93-6.94 gives the following source term for the acoustic equation:

$$\nabla \cdot \mathbf{f} = \varepsilon_0 \gamma_e q^2 \left[A_1 A_2^* e^{i(qz - \Omega t)} + c.c. \right]. \quad (6.102)$$

If we substitute Eq.6.100 and Eq.6.102 back to the acoustic wave equation Eq.6.92 and assume SVEA for the acoustic amplitude we get:

$$-2i\Omega \frac{\partial \rho_A}{\partial t} + (\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B)\rho_A - 2iqv^2 \frac{\partial \rho_A}{\partial z} = \varepsilon_0 \gamma_e q^2 A_1 A_2^* \quad (6.103)$$

Here $\Gamma_B = q^2 \Gamma'$ is the *Brillouin linewidth* which is inversely proportional to the phonon lifetime: $\tau_p = \Gamma_B^{-1}$.

Usually when treating this equation it is customary to drop the term containing $\frac{\partial \rho_A}{\partial z}$ which states that the change in amplitude of the acoustic wave is negligible compared to the changes in amplitude of the optical waves³.

So - dropping the spatial derivative of the acoustic wave we would look for the steady state solution (that is - we also set to zero the temporal derivative) to get:

$$\rho_A(z, t) = \varepsilon_0 \gamma_e q^2 \frac{A_1(z, t) A_2^*(z, t)}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B}. \quad (6.104)$$

Now we can turn to the evolution of the optical fields, which are governed by the usual wave equations:

$$\frac{\partial^2 E_i}{\partial z^2} - \frac{n^2}{c^2} \frac{\partial^2 E_i}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P_i}{\partial t^2}; \quad i = 1, 2 \quad (6.105)$$

The total nonlinear polarization, assuming instantaneous response of the acoustic wave is:

$$P = \varepsilon_0 \Delta \chi E = \varepsilon_0 \Delta \varepsilon E = \varepsilon_0 \gamma_e \rho_0^{-1} \rho E. \quad (6.106)$$

where ρ is the change in density ($\Delta \rho$ from Eq.6.87⁴) which is proportional to the square of the field.

³ Notice that in Boyd's book there is a different argument - that the phonons actually are damped very fast - on micron scale. This statement is problematic since it suggests that the variations in ρ_A are actually very fast and we cannot neglect the term under discussion

⁴ In that equation ρ takes the role of ρ_0 here.

The polarization field includes components oscillating at different frequencies. We choose the two components which are at the frequencies of the drive (ω_1) component:

$$P_1 = p_1 e^{i(k_1 z - \omega_1 t)} + c.c. \quad (6.107)$$

and the Stokes component (ω_2):

$$P_2 = p_2 e^{i(k_2 z - \omega_2 t)} + c.c. \quad (6.108)$$

Using the form of E (Eq.6.101) and ρ (Eq.6.100 in Eq.6.106 we find that:

$$p_1 = \varepsilon_0 \gamma_e \rho_0^{-1} \rho_A A_2, \quad (6.109)$$

and

$$p_2 = \varepsilon_0 \gamma_e \rho_0^{-1} \rho_A^* A_1. \quad (6.110)$$

Substituting the polarizations and the amplitude of the acoustic wave Eq.6.104 into the wave equations Eq.6.105 and also using an SVEA we finally get:

$$\frac{\partial A_1}{\partial z} + \frac{n}{c} \frac{\partial A_1}{\partial t} = \frac{i\omega \gamma_e}{2nc\rho_0} \rho_A A_2, \quad (6.111)$$

and

$$-\frac{\partial A_2}{\partial z} + \frac{n}{c} \frac{\partial A_2}{\partial t} = \frac{i\omega \gamma_e}{2nc\rho_0} \rho_A^* A_1. \quad (6.112)$$

For these two equations we approximated $\omega_1 \approx \omega_2 = \omega$.

For the steady state case we can drop the time derivative and use Eq.6.104 to get:

$$\frac{dA_1}{dz} = \frac{i\varepsilon_0 \omega q^2 \gamma_e^2}{2nc\rho_0} \frac{|A_2|^2 A_1}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B}, \quad (6.113)$$

$$\frac{dA_2}{dz} = -\frac{i\varepsilon_0 \omega q^2 \gamma_e^2}{2nc\rho_0} \frac{|A_1|^2 A_2}{\Omega_B^2 - \Omega^2 + i\Omega\Gamma_B}, \quad (6.114)$$

We can recast these equations in terms of intensities using $I_i = 2n\varepsilon_0 c |A_i|^2$ to give us:

$$\frac{dI_1}{dz} = -g I_1 I_2 \quad (6.115)$$

$$\frac{dI_2}{dz} = -g I_1 I_2 \quad (6.116)$$

where g , known as the *SBS gain factor* is a Lorentzian in the frequency Ω , given with:

$$g = g_0 \frac{(\Gamma_B/2)^2}{(\Omega_B - \Omega)^2 + (\Gamma_B/2)^2}, \quad (6.117)$$

with

$$g_0 = \frac{\gamma_e^2 \omega^2}{nvc^3 \rho_0 \Gamma_B}. \quad (6.118)$$

The resonance of the Lorentzian is at $\Omega = \Omega_B$ and its FWHM is Γ_B . Notice that Γ_B is itself proportional to ω^2 (because it is proportional to q^2 and q is proportional to ω) and so g_0 is independent of the laser frequency ω .

For the simple case in which the pump is undepleted $I_1 = \text{const}$ the solution to the Stokes wave injected to the medium at coordinate $z = L$ is:

$$I_2(z) = I_2(L) e^{gI_1(L-z)}, \quad (6.119)$$

that is - the Stokes wave grows exponentially as it propagates through the medium.

If we repeat the analysis given here for anti-Stokes scattering in which case ω_2 is the pump wave and $\omega_1 = \omega_2 + \Omega$ is the anti-Stokes wave, then it can be shown that the undepleted pump case for which $I_2(z) = \text{const}$ leads to the anti-Stokes wave to experience exponential attenuation $I_1(z) = I_1(0)e^{-gI_2 z}$.

For the nondepleted case we first notice from Eq.6.115-6.116 that:

$$\frac{dI_1}{dz} = \frac{dI_2}{dz} \rightarrow I_1(z) = I_2(z) + C, \quad (6.120)$$

where the constant C is dependent on the boundary conditions: $C = I_1(0) - I_2(0)$.

This situations is shown in Fig.6.15.

We can use the last result with Eq.6.116 to get:

$$\frac{dI_2}{I_2(I_2 + C)} = -gdz. \quad (6.121)$$

Integrating this equation we get:

$$\int_{I_2(0)}^{I_2(z)} \frac{dI_2}{I_2(I_2 + C)} = - \int_0^z g dz' \quad (6.122)$$

which is easily solved to yield:

$$\ln \left\{ \frac{I_2(z)[I_2(0) + C]}{I_2(0)[I_2(z) + C]} \right\} = -gCz. \quad (6.123)$$

Isolating $I_2(z)$ and substituting $C = I_1(0) - I_2(0)$ we get:

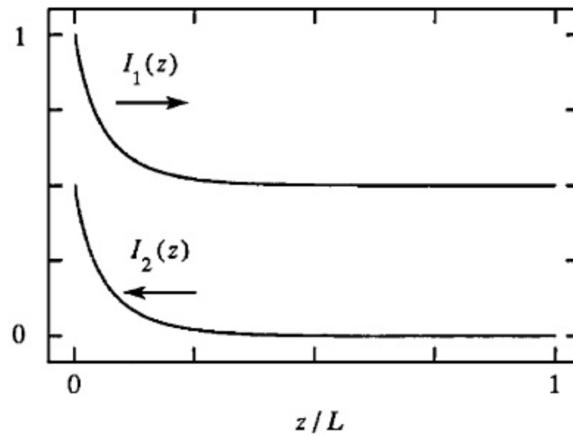


Fig. 6.15 Dynamics of the intensities for the SBS amplifier.

$$I_2(z) = \frac{I_2(0)[I_1(0) - I_2(0)]}{I_1(0)\exp\{gz[I_1(0) - I_2(0)]\} - I_2(0)}. \quad (6.124)$$

Of course from Eq.6.120 we can also find $I_1(z)$:

$$I_1(z) = I_2(z) + I_1(0) - I_2(0). \quad (6.125)$$

Now, notice that the configuration we analyze here involves an amplification through Stimulated Brillouin Scattering (and so it is called an SBS amplifier) in which the Stokes wave I_2 comes from the opposite side to the pump I_1 as depicted in Fig.6.16. This means that actually we know in advance $I_2(L)$ and not $I_2(0)$. We can find $I_2(0)$ using Eq.6.124 while substituting $I_2(z) = I_2(L)$. This can be solved numerically (the equation is transcendental) for finding $I_2(0)$.

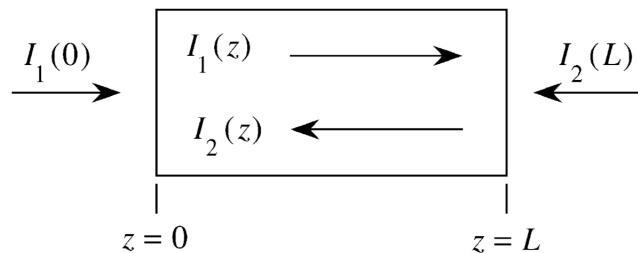


Fig. 6.16 Configuration for SBS amplifier.

The performance of the SBS amplifier is quantified through the intensity transfer defined as:

$$\frac{I_2(0) - I_2(L)}{I_1(0)} \quad (6.126)$$

as a function of the exponential gain experienced by a weak Stokes input, which is $G = gI_1(0)L$. Graphs for the intensity transfer as a function of G for various ratios of input intensities $I_2(L)/I_1(0)$ are shown in Fig.6.17. We see that large enough gain allows to transfer virtually all of the energy in I_1 to I_2 such that $I_2(0) - I_2(L) = I_1(0)$. Also - the threshold is lower when the Stokes seed is higher (that is - when $I_2(L)$ is higher).

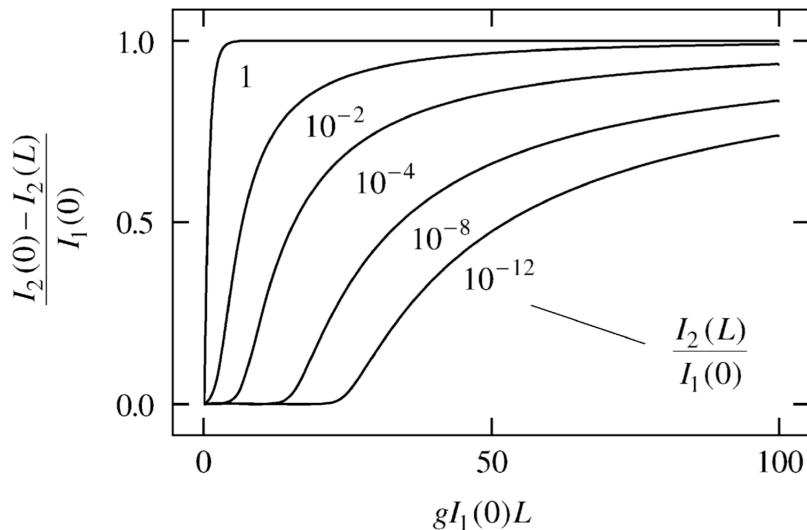


Fig. 6.17 Intensity transfer of an SBS amplifier.

Electrostriction-induced Stimulated Brillouin Scattering - SBS Generator

Here we consider the case in which there is no Stokes field seed which is injected externally. In this case, the SBS process is initiated by Stokes photons that are created by spontaneous Brillouin scattering. If we work in the parameter range which is below the gain threshold (before the abrupt rise in Fig.6.17) then we can assume that the transfer of energy from the pump to the Stokes wave is small such that we approximate $I_1(z) \approx \text{const}$. Then from Eq.6.116:

$$\frac{dI_2}{dz} = -gI_1(0)I_2 \rightarrow I_2(z) = I_2(0)e^{-gI_1(0)z}, \quad (6.127)$$

So:

$$I_2(L) = I_2(0)e^{-G}, \quad (6.128)$$

with $G = gI_1(0)L$. This of course can also be set as:

$$I_2(0) = I_2(L)e^G, \quad (6.129)$$

which better reflects the buildup of the Stokes component along the material.
The reflectivity of SBS is defined as:

$$R = \frac{I_2(0)}{I_1(0)}. \quad (6.130)$$

It is customary to define the SBS threshold as the case in which $R_{th} = 0.01$. Experiments set the values of G_{th} to be between 25 to 30.

For the general case in which $G > G_{th}$ we can no longer assume that $I_1(z)$ stays constant. For this reason we need to use the more general expression we already got Eq.6.124 by substituting $z = L$. With a little bit of algebra this equation can be cast in the form:

$$\frac{I_2(L)}{I_1(0)} = \frac{R(1-R)}{e^{G(1-R)} - R}. \quad (6.131)$$

We can also use Eq.6.125 at the boundary:

$$I_1(L) - I_2(L) = I_1(0) - I_2(0). \quad (6.132)$$

We can use the last two equations with the following approximations: $R \ll e^{G(1-R)}$ and $I_2(L)/I_1(L) \ll 1$ together with the approximation of $G_{th} = -\ln \{I_2(L)/I_1(L)\}$ to find the following expression:

$$\frac{G}{G_{th}} = \frac{(\ln R)/G_{th} + 1}{1 - R} \quad (6.133)$$

The reflectivity as a function of the gain can be calculated numerically from this equation, leading to the behavior shown in Fig.6.18. For $G < G_{th}$ there is essentially no reflectivity - no Stokes reflection, while for $G > G_{th}$ the reflectivity rises rapidly and then saturates.

6.3 Raman Scattering

Raman scattering as opposed to Rayleigh scattering is a non-parametric process, where the energy of the electron in the medium (a gas of molecules) does not stay the same after the process (see Fig.6.19). In Stokes Raman scattering the molecule is left at an excited state while the emitted photon has less energy than the pump

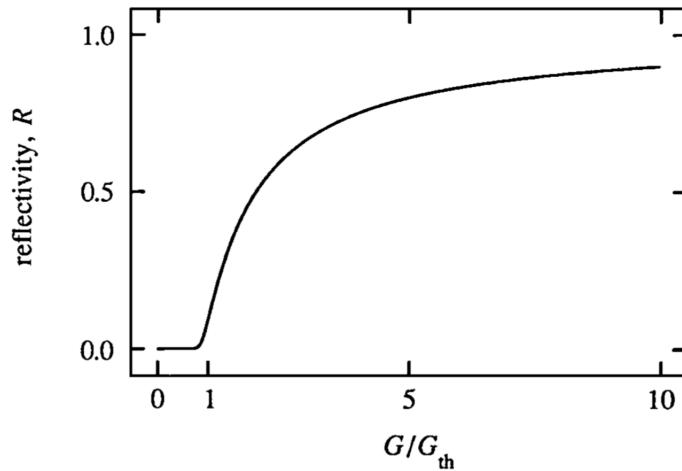


Fig. 6.18 SBS generator reflectivity vs weak-signal gain.

photon, while in the Anti-Stokes scattering the medium is left in a lower state while the emitted photon has higher energy than the pump photon. In spontaneous Raman scattering the Stokes component is much stronger because in usual thermal equilibrium conditions the population in the low state is larger than the population in the excited state - and so the probability for a process starting from this level is higher.

6.3.1 Stimulated Raman Scattering

Here we present a classical, scalar model for stimulated Raman scattering. In this model the incoming optical field interacts with a vibrational mode of a molecule where this mode is represented as an oscillator (see Fig. 6.20) with a displacement $q(t)$ from the equilibrium value q_0 . The equation of motion for the displacement is very similar to the Lorentz model:

$$\frac{d^2q(t)}{dt^2} + 2\gamma \frac{dq}{dt} + \omega_\nu^2 q(t) = \frac{F(t)}{m}, \quad (6.134)$$

where ω_ν is the resonance frequency and γ is a damping constant. The form of the driving force $F(t)$ would be different than what we have seen for the Lorentz model describing the movement of an electron due to the application of an optical field. Here the driving force is mediated through the *polarizability* α of the molecule which connects between the applied field and the induced polarization of the molecule:

$$\mathbf{p}(z, t) = \varepsilon_0 \alpha \mathbf{E}(z, t). \quad (6.135)$$

The dependency on the coordinate z allows us to analyze macroscopic effects.

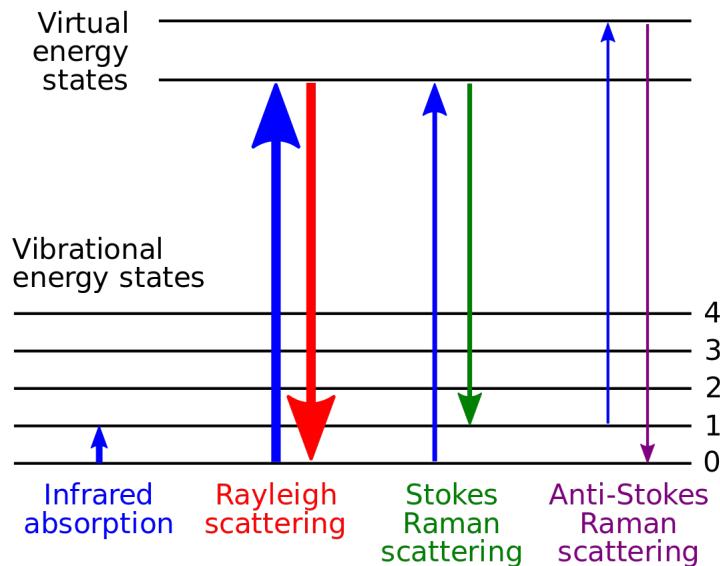


Fig. 6.19 Energy level transitions for various processes.

Also, for a density of N molecules per volume unit the dielectric constant is:

$$\varepsilon = 1 + N\alpha(t) \quad (6.136)$$

Now, to find the driving force we first note that the energy required to build the oscillation of the induced dipole moment is given with:

$$W = \frac{1}{2} \langle \mathbf{p}(z, t) \cdot \mathbf{E}(z, t) \rangle = \frac{1}{2} \varepsilon_0 \alpha \langle E^2(z, t) \rangle \quad (6.137)$$

where $\langle \cdot \rangle$ stands for time averaging.

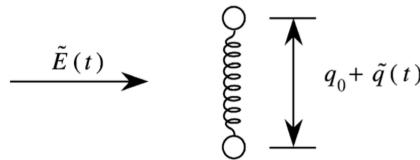


Fig. 6.20 Classical model for stimulated Raman scattering.

Now, it is easy to calculate the force. If we assume that the polarizability is to first order linear in the displacement:

$$\alpha(t) = \alpha_0 + \left(\frac{\partial \alpha}{\partial q} \right)_0 q(t) \quad (6.138)$$

then:

$$F(z, t) = \frac{dW}{dq} = \frac{\varepsilon_0}{2} \left(\frac{\partial \alpha}{\partial q} \right)_0 \langle E^2(z, t) \rangle. \quad (6.139)$$

Now, qualitatively there are two coupled processes in the medium: the driven oscillations of the molecule modulates in time the index of refraction of the material, leading to Stokes and anti-Stokes sidebands (see Fig.6.21(a)) while at the same time the time-beating of two frequencies set ω_ν , apart excites the molecular vibration.

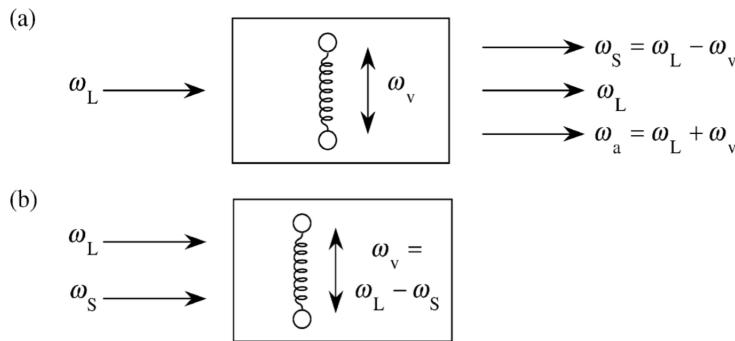


Fig. 6.21

Qualitatively description of SRS. (a) Modulation of the index of refraction leads to frequency side bands. (b) Intensity beating of two frequencies lead to oscillations (vibrations) of the molecule.

Let us analyze the interaction for the input laser field at frequency ω_L with the Stokes component oscillating at frequency $\omega_S = \omega_L - \omega_\nu$. (At the end we'll also consider the effect of the anti-Stokes component.) The total field is:

$$E(z, t) = A_L e^{i(k_L z - \omega_L t)} + A_S e^{i(k_S z - \omega_S t)} + c.c. \quad (6.140)$$

Using Eq.6.139 the force acting on the molecule is:

$$F(z, t) = \varepsilon_0 \left(\frac{\partial \alpha}{\partial q} \right)_0 \left[A_L A_S^* e^{i(Kz - \Omega t)} + c.c. \right], \quad (6.141)$$

where $K = k_L - k_S$ and $\Omega = \omega_L - \omega_S$.

We assume a solution to the oscillator equation in the form of:

$$q(z, t) = q(\Omega) e^{i(Kz - \Omega t)} + c.c. \quad (6.142)$$

Inserting this form of the solution and the form of the force Eq.6.141 into the oscillator equation of motion Eq.6.134 we get:

$$-\Omega^2 q(\Omega) - 2i\Omega\gamma q(\Omega) + \omega_\nu^2 q(\Omega) = \frac{\varepsilon_0}{m} \left(\frac{\partial \alpha}{\partial q} \right)_0 A_L A_S^*. \quad (6.143)$$

Solving for the amplitude $q(\Omega)$ we find:

$$q(\Omega) = \frac{(\varepsilon_0/m)(\partial \alpha / \partial q)_0 A_L A_S^*}{\omega_\nu^2 - \Omega^2 - 2i\Omega\gamma}. \quad (6.144)$$

The macroscopic polarization of the medium is (using Eq.6.135 and Eq.6.138):

$$\begin{aligned} P(z, t) &= N p(z, t) = \varepsilon_0 N \alpha(z, t) E(z, t) \\ &= \varepsilon_0 N \left[\alpha_0 + \left(\frac{\partial \alpha}{\partial q} \right)_0 q(z, t) \right] E(z, t) \\ &= P^{(1)} + P^{NL}, \end{aligned} \quad (6.145)$$

with

$$\begin{aligned} P^{NL}(z, t) &= \varepsilon_0 N \left(\frac{\partial \alpha}{\partial q} \right)_0 \left[q(\Omega) e^{i(Kz - \Omega t)} + c.c. \right] \\ &\quad \times \left[A_L e^{i(k_L z - \omega_L t)} + A_S e^{i(k_S z - \omega_S t)} + c.c. \right] \end{aligned} \quad (6.146)$$

We can see that the nonlinear polarization contains terms oscillating in various frequencies. Let us look at the term oscillating with frequency ω_S , which serves as the source term for the Stokes wave:

$$P_S^{NL}(z, t) = P(\omega_S) e^{-i\omega_S t} + c.c. \quad (6.147)$$

where

$$P(\omega_S) = \varepsilon_0 N \left(\frac{\partial \alpha}{\partial q} \right)_0 q^*(\Omega) A_L e^{ik_S z}. \quad (6.148)$$

Using the Stokes wave amplitude Eq.6.144 in the last equation we get:

$$P(\omega_S) = \frac{(\varepsilon_0^2 N/m)(\partial \alpha / \partial q)_0^2 |A_L|^2 A_S}{\omega_\nu^2 - \Omega^2 + 2i\Omega\gamma} e^{ik_S z}. \quad (6.149)$$

Looking at the amplitudes involved in this expression we see that this polarization has the form of a four-wave mixing process. This is made explicit by writing:

$$P(\omega_S) = 6\varepsilon_0 \chi_R(\omega_S) |A_L|^2 A_S e^{ik_S z}. \quad (6.150)$$

where the factor of 6 is the associated degeneracy factor and $\chi_R(\omega_S) = \chi_R(\omega_S = \omega_L - \Omega) = \chi_R(\omega_S = \omega_L - (\omega_L - \omega_S)) = \chi_R(\omega_S = \omega_S + \omega_L - \omega_L)$ which, with the last form, shows the susceptibility described solely by the optical fields.

The susceptibility can be written using the last two equations and $\Omega = \omega_L - \omega_S$ as:

$$\chi_R(\omega_S) = \frac{(\varepsilon_0 N/6m)(\partial\alpha/\partial q)_0^2}{\omega_\nu^2 - (\omega_L - \omega_S)^2 + 2i(\omega_L - \omega_S)\gamma} = \chi'_R(\omega_S) + i\chi''_R(\omega_S). \quad (6.151)$$

Here $\chi'_R(\omega_S)$ and $\chi''_R(\omega_S)$ are the real and imaginary parts of the susceptibility. Their form is shown in Fig.6.22. This nonlinearity is atomic and not electronic in nature - that is - it is due to the motion of atoms within a molecule and not the motion of an electron within an atom. We also note that at exact resonance $\omega_S = \omega_L - \omega_\nu$ this nonlinear susceptibility is purely negative imaginary (which, as will see later - leads to amplification of the Stokes wave).

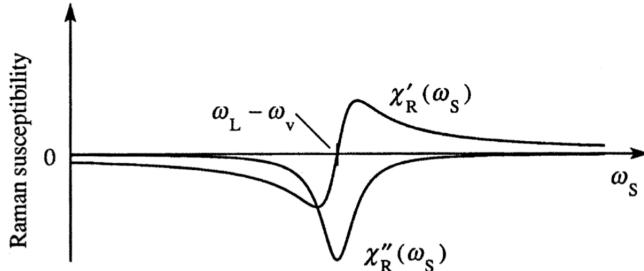


Fig. 6.22

Form of the SRS Stokes nonlinear susceptibility. The frequency $\omega_S = \omega_L - \omega_\nu$ is the resonance frequency.

We can now use the form of the optical fields Eq.6.140 and polarization Eq.6.148-Eq.6.150 in the wave equation Eq.1.17 for the Stokes mode to get:

$$\frac{dA_S}{dz} = -\alpha_S A_S, \quad (6.152)$$

with

$$\alpha_S = -3i \frac{\omega_S}{n_S c} \chi_R(\omega_S) |A_L|^2 \quad (6.153)$$

being an "absorption" coefficient. However, at resonance, as we noted above, the imaginary part of $\chi_R(\omega_S)$ is negative and there is no real part, and so $-\alpha_S$ would be positive leading to an exponential gain in the Stokes wave.

We note that this wave equations does not contain any phase mismatch, which is another way to state that this is an efficient gain process.

Now, we can repeat this whole analysis that started with Eq.6.140 but for the anti-Stokes wave with $\Omega = \omega_a - \omega_L$ and find that the susceptibility for the anti-Stokes wave is given with:

$$\chi_R(\omega_a) = \frac{(\varepsilon_0 N / 6m)(\partial \alpha / \partial q)_0^2}{\omega_\nu^2 - (\omega_L - \omega_a)^2 + 2i(\omega_L - \omega_a)\gamma} = \chi'_R(\omega_a) + i\chi''_R(\omega_a). \quad (6.154)$$

which has exactly the same form as for the Stokes wave but we note that

$$\omega_L - \omega_S = -(\omega_L - \omega_a), \quad (6.155)$$

and so:

$$\chi_R(\omega_a) = \chi_R^*(\omega_S) \quad (6.156)$$

The full form of the SRS nonlinear susceptibility is shown in Fig.6.23

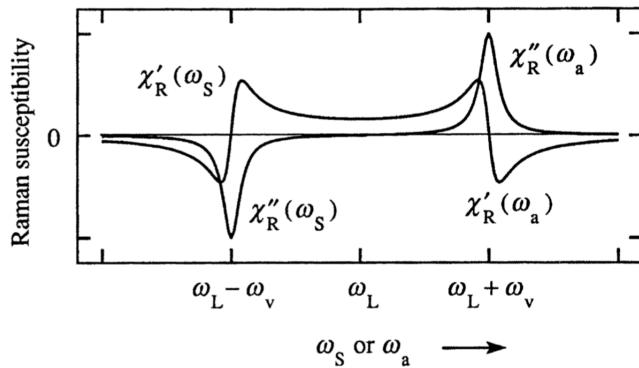


Fig. 6.23

Form of the full (Stokes and anti-Stokes) SRS nonlinear susceptibility.

The wave equation for the anti-Stokes wave is given with:

$$\frac{dA_a}{dz} = -\alpha_a A_S, \quad (6.157)$$

with

$$\alpha_a = -3i \frac{\omega_a}{n_a c} \chi_R(\omega_a) |A_L|^2 \quad (6.158)$$

However - here there is actually an absorption (attenuation) for the anti-Stokes wave in resonance.

Somewhat surprisingly it turns out that the anti-Stokes wave is actually being generated quite efficiently. This means that we did not discuss all the components our model can give us which can to the Stokes and anti-Stokes waves.

If we go back to the nonlinear polarization term in our model Eq.6.146 and inspect it carefully we would find an additional term oscillating at the anti-Stokes frequency ω_a :

$$P_a^{NL}(z) = P(\omega_a)e^{-i\omega_a t} + c.c. \quad (6.159)$$

with

$$P(\omega_a) = N\varepsilon_0 \left(\frac{\partial\alpha}{\partial q} \right)_0 q(\Omega) A_L. \quad (6.160)$$

Substituting the form of $q(\Omega)$ from Eq.6.144 we find:

$$P(\omega_a) = \frac{(N\varepsilon_0^2/m) (\partial\alpha/\partial q)_0^2 A_L^2 A_S^* e^{i(2k_L - k_S)z}}{\omega_\nu^2 - \Omega^2 - 2i\Omega\gamma}. \quad (6.161)$$

The dependency on ω_a in the above expression is found in $\Omega = \omega_L - \omega_S = \omega_a - \omega_L$.

What we have found here is again a Four-wave mixing process, but this time of: $\omega_a = \omega_L + \omega_L - \omega_S$ with the difference from what we had until now is that here we have a completely parametric process with no involvement of an acoustic wave. Here the anti-Stokes waves is directly fed by both the Stokes component and the laser pumping the system. As a result this process can be phase mismatched as shows the term $2k_L - k_S$. Anyway, as this is a FWM term we describe it with $\chi_F(\omega_a)$ such that:

$$P(\omega_a) = 3\varepsilon_0 \chi_F(\omega_a) A_L^2 A_S^* e^{i(2k_L - k_S)z} \quad (6.162)$$

where the prefactor of 3 is related to the degeneracy factor and:

$$\chi_F(\omega_a) = \frac{(N\varepsilon_0^2/3m) (\partial\alpha/\partial q)_0^2}{\omega_\nu^2 - (\omega_L - \omega_a)^2 + 2i(\omega_L - \omega_a)\gamma} \quad (6.163)$$

So, summing the contributions to the anti-Stokes wave we have altogether:

$$P(\omega_a) = 6\varepsilon_0 \chi_R(\omega_a) |A_L|^2 A_a e^{ik_a z} + 3\varepsilon_0 \chi_F(\omega_a) A_L^2 A_S^* e^{i(2k_L - k_S)z}. \quad (6.164)$$

So again - the first term is the feedback in which the anti-Stokes feeds an acoustic wave and the acoustic wave feeds the anti-Stokes wave in turn, and the second term is the FWM process fed by the laser and Stokes components.

In a very similar manner we can return to the Stokes wave and find that it also has a FWM contribution, described with:

$$\chi_F(\omega_S) = \frac{(N\varepsilon_0^2/3m) (\partial\alpha/\partial q)_0^2}{\omega_\nu^2 - (\omega_L - \omega_S)^2 + 2i(\omega_L - \omega_S)\gamma} \quad (6.165)$$

and the total polarization for Stokes wave is:

$$P(\omega_S) = 6\varepsilon_0 \chi_R(\omega_S) |A_L|^2 A_S e^{ik_S z} + 3\varepsilon_0 \chi_F(\omega_S) A_L^2 A_a^* e^{i(2k_L - k_a)z}. \quad (6.166)$$

We can now extend the evolution equation for the Stokes (Eq.6.152) and anti-Stokes (Eq.6.157) waves with the more complete polarization terms that we got here to get:

$$\frac{dA_S}{dz} = -\alpha_S A_S + \kappa_S A_a^* e^{i\Delta kz}, \quad (6.167)$$

$$\frac{dA_a}{dz} = -\alpha_a A_S + \kappa_a A_S^* e^{i\Delta kz}, \quad (6.168)$$

where

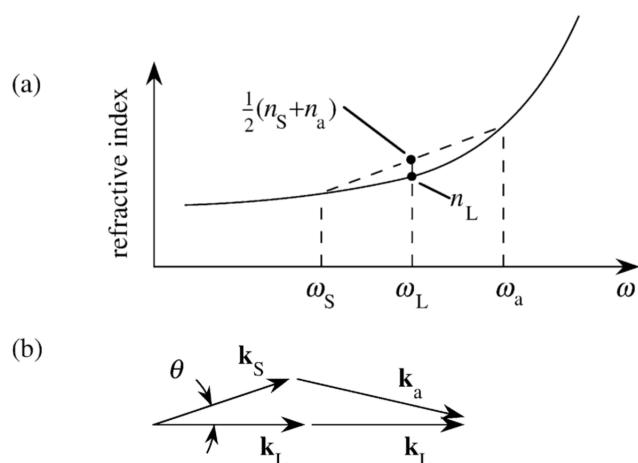
$$\alpha_j = -\frac{3i\omega_j}{n_j c} \chi_R(\omega_j) |A_L|^2, \quad j = S, a, \quad (6.169)$$

$$\kappa_j = \frac{3i\omega_j}{2n_j c} \chi_F(\omega_j) A_L^2, \quad j = S, a, \quad (6.170)$$

and the phase mismatch is given with:

$$\Delta k = \Delta \mathbf{k} \cdot \hat{\mathbf{z}} = (2\mathbf{k}_L - \mathbf{k}_S - \mathbf{k}_a) \cdot \hat{\mathbf{z}}. \quad (6.171)$$

The forms of these equations show that both the Stokes and anti-Stokes waves experience Raman gain or loss and a FWM term which can have some mismatch. In order for the FWM term to be efficient we require that the phase mismatch would be zero. We notice that in a material with normal dispersion, as $\omega_S < \omega_L < \omega_a$ and $n_S < n_L < n_a$ (leading to $n_L < (n_S + n_a)/2$, see Fig.6.24(a)) we'll have overall $2k_L < k_S + k_a$ - which is simply the triangle inequality provided k_L, k_S and k_a stand for the length of vectors - which of course - they do. This means that we can always phase match the FWM part of the interaction in a non co-linear setting as depicted in Fig.6.24(b).

**Fig. 6.24**

Phase matching for Stokes and anti-Stokes coupling in SRS. (a) The various interacting components in a material with normal dispersion. (b) Phase matching diagram.

7

Extreme nonlinear optics

In this chapter we would consider the application of optical forces (applied through the electric field) which easily compete with the Coulomb binding of electrons to the material. This means that the perturbative regime that we used until this point is no longer valid and we enter the regime of *extreme nonlinear optics*. In particular we would examine an optical upconversion process known as *high harmonic generation* (HHG) in which many photons of the pump pulse can be converted to a single photon in a measurable flux. We would consider for simplicity such an interaction for a gas of atoms.

In terms of energy, the potential Coulombic energy is given with:

$$U_0(r) = -\frac{e^2}{4\pi\epsilon_0 r} \quad (7.1)$$

with r the distance between the ion and electron.

Using a one dimensional approximating model:

$$U_0(x) = -\frac{e^2}{4\pi\epsilon_0|x|} \quad (7.2)$$

The potential energy of the electric field in this 1D model is associated with the dipole moment of the electron $d = -ex$ (in 1D) given with:

$$U_E = -d \cdot E = -eE(t)x. \quad (7.3)$$

So the total potential energy of the electron is:

$$U(x) = -\frac{e^2}{4\pi\epsilon_0|x|} - eE(t)x. \quad (7.4)$$

The three step model is a simple model that describes the process of HHG. Examining the expression above - when there is no applied electric field - the electron is stationary in its ground state. When the field is applied in the form of a strong oscillating pulse - eventually when the field is strong enough there is a high probability for the electron to tunnel out of the combined potential (see Fig.7(a)). Now the freed electron is accelerated by the laser pulse - first one way and then (because the electric field is oscillating) back towards its parent ion (Fig.7(b)). During this acceleration the electron can acquire a high value of kinetic energy. Finally, when the electron is back at the ion it is some probability to recombine with it, getting back to the ground state of the atom while in the process releasing its excess energy

(kinetic energy plus the binding or ionization energy) in the form of a single photon (Fig.7(c))). The energy of this photon can be the sum of energies of a high number of photons of the pump beam - hence the name - high harmonic generation.

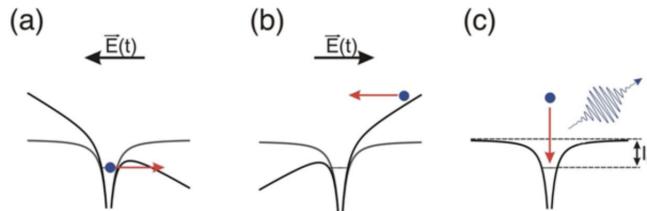


Fig. 7.1

The three step model for high harmonic generation. (a) Tunneling (b) acceleration (c) recombination. I_p is the ionization potential - the energy required to ionize the electron from its ground state.

We would now consider an even simpler model to allow us to analyze the process of tunneling during HHG. For this we would use a simpler model for the potential - of a field-free finite well potential (as shown on the left side of Fig.7).

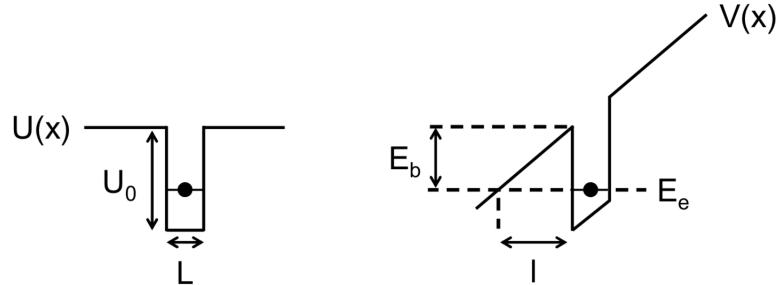


Fig. 7.2

Electric field induced tunneling. Left: a model potential well with an electron in its ground state in a finite potential well of depth U_0 . Right: For some large value of the induced optical field the electron needs to tunnel over a distance of l and an energy barrier of height E_b which is equal to the ionization potential.

Despite the assumption that tunneling is an instantaneous event we model it as a dynamic process during which the electron possesses a kinetic energy, so its total energy U_e is given with:

$$U_e = \frac{1}{2}mv^2 + U(x) \quad (7.5)$$

As long as the electron still resides under the potential it must have $U(x) > U_e$ so this kinetic energy must be negative! This means that this proposed dynamics is governed with a purely imaginary velocity with an absolute value of:

$$|v(x)| = \sqrt{\frac{2}{m}[U(x) - U_e]} \quad (7.6)$$

At the highest point of the potential the electron "experiences" during its tunneling process :

$$U(x)_{max} - U_e = I_p \quad (7.7)$$

so:

$$|v|_{max} = \sqrt{\frac{2I_p}{m}} \quad (7.8)$$

During the tunneling process the electron "slows" down until it reaches the point $U(x) = U_e$ for which $|v|_{min} = 0$. As in our simple model $U(x)$ is linear, the average velocity of the electron during the tunneling process is given with:

$$\langle |v| \rangle = \frac{1}{2} \sqrt{\frac{2I_p}{m}} = \sqrt{\frac{I_p}{2m}}. \quad (7.9)$$

Now, the width l of the barrier obeys the condition:

$$U_e(x = -l) = -I_p \quad (7.10)$$

But at that point the electron has kinetic energy and is no longer in the potential well - it is just under the influence of the potential induced by the electric field: $U_e(x = -l) = U_E(x = -l)$ so by using Eq.7.3 and the above equation we get:

$$-eE(t_0)(-l) = -I_p \quad (7.11)$$

where t_0 is the point in time given the total potential depicted in Fig.7. We denote $E(t = t_0) = E_0$ so:

$$l = \frac{I_p}{e \cdot E_0} \quad (7.12)$$

With Eq.7.9 and Eq.7.12 we can define the tunneling time to be:

$$t_{tun} = \frac{l}{\langle |v| \rangle} = \frac{\sqrt{2mI_p}}{e \cdot E_0} \quad (7.13)$$

In order that this view of tunneling would be valid we require that the tunneling time would be much shorter than than the typical time for the dynamics of the field. Equivalently we can require that the frequency associated with tunneling $\Omega_{tun} = 2\pi/t_{tun}$ would be much faster than the frequency of the field ω_0 : $\Omega_{tun} \gg \omega_0$. Equivalently, we define the *Keldysh parameter*:

$$\gamma = \frac{\omega_0}{\Omega_{tun}} = \frac{\omega_0 \sqrt{2mI_p}}{e \cdot E_0} \quad (7.14)$$

and require that $\gamma \ll 1$ in order for this "tunneling regime" to be effective. As the field gets stronger it is more probable for the electron to tunnel out of the potential. We can estimate this probability with a very crude model, as follows: let us assume that the wave function of the electron under the potential barrier is given with:

$$\psi(x) \propto e^{-|k(x)||x|} \quad (7.15)$$

That is - it is an evanescent wave. We now approximate the wave vector with its average value:

$$|k(x)| \cong \langle |k(x)| \rangle = m \langle |v| \rangle / \hbar. \quad (7.16)$$

Using Eq.7.9 for the expression of the wave vector and substituting back into the expression for the wave function we get:

$$\psi(x) \propto e^{-\frac{1}{\hbar} \sqrt{\frac{mI_p}{2}} |x|} \quad (7.17)$$

We would also estimate very roughly that the probability for ionization is approximated with:

$$P_{tun} \propto |\psi(x = -l)|^2 \quad (7.18)$$

Using the value of l from Eq.7.12 and the expression for $\psi(x)$ we get:

$$P_{tun} \propto e^{-\frac{\sqrt{2m}}{\hbar e} I_p^{3/2} \frac{1}{|E_0|}} \quad (7.19)$$

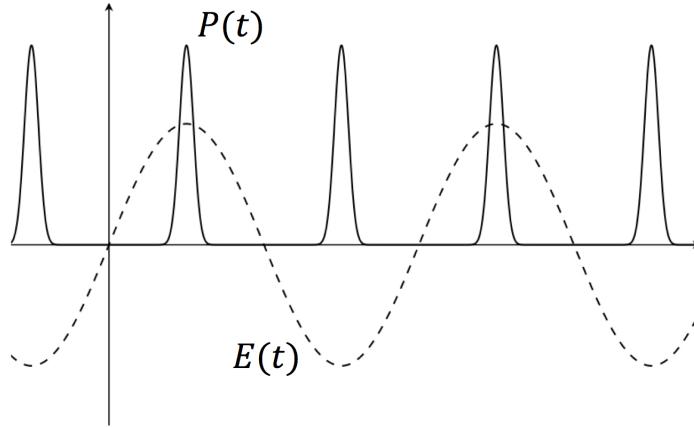
If we parametrize this expression as a function of time by substituting $E_0 \rightarrow E(t)$ we get:

$$P_{tun} \propto \exp \left[-\frac{\sqrt{2m}}{\hbar e} I_p^{3/2} \frac{1}{|E(t)|} \right] \quad (7.20)$$

A typical scenario is shown in Fig.7. This case of relatively isolated bursts of the probability of ionization happens when the values in the exponent in the above equation are just around the "knee" of the exponent - where it starts to change fast.

Following the ionization of the electron, we can analyze its movement under the influence of the laser field $E(t)$ (the same field that ionized it) using simple Newton's equation of motion:

$$m\ddot{x} = -eE(t) \quad (7.21)$$

**Fig. 7.3**

Ionization dynamics. A depiction of the electric field $E(t)$ (dashed line) and of the probability for ionization $P(t)$ (in a continuous line).

For simplicity we assume that the field is a CW, so our conclusions would be adequate for a single oscillation of the field. The field is given now with:

$$E(t) = E_0 \cos(\omega_0 t + \phi). \quad (7.22)$$

For a CW field there is no real meaning for the carrier-envelope-phase (CEP) ϕ so we'll set it to zero (for any other value we'll get the same set of solutions but permuted cyclically). We also assume the following initial conditions:

$$x(t_0) = 0. \quad (7.23)$$

$$v(t_0) = 0. \quad (7.24)$$

This means that the electron was ionized at some time t_0 where it is released very close to the ion set at coordinate $x = 0$ and there is no excess kinetic energy following the tunneling ionization process. We integrate the equation of motion to find the velocity:

$$\dot{x}(t) = v(t) = -\frac{eE_0}{m\omega_0} [\sin(\omega_0 t) - \sin(\omega_0 t_0)], \quad (7.25)$$

and integrate again to find the coordinate:

$$x(t) = \frac{eE_0}{m\omega_0^2} [\cos(\omega_0 t) - \cos(\omega_0 t_0) + \sin(\omega_0 t_0)(\omega_0 t - \omega_0 t_0)]. \quad (7.26)$$

The kinetic energy of the electron at time t is:

$$E_k(t) = \frac{1}{2}mv^2(t) = 2\langle E_{kin} \rangle [\sin(\omega_0 t) - \sin(\omega_0 t_0)]^2, \quad (7.27)$$

where

$$\langle E_{kin} \rangle = \frac{1}{2}m\langle v^2(t)|_{t_0=0} \rangle = \frac{1}{4}\frac{e^2E_0^2}{m\omega_0^2} \triangleq U_p \quad (7.28)$$

is the averaged kinetic energy of the electron in the applied electric field, also known as the *Pondermotive energy* or *Pondermotive potential*.

Above we used:

$$\langle v^2(t)|_{t_0=0} \rangle = \frac{1}{T} \int_0^T v^2(t)|_{t_0=0} dt, \quad (7.29)$$

with $T = 2\pi/\omega_0$.

It can be verified numerically that the maximum possible value of the kinetic energy is:

$$E_k|_{max} = E_k(\omega_0 t_{max} = 0.05 \cdot 2\pi) \approx 3.17U_p. \quad (7.30)$$

We note that with the Pondermotive energy (or potential) we can rewrite the Keldysh parameter (Eq.7.14) as:

$$\gamma = \sqrt{\frac{I_p}{2U_p}}. \quad (7.31)$$

Eq.7.26 describes the possible classical trajectories of the electron. If the electron returns at some time t_1 to its parent ion, it can recombine and give rise to HHG. We call t_1 in this case the recombination time. So the condition for the emission of new radiation components in this model is:

$$x(t_0) = x(t_1) = 0. \quad (7.32)$$

The trajectories equation can be solved numerically in order to find all possible pairs of time values (t_0, t_1) that obey Eq.7.32. The result of this calculation is shown in Fig.7.

For each such trajectory we can use Eq.7.27 to calculate the kinetic energy of the electron in its moment of recombination $E_k(t_1)$. Now, it is of course enough to calculate all possible returning trajectories over one cycle of the driving field: $-\pi \leq \omega_0 t_0 \leq \pi$. The result of such a numerical calculation is shown in Fig.7. In this figure we can recognize the maximum value with the one given in Eq.7.30. The maximal kinetic energy gives the maximum photon energy available in the HHG process by adding to it the binding energy I_p (the energy required for ionization is equal to the energy released when the electron is bounded back to the ground state):

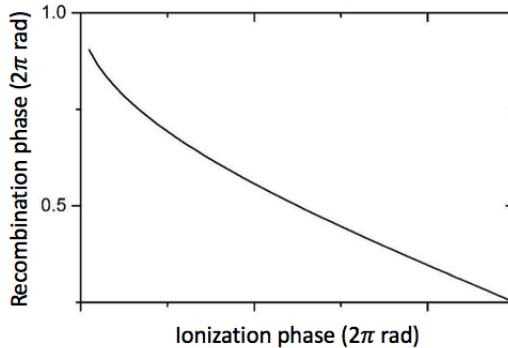


Fig. 7.4

Recombination time vs ionization time for returning electron trajectories.

$$\hbar\omega_{max} = I_p + 3.17U_p. \quad (7.33)$$

This maximal photon energy (or maximal possible kinetic energy for the electron) is known as the *cut-off energy*.

Further we notice that for every value of kinetic energy below the maximum - there are two possible recombination (emission) times. From Fig.7 we see that earlier ionization time have greater delay until they recombine. For this reason we term the two trajectories giving rise to the same kinetic energy - as short and long trajectories. The long trajectory is released earlier. Notice that if we look at the kinetic energy as a function of the emission of the HHG photons we'll get a mirror image - the short trajectories would appear first, followed by the long trajectories.

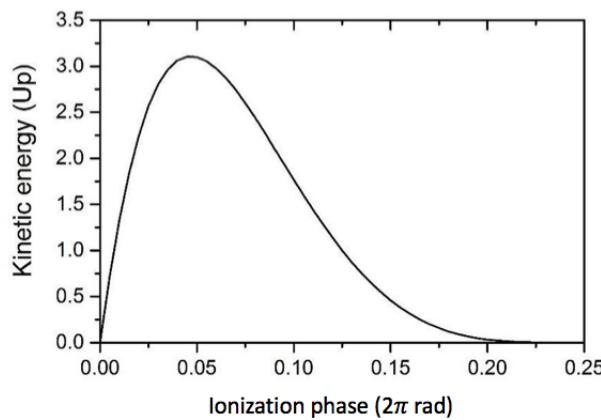
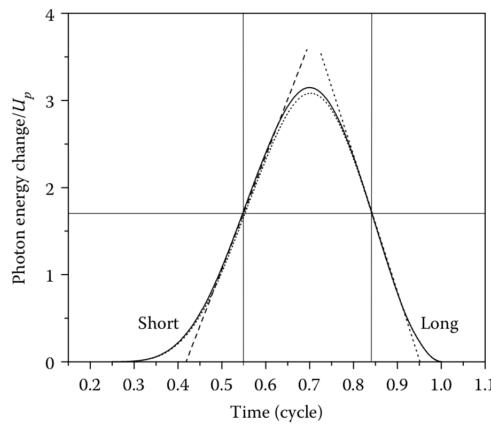


Fig. 7.5

The returning electron kinetic energy in units of U_p as a function of the phase of the field at the moment of ionization phase $\omega_0 t_0$.

As the photon energy is changing as a function of time - the HHG radiation is inherently chirped. The short trajectories emission is positively chirped (frequency rises with time) while the long trajectories emission is negatively chirped. This phenomenon is known as *attosecond chirp*. Atto is ten to the power of -18 , and the name attosecond is given here as the dynamics of the HHG emission (and electron trajectories) takes place over time intervals which are less than half a cycle of the driving field. Generally such time intervals measure hundreds of attosecond. For this reason HHG is part of the so called atto-science.

**Fig. 7.6**

Attosecond chirp of HHG. Photon energy (change) of HHG as a function of emission time.

As the emission of HHG repeats every half cycle of the driving laser - we have a semi-periodic (considering a long pulse) emission pattern with period $T/2 = \pi/\omega_0$. In the spectrum we thus have a periodic train of peaks with separation of $2\pi/(T/2) = 2\pi/(\pi/\omega_0) = 2\omega_0$. This is true approximately up to the cutoff harmonic order. Further the harmonic orders that we'll see are typically the odd-order harmonics, due to the inversion symmetry of atoms in the medium (or due to averaging effect of randomly oriented molecules). A typical structure of HHG spectrum is shown in Fig.7. Notice the "plateau" of harmonics. Unlike the power law observed in perturbative non-linear optics, here many harmonic orders can have about the same intensity. This is a strong indication that the process is non-perturbative and so belongs to the strong field regime.

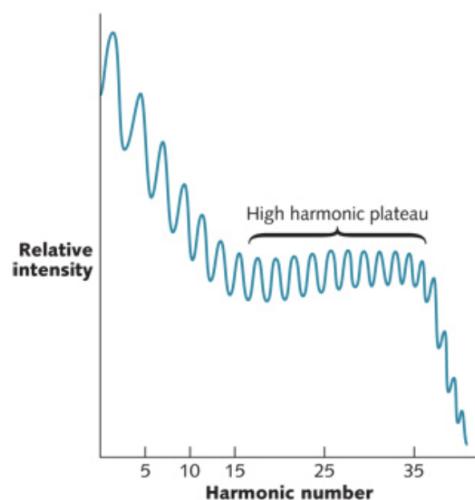


Fig. 7.7 Typical structure of HHG spectrum.

Semiconductor materials are quite important in both linear and nonlinear optics as devices can be created from these materials. Additionally semiconductors have relatively large nonlinearities. The energy levels of semiconductors are ordered in bands, where the relevant bands for optics are the valence band (which is the top band filled with electrons in absolute zero temperature) and the conducting band - which is the closest band above the valence band. The energy gap between the bands is denoted with E_g (see Fig.8(a)) In room temperature there are always some excited electrons that populate the conduction band. There are two types of electron transitions that can contribute to a nonlinear response: band-to-band (interband) transitions (Fig.8(b)) which is possible when the energy of incoming photons are larger than E_g , and virtual transitions (Fig.8(c)) when the incoming photon energy is smaller than E_g .

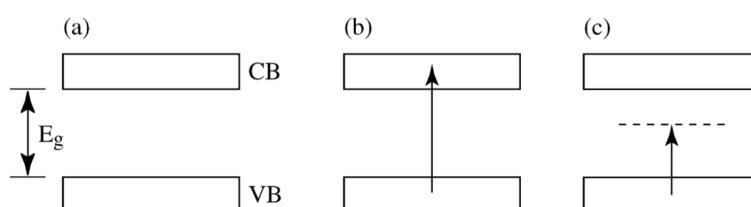


Fig. 8.1

Nonlinear optics in semiconductors. (a) The relevant energy bands of a semiconductor are the valence band (VB) and conduction band (CB), which are separated by the energy gap E_g . (b) If the incoming radiation consists of photon with energy $\hbar\omega > E_g$ then the nonlinear response is due to transfer of electrons to the conduction band. (c) Otherwise we have nonlinear response due to virtual transitions.

8.1 Interband transitions nonlinearities

The premise here is that the index of refraction of the material depends on the number of electrons in the conduction band. The population (density) N_c obeys a rate equation of the form:

$$\frac{dN_c}{dt} = \frac{\alpha I}{\hbar\omega} - \frac{N_c - N_c^{(0)}}{\tau_R}, \quad (8.1)$$

where α is an absorption coefficient for the radiation at frequency ω , $N_c^{(0)}$ is the population of the conduction band in thermal equilibrium and τ_R is the electron-hole recombination time.

The steady state solution, applicable for long enough pulses, is:

$$N_c = N_c^{(0)} + \frac{\alpha I \tau_R}{\hbar\omega}. \quad (8.2)$$

For pulses with duration shorter than τ_R the above equation is irrelevant, and the population just grows monotonically during the laser pulse.

The change in the electron population in the conduction band can change the index of refraction through several different mechanisms that we describe below.

8.1.1 Modification of optical properties due to the presence of free electrons

Electrons in the conduction band can be approximated as a free electron gas which responds to an applied optical field quite similarly to plasma. In this case the dielectric constant is given with:

$$\varepsilon(\omega) = \varepsilon_b - \frac{\omega_p^2}{\omega(\omega + i/\tau)}, \quad (8.3)$$

where ε_b is the contribution from bound charges, $\omega_p^2 = N_c e^2 / \varepsilon_0 m$ is the plasma frequency and τ is an optical response time which is usually much shorter than τ_R .

The increase in N_c with the laser intensity decreases the dielectric constant. In the steady-state limit we can derive an approximate expression for the index of refraction in the regular Kerr form of $n = n_0 + n_2 I$ with:

$$n_0^2 = \varepsilon_b - \frac{N_c(0)e^2}{\varepsilon_0 m \omega (\omega + i/\tau)} \quad (8.4)$$

and with:

$$n_2 = -\frac{e^2 \alpha \tau_R}{2 \varepsilon_b n_0 m \hbar \omega^3}. \quad (8.5)$$

The mass m in the last two equations is the effective mass of the electron in the conduction band. We note that the nonlinear effect goes as ω^{-3} which means it is more dominant for long wavelengths.

8.1.2 Modification of optical properties by plasma screening effects

When electrons have moved to the conducting band - the semiconductor becomes weakly conductive. Charges can flow and reduce the effective Coulomb potential between charges. It can be shown that the effective potential between two charges of charge e is now given with:

$$V = \frac{e^2}{4\pi\epsilon_0\epsilon r} e^{-\kappa r}, \quad (8.6)$$

with ϵ being the dielectric constant of the semiconductor and with

$$\kappa = \sqrt{\frac{N_c e^2}{\epsilon_0 k T}} \quad (8.7)$$

which is known as the *Debye-Hückel* screening wavenumber.

This screening effect reduces the strength of excitons. An exciton is a bound state of an electron-hole pair where the electron is in the conduction band and the hole is in the valence band. Typically the energy of these bound states lies below the conduction band and they can give rise to a feature in the absorption spectra of the material. However, the screening effect can weaken the attracting potential between the electron and hole, destroying the exciton. The change in the absorption at a given frequency (of the excitonic transition) changes (via the Kramers-Kronig relations) the index of refraction of the material. Thus the intensity of the laser responsible for screening modifies the optical properties of the crystal.

Just a reminder for the Kramers-Kronig relations - a change in absorption of value $\Delta\alpha$ would lead to a change in the refractive index of value:

$$\Delta n(\omega) = \frac{c}{\pi} \mathbb{P} \int_0^\infty \frac{\Delta\alpha(\omega') d\omega'}{\omega'^2 - \omega^2}, \quad (8.8)$$

where \mathbb{P} denotes principle value integration.

8.1.3 Modification of optical properties due to band filling effects

As electrons are excited to the conduction band, they start to fill the band, and the probability for further excitations is dropping. Hence the absorption is dropping and as a consequence the index of refraction (again through the Kramers-Kronig relations) gets modified.

8.1.4 Modification in optical properties due to band-gap renormalization

This is a subtle effect in which when the density of electrons in the conduction band rises, the energy-gap is getting smaller, changing the optical absorption spectrum.

8.2 Virtual transitions nonlinearities

When the energy of incoming photons is smaller than the energy gap (Fig.8.2(a)) only virtual transitions can occur. Nonlinear virtual processes, which are akin to four-wave-mixing, are illustrated in Fig.8.2(b)-(c). Such processes behave as a Kerr effect - modifying the index of refraction as a function of the light intensity. Usually the process illustrated in Fig.8.2(b) is more dominant than the one illustrated in Fig.8.2(c) (except when the photon energies approach the band-gap energy).

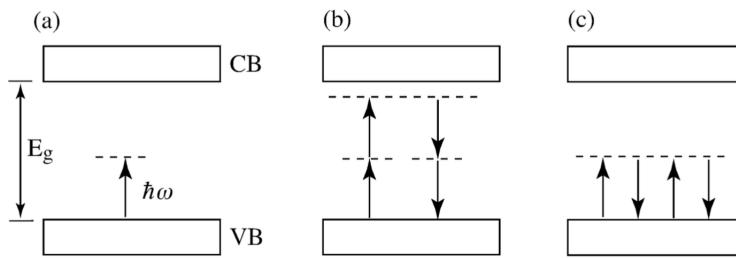


Fig. 8.2

Virtual transitions in a semiconductor. (a) Relevant scenario is when $\hbar\omega < E_g$. (b) A "four-wave-mixing" process via two-photon virtual transitions. (c) A "four-wave-mixing" process via single-photon virtual transitions.