

# Third Order Nonlinear Processes

## P A R T 1

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$$P_{tot} = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \dots$$

Linear + 2<sup>nd</sup> + 3<sup>rd</sup> + Higher

**Third Harmonic Generation (THG)**

**Kerr Effect**

**Self Focusing**

**Self- Diffraction**

**Self- Phase Modulation**

**Generation of Temporal Solitons**

**Stimulated Brillouin Scattering (SBS)**

**Stimulated Raman Techniques**

**Optical Phase Conjugation**

# Third Order Effects

Let us next examine the form of the nonlinear polarization

$$\tilde{P}^{(3)}(t) = \epsilon_0 \chi^{(3)} \tilde{E}^3(t)$$

induced by an applied field that consists of three frequency components:

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + \text{c.c.}$$

The third order polarization will have **22 components** which their frequency dependent are:

**Note:** There are  $6 \times 6 \times 6 = 216$  terms, can be sorted to 22 frequencies.

$$\begin{aligned} &\omega_1, \omega_2, \omega_3, 3\omega_1, 3\omega_2, 3\omega_3, (\omega_1 + \omega_2 + \omega_3), (\omega_1 + \omega_2 - \omega_3), \\ &(\omega_1 + \omega_3 - \omega_2), (\omega_2 + \omega_3 - \omega_1), (2\omega_1 \pm \omega_2), (2\omega_1 \pm \omega_3), (2\omega_2 \pm \omega_1), \\ &(2\omega_2 \pm \omega_3), (2\omega_3 \pm \omega_1), (2\omega_3 \pm \omega_2), \end{aligned}$$

## Third order polarizations (General)

$$1 \quad P(\omega_1) = \epsilon_0 \chi^{(3)} (3E_1 E_1^* + 6E_2 E_2^* + 6E_3 E_3^*) E_1,$$

$$2 \quad P(\omega_2) = \epsilon_0 \chi^{(3)} (6E_1 E_1^* + 3E_2 E_2^* + 6E_3 E_3^*) E_2,$$

$$3 \quad P(\omega_3) = \epsilon_0 \chi^{(3)} (6E_1 E_1^* + 6E_2 E_2^* + 3E_3 E_3^*) E_3,$$

$$4 \quad P(3\omega_1) = \epsilon_0 \chi^{(3)} E_1^3, \quad 5 \quad P(3\omega_2) = \epsilon_0 \chi^{(3)} E_2^3, \quad 6 \quad P(3\omega_3) = \epsilon_0 \chi^{(3)} E_3^3,$$

$$7 \quad P(\omega_1 + \omega_2 + \omega_3) = 6\epsilon_0 \chi^{(3)} E_1 E_2 E_3,$$

$$8 \quad P(\omega_1 + \omega_2 - \omega_3) = 6\epsilon_0 \chi^{(3)} E_1 E_2 E_3^*,$$

$$9 \quad P(\omega_1 + \omega_3 - \omega_2) = 6\epsilon_0 \chi^{(3)} E_1 E_3 E_2^*,$$

$$10 \quad P(\omega_2 + \omega_3 - \omega_1) = 6\epsilon_0 \chi^{(3)} E_2 E_3 E_1^*,$$

$$11 \quad P(2\omega_1 + \omega_2) = 3\epsilon_0 \chi^{(3)} E_1^2 E_2, \quad P(2\omega_1 + \omega_3) = 3\epsilon_0 \chi^{(3)} E_1^2 E_3, \quad 12$$

$$13 \quad P(2\omega_2 + \omega_1) = 3\epsilon_0 \chi^{(3)} E_2^2 E_1, \quad P(2\omega_2 + \omega_3) = 3\epsilon_0 \chi^{(3)} E_2^2 E_3, \quad 14$$

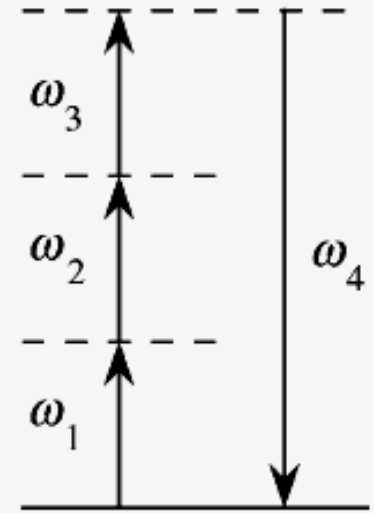
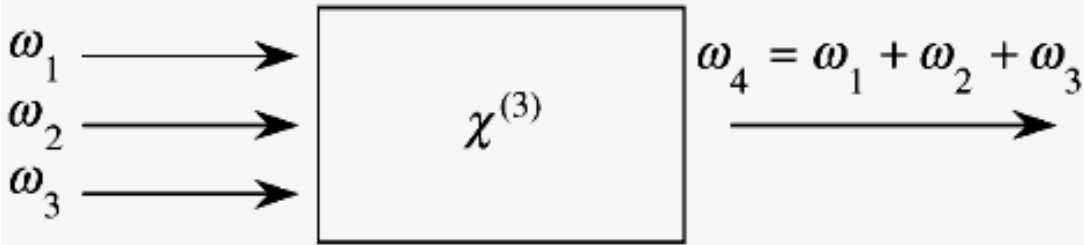
$$15 \quad P(2\omega_3 + \omega_1) = 3\epsilon_0 \chi^{(3)} E_3^2 E_1, \quad P(2\omega_3 + \omega_2) = 3\epsilon_0 \chi^{(3)} E_3^2 E_2, \quad 16$$

$$17 \quad P(2\omega_1 - \omega_2) = 3\epsilon_0 \chi^{(3)} E_1^2 E_2^*, \quad P(2\omega_1 - \omega_3) = 3\epsilon_0 \chi^{(3)} E_1^2 E_3^*, \quad 18$$

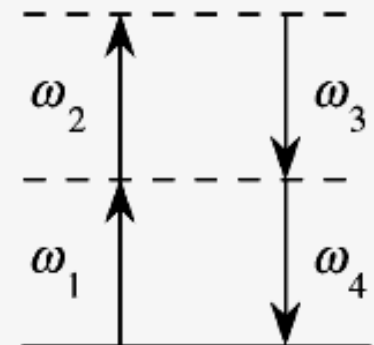
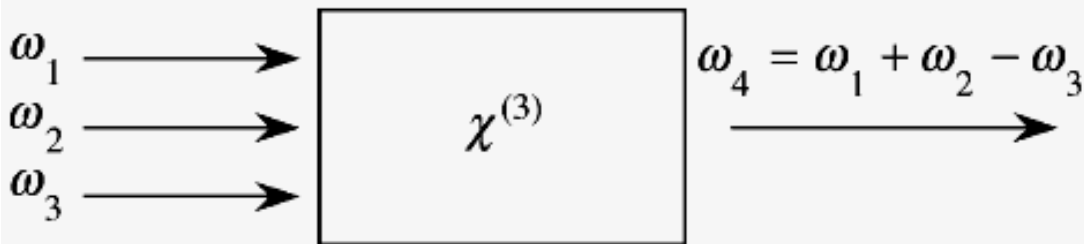
$$19 \quad P(2\omega_2 - \omega_1) = 3\epsilon_0 \chi^{(3)} E_2^2 E_1^*, \quad P(2\omega_2 - \omega_3) = 3\epsilon_0 \chi^{(3)} E_2^2 E_3^*, \quad 20$$

$$21 \quad P(2\omega_3 - \omega_1) = 3\epsilon_0 \chi^{(3)} E_3^2 E_1^*, \quad P(2\omega_3 - \omega_2) = 3\epsilon_0 \chi^{(3)} E_3^2 E_2^* \quad 22$$

(a)



(b)



**Two possible mixing processes that can occur when 3 input waves interact in  $\chi^{(3)}$  medium**

## 4.5 Third-Order Effects

Third-order nonlinear polarization  $P^{(3)}$  is a function of the third power of the incident electric light field  $E_{\text{inc}}$ :

$$P^{(3)} = \varepsilon_0 \chi^{(3)} E_{\text{inc},1} E_{\text{inc},2} E_{\text{inc},3} \quad (4.88)$$

with the third-order nonlinear susceptibility  $\chi^{(3)}$  which is a four-dimensional tensor which can in general be complex.

The three light fields  $E_{\text{inc},i}$  can be components of the same light beam but can also be three different light beams which overlap in the nonlinear material. If a fourth beam is used to detect the changes in the third-order nonlinear material *four wave mixing (FWM)* takes place (see Fig. 4.22).

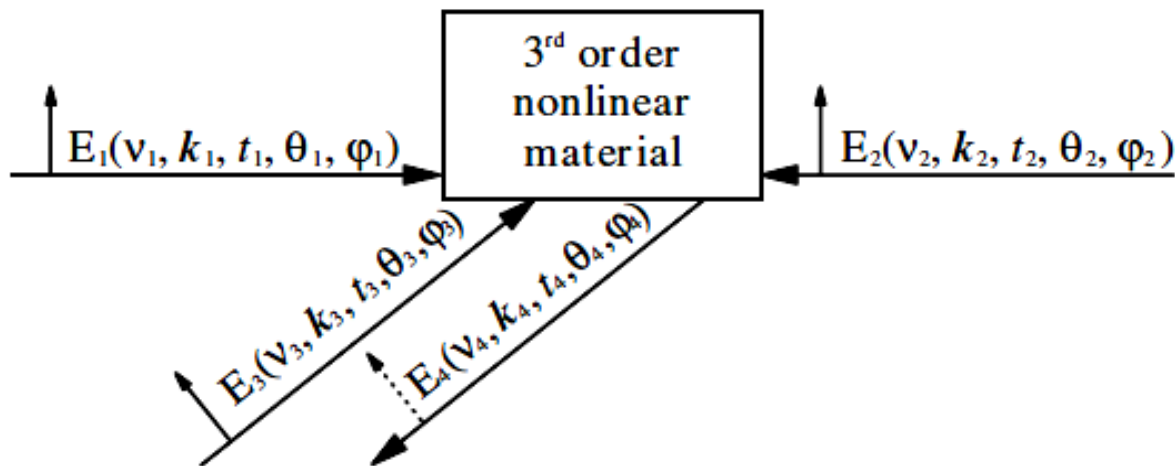


Fig. 4.22. Schematic of four-wave mixing as third order nonlinear process in suitable matter

By choosing different frequencies  $\nu_m$ , propagation directions  $\mathbf{k}_m$ , timing  $t_m$ , polarization  $\phi_i$  and phases  $\varphi_m$  more than hundred prominent schemes of FWM can be applied. Usually the transversal intensity distribution is neglected and plane waves are assumed for easier modeling. In one of the simplest cases all light waves have the same frequency and this process is called *degenerate four-wave mixing (DFWM)*.

The three components of  $\mathbf{P}^{(3)}$  in the  $x$ ,  $y$  and  $z$  direction follow from:

$$P_m^{(3)} = \frac{\varepsilon_0}{4} \left[ \sum_{p,q,r} \left\{ \chi_{mpqr}^{(3,1)} E_p E_q E_r + \chi_{mpqr}^{(3,2)} E_p^* E_q E_r \right. \right. \\ \left. \left. + \chi_{mpqr}^{(3,3)} E_p E_q^* E_r + \chi_{mpqr}^{(3,4)} E_p E_q E_r^* \right\} \right] \\ \text{with } m, p, q, r = x, y, z. \quad (4.89)$$

For nonabsorbing materials the susceptibility tensor  $\chi^{(3)}$  has 81 real components and the complex products of the electric field components disappear. Again as in second-order nonlinearity for symmetry reasons of the allowed permutations of the  $p$ ,  $q$ ,  $r$  of the electric field vectors the number of distinguishable tensor components of  $\chi^{(3)}$  is reduced. Only 30 different values

remain for this reason. The components of the third-order nonlinear polarization can be written as:

$$\begin{pmatrix} P_x^{(3)} \\ P_y^{(3)} \\ P_z^{(3)} \end{pmatrix} = \varepsilon_0 \cdot \begin{pmatrix} e_{11} & e_{12} & e_{13} & e_{14} & e_{15} & e_{16} & e_{17} & e_{18} & e_{19} & e_{1\ 10} \\ e_{21} & e_{22} & e_{23} & e_{24} & e_{25} & e_{26} & e_{27} & e_{28} & e_{29} & e_{2\ 10} \\ e_{31} & e_{32} & e_{33} & e_{34} & e_{35} & e_{36} & e_{37} & e_{38} & e_{39} & e_{3\ 10} \end{pmatrix} \cdot \begin{pmatrix} E_x^3 \\ E_y^3 \\ E_z^3 \\ 3E_x E_y^2 \\ 3E_x E_z^2 \\ 3E_y E_x^2 \\ 3E_y E_z^2 \\ 3E_z E_x^2 \\ 3E_z E_y^2 \\ 6E_x E_y E_z \end{pmatrix} \quad (4.90)$$

with the additional internal relations between the  $e$  matrix elements:

$$\begin{aligned} e_{12} &= e_{24} & e_{13} &= e_{35} & e_{14} &= e_{26} & e_{15} &= e_{38} & e_{16} &= e_{21} \\ e_{17} &= e_{25} = e_{3\ 10} & e_{19} &= e_{2\ 10} = e_{34} & e_{1\ 10} &= e_{28} = e_{36} \\ e_{18} &= e_{31} & e_{23} &= e_{37} & e_{27} &= e_{39} & e_{29} &= e_{32} \end{aligned} \quad (4.91)$$

considering the further symmetry rules  $\chi_{mpqr} = \chi_{pmqr}$ . Finally 15 components are relevant for describing the third-order nonlinear processes which are functions of the applied light frequency.



If the nonabsorbing crystals are of cubic symmetry only two different values are distinguishable:

$$P_m^{(3)} = \varepsilon_0 E_m \{e_{11} E_m^2 + 3e_{14} (E_p^2 + E_q^2)\} \quad \text{with } m \neq p \neq q = x, y, z \quad (4.92)$$

and if the material is isotropic the additional relation  $e_{11} = 3e_{14}$  is valid and the third-order nonlinear polarization reduces to:

$$P_m^{(3)} = \varepsilon_0 e_{11} E_m (\mathbf{E}\mathbf{E}) \quad \text{with } m = x, y, z. \quad (4.93)$$

Isotropic matter and materials of cubic symmetry will not show any second-order nonlinear effect and thus the third order effects are dominant. High intensities can usually be applied without other nonlinear losses in these materials. In particular isotropic matter such as gases, liquids or solutions and amorphous solids can be used for third-order nonlinear effects with high efficiencies in photonic applications. Optical phase conjugation (see Sect. 6.11.3, p. 480) and stimulated Raman scattering for frequency conversion (see Sect. 6.15.3, p. 528) may serve as examples.

# Third Harmonic Generation (THG)

## (Frequency Tripling)

Frequency tripling is a process of NL frequency conversion where the resulting optical frequency is three times that of the input laser beam.

This can be achieved with  $\chi^{(3)}$  nonlinearity for direct THG, but this is difficult due to the small  $\chi^{(3)}$  nonlinearity of optical media and phase-matching constraints.

Thus, frequency tripling is usually realized as a cascaded process, beginning with frequency doubling of the input beam and subsequent sum frequency generation of both waves, with both processes being based on nonlinear crystal materials with  $\chi^{(2)}$  nonlinearity.

# THG

If the medium possesses cubic nonlinearity, under the action of **2** monochromatic waves  $\omega_1$  and  $\omega_2$  the polarization  $P^{(3)}$  would contain the components with frequencies:

$$3\omega_1, 3\omega_2, 2\omega_1 \pm \omega_2, 2\omega_2 \pm \omega_1$$

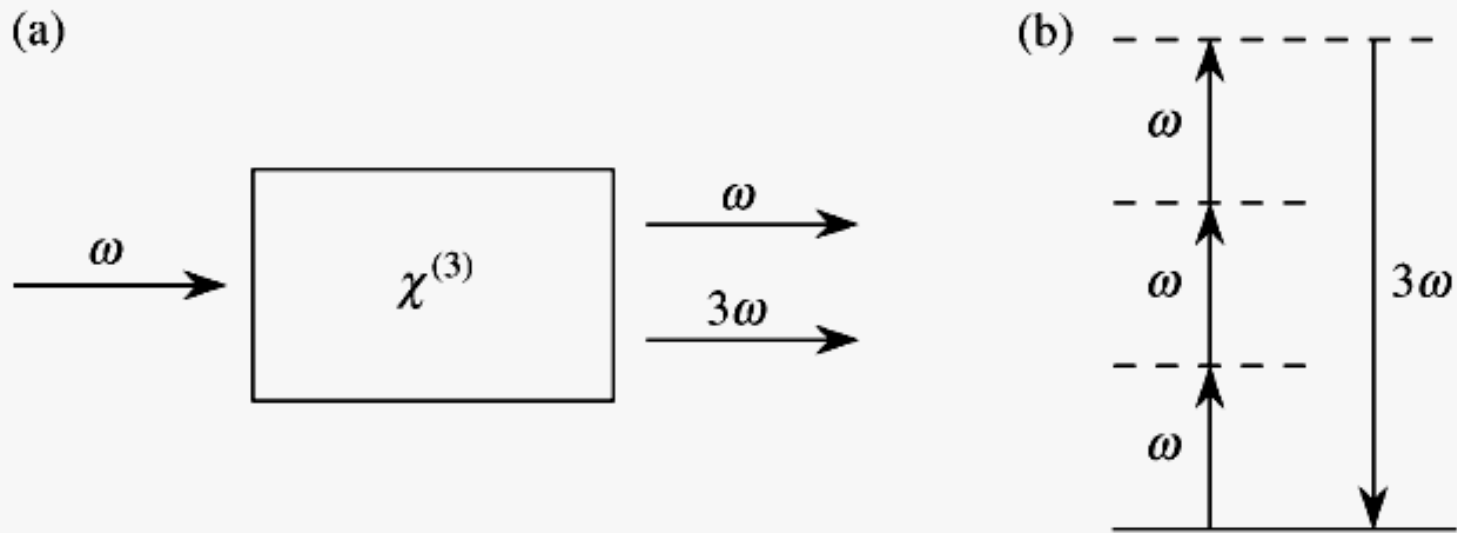
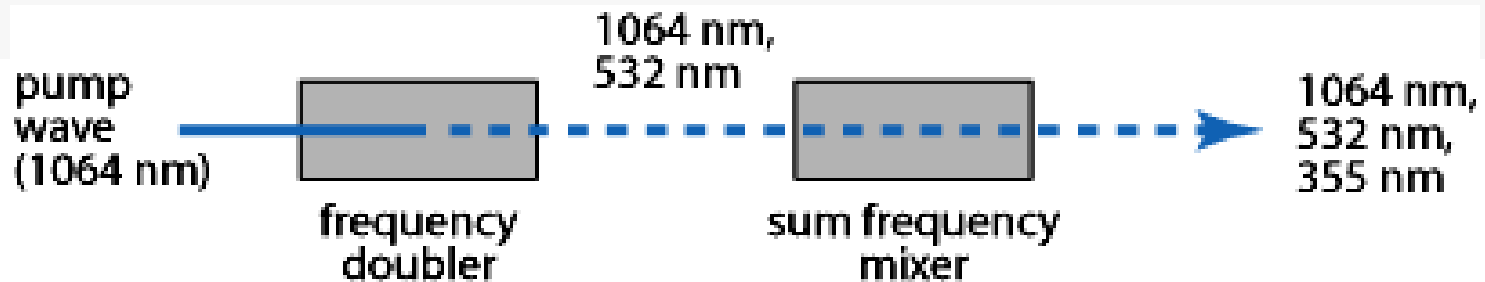


FIGURE Third-harmonic generation. (a) Geometry of the interaction. (b) Energy-level description.



### Typical configuration for frequency tripling

An infrared input beam at **1064 nm** generates a green **532 nm** wave, and these two mix in a second crystal to obtain **355 nm** light.

### 4.5.1 Generation of the Third Harmonic

Analogous to the generation of second harmonics the third-order nonlinearity of (4.90) can be applied for the generation of light with three times the frequency of the incident light wave. This process is called third harmonic generation (THG):

$$\text{THG} \quad I_{\text{out}}(3\nu_{\text{inc}}) \stackrel{\chi_{\text{THG}}^{(3)}}{\leftarrow} I_{\text{inc}}(\nu_{\text{inc}}). \quad (4.94)$$

If this process is based on the third-order susceptibility  $\chi^{(3)}$  as given above the efficiency for the known materials is quite low (typically  $< 10^{-4}$ ). Thus this method is mostly applied for the determination of third-order nonlinearity itself and the characterization of the material.

In photonics the most commonly used generation of third harmonic light is therefore based on the much more efficient two-step procedure of second harmonic generation (SHG) in a first step and frequency mixing of this second harmonic with the residual incident wave as the second step. Both steps are nonlinear to second order:

$$\text{THG II} \quad I_{\text{THG}}(3\nu_{\text{inc}}) \stackrel{\chi_{\text{mix}}^{(2)}}{\leftarrow} I_{\text{SHG}}(2\nu_{\text{inc}}), \quad I_{\text{res}}(\nu_{\text{inc}}) \stackrel{\chi_{\text{SHG}}^{(2)}}{\leftarrow} I_{\text{inc}}(\nu_{\text{inc}}). \quad (4.95)$$

The total efficiency for this type of third harmonic generation can be as high as about 80%. For optimal total efficiency, the efficiency of second harmonic generation has to be 2/3 for enough residual intensity of the fundamental wave  $I_{\text{res}}(\nu_{\text{inc}})$  to achieve the optimal photon numbers for the two different wavelengths.

# Kerr Effect

- At high intensities such as on the peak of an ultrashort laser pulse, the refractive index of any medium becomes a function of the incident intensity.
- The Kerr effect is a change in the refractive index of a material in response to an electric field. All materials show a Kerr effect, but certain liquids display the effect more strongly than other materials do. The Kerr effect was discovered in 1875 by John Kerr, a Scottish physicist.
- **Strong applied electric field can induce birefringence in materials which are isotropic without the field. The refractive index becomes a function of light intensity**

$$n_{non-linear} = \sqrt{(1 + \chi^{(1)} + \chi^{(3)})} \cong n_0 + n_2 I$$

# The Kerr effect:

The polarization rotation is proportional to the Kerr constant  $K$  and  $E^2$

$$\Delta n = \lambda_0 K E^2$$

where  $\Delta n$  is the induced birefringence,  
 $E$  is the electric field strength,  $K$  is the **Kerr constant** of the material.

**TABLE** Kerr constants for some selected liquids (20°C,  $\lambda_0 = 589.3$  nm).

Substance		$K$ (in units of $10^{-7}$ cm statvolt $^{-2}$ )
Benzene	$C_6H_6$	0.6
Carbon disulfide	$CS_2$	3.2
Chloroform	$CHCl_3$	-3.5
Water	$H_2O$	4.7
Nitrotoluene	$C_7H_7NO_2$	123
Nitrobenzene	$C_6H_5NO_2$	220

The **AC Kerr Effect** creates birefringence using intense fields of a light wave. Usually very high irradiances from ultra-short laser pulses are required to create quarter-wave rotations.

# Intensity Dependent Refractive Index

- The incident optical field

$$\tilde{E}(t) = E(\omega)e^{-i\omega t} + \text{C.C.}$$

- Third order nonlinear polarization

$$P^{(3)}(\omega) = 3\chi^{(3)}(\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega)$$



The **total polarization** can be written as

$$P^{\text{TOT}}(\omega) = \chi^{(1)} E(\omega) + 3\chi^{(3)} (\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega)$$

One can define an **effective susceptibility**

$$\chi_{\text{eff}} = \chi^{(1)} + 4\pi |E(\omega)|^2 \chi^{(3)}$$

The **refractive index** can be defined as

$$n^2 = 1 + 4\pi\chi_{\text{eff}}$$

By definition

$$n = n_0 + n_2 I$$

where

$$I = \frac{n_0 c}{2\pi} |E(\omega)|^2$$

and

$$n_2 = \frac{12\pi^2}{n_0^2 c} \chi^{(3)}$$

## 4.5.2 Kerr Effect

Based on third-order nonlinear polarization a strong applied electric field can induce optical birefringence in materials which are optically isotropic without the field [4.266–4.280]. Thus the refractive index tensor of the material becomes a function of the light intensity. Mostly this Kerr effect is achieved on the microscopic scale by the orientation of electric dipoles in the electric field. This electric field  $E_{\text{ext}} = U/d$  can be applied as an oscillating field of a strong incident light wave or as an external field as in the Pockels effect but with transversal orientation to the light propagation direction as shown in Fig. 4.23. For the following analysis polarized incident light with an electric field vector in the  $x$  direction with  $E_{\text{inc},x}(\nu_{\text{inc}})$  propagating in  $z$  direction through a third-order nonlinear material is assumed as in Fig. 4.23.

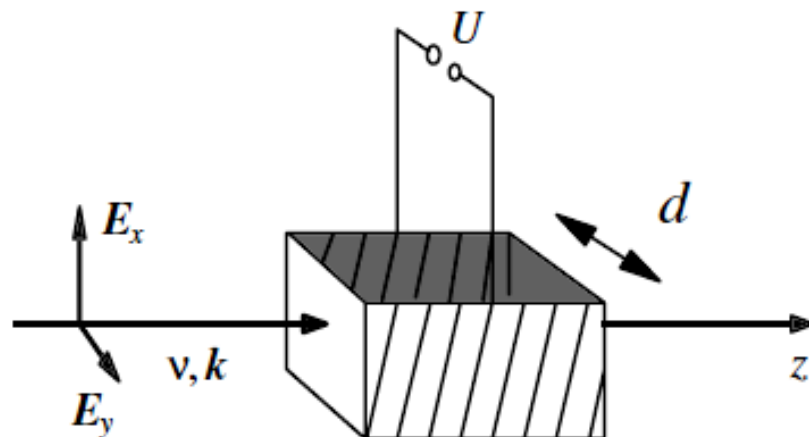


Fig. 4.23. Optical Kerr effect in an isotropic third-order nonlinear material inducing optical birefringence

The total polarization  $P^{(\text{tot})}$  is the sum over the linearly induced polarization and the third-order nonlinear polarization which both have a component in the  $x$  direction:

$$\begin{aligned}
 P_x^{(\text{tot})}(\nu_{\text{inc}}) &= P_{\text{lin},x}^{(1)} + P_{\text{nl},x}^{(3)} \\
 &= \varepsilon_0 \chi^{(1)} E_{\text{inc},x} + \varepsilon_0 \chi^{(3)} E_{\text{inc},x} \left( \frac{3}{4} E_{\text{inc},x}^2 + E_{\text{ext}}^2 \right) \\
 &= \varepsilon_0 E_{\text{inc},x} \left[ \chi^{(1)} + \chi^{(3)} \left( \frac{3}{4} E_{\text{inc},x}^2 + E_{\text{ext}}^2 \right) \right] \tag{4.96}
 \end{aligned}$$

$$P_y^{(\text{tot})}(\nu_{\text{inc}}) = P_z^{(\text{tot})}(\nu_{\text{inc}}) = 0$$

with

$$P_{\text{nl},x}^{(3)}(\nu_{\text{inc}}) = \varepsilon_0 e_{11} E_{\text{inc},x} \left( \frac{3}{4} E_{\text{inc},x}^2 + E_{\text{ext}}^2 \right). \tag{4.97}$$

The refractive index  $n = \sqrt{1 + \chi}$  in the  $x$  direction is changed by this nonlinear polarization:

$$n_{\text{nl},x} = \sqrt{n_0^2 + e_{11} \left( \frac{3}{4} E_{\text{inc},x}^2 + E_{\text{ext}}^2 \right)} \tag{4.98}$$

and is constant in the  $y$  and  $z$  directions. The small change  $\Delta n_x$  in the  $x$  direction results in:

$$\Delta n_x = \frac{e_{11}}{2n_0} \left( \frac{3}{4} E_{\text{inc},x}^2 + E_{\text{ext}}^2 \right). \quad (4.99)$$

Assuming low light intensities the change in the refractive index as a function of the external cw electric field  $E_{\text{ext}}^2$  results in:

$$\Delta n_x = \frac{e_{11}}{2n_0} E_{\text{ext}}^2 \quad (4.100)$$

The light polarization can also be applied parallel to the external electric field which is the  $y$  direction in Fig. 4.23 (p.209). The resulting refractive index for this polarization is then given by:

$$n_{\text{nl},y} = \sqrt{n_0^2 + e_{11} \left( \frac{3}{4} E_{\text{inc},y}^2 + 3E_{\text{ext}}^2 \right)} \quad (4.101)$$

which leads to a change in the refractive index  $\Delta n_y$  in this  $y$ -polarization direction (which is called *extraordinary* analogous to natural birefringence):

$$\Delta n_y = \frac{3e_{11}}{2n_0} E_{\text{ext}}^2 \quad (4.102)$$

The difference between the two components results in

$$\Delta n_{\text{Kerr}} = \Delta n_x - \Delta n_y = \frac{e_{11}}{n_0} E_{\text{ext}}^2 = K_{\text{Kerr}} \lambda_{\text{inc}} E_{\text{ext}}^2 \quad (4.103)$$

with the Kerr constant

$$\text{Kerr constant } K_{\text{Kerr}} = \frac{e_{11}}{n_0 \lambda_{\text{inc}}} \quad (4.104)$$

describing the Kerr effect for slowly changing fields. Thus, with the external electric field the birefringence of the Kerr material can be changed or in isotropic materials an optical birefringence can be induced. This can be used in a setup of a Kerr cell between two crossed polarizers as in Fig. 4.24. This is an electrically controlled light gate, a Kerr shutter.

If only fast oscillating fields are applied, different parameters for the Kerr effect are observed. In this case the reorientation and diffusion processes to reach the steady-state under the applied electric field may not be completed. This fast Kerr effect is typically described with the constant  $\gamma$  or  $\gamma_I$  as defined in relation to  $e_{11}$  in the next section.

Using this type of induced optical birefringence of the Kerr effect very fast optical switches can be made. The setup is sketched in Fig. 4.24.

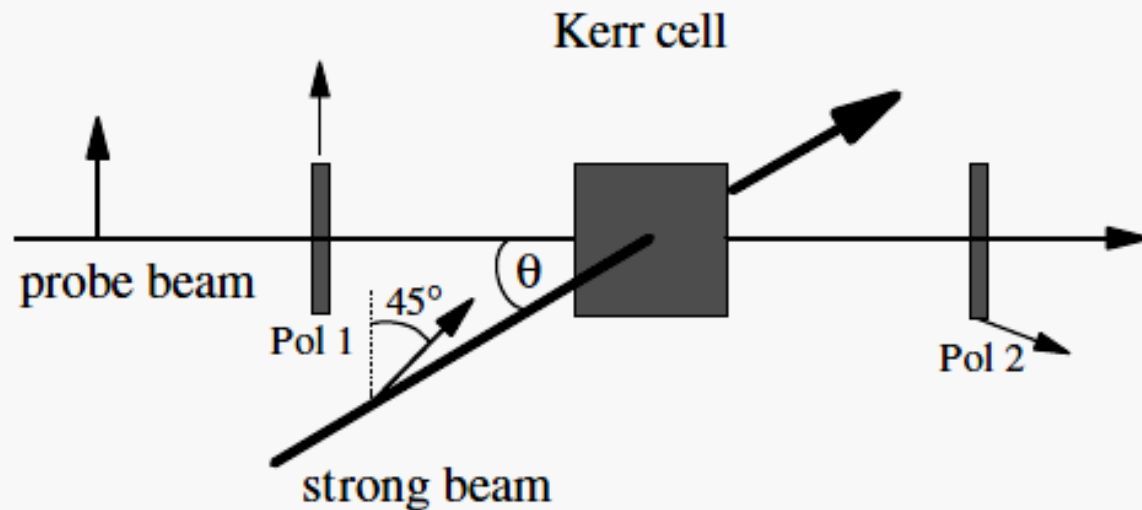


Fig. 4.24. Fast optical shutter based on the optical Kerr effect

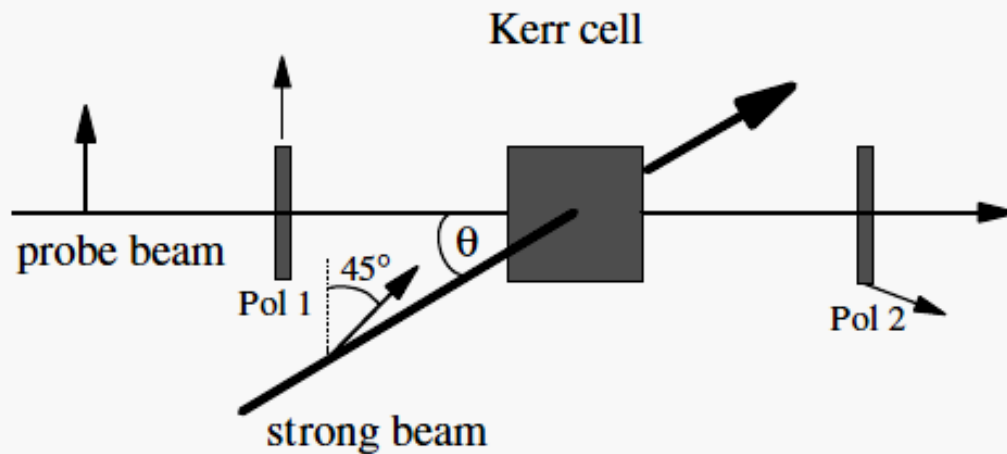


Fig. 4.24. Fast optical shutter based on the optical Kerr effect

The Kerr active material, e.g.  $\text{CS}_2$ , is placed between two crossed polarizers Pol 1 and Pol 2. Thus the incident probe light beam polarized in the direction of polarizer Pol 1 cannot pass the setup. In addition a short and strong light pulse is applied to switch on the transparency of this device. Therefore the direction of its polarization has to be  $45^\circ$  to the incident polarization and the angle  $\theta$  between the propagation directions of the probe and the strong pump beams should be as small as possible.

If the intensity of the switch beam is suitably chosen the incident probe light will be circularly polarized behind the nonlinear cell. Thus 50% of the incident probe beam intensity can pass the second polarizer at maximum in this case. If the intensity is even higher a  $90^\circ$  rotation of the polarization may be possible and then almost all probe light can pass the shutter.



# Self Focusing

**Self focusing of light** is the process in which an intense beam of light modifies the optical properties of a material medium in such a manner that the beam is caused to come to a focus within the material.



# Self Focusing

It is a NL optical process induced by the change in refractive index of materials exposed to intense light. **A medium whose refractive index increases with the electric field intensity acts as a focusing lens** for an EM wave characterized by an initial transverse intensity gradient (as in a laser beam).

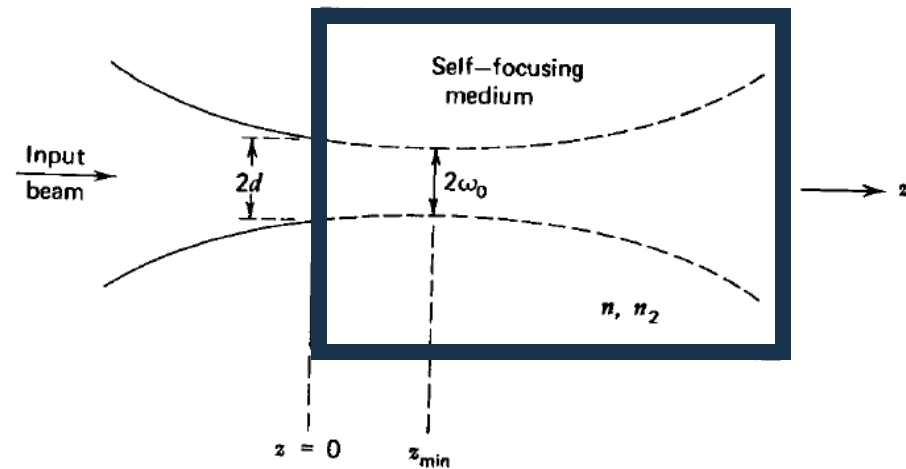
# Self Focusing and Self Defocusing

Due to a Kerr lens, an intense optical pulse propagating in a nonlinear medium can experience self-focusing: the beam diameter is decreased.

**The mechanism is based on a Kerr nonlinearity with positive  $\chi^{(3)}$ . The higher optical intensities on the beam axis, as compared with the wings of the spatial intensity distribution, cause an effectively increased refractive index for the inner part of the beam. This modified refractive index distribution then acts like a focusing lens.**

A related effect, occurring in the case of a **negative  $\chi^{(3)}$**  nonlinearity, is *self-defocusing*.

The laser beam has Gaussian intensity profile. It can induce a Gaussian refractive index profile inside the NLO sample.



**FIGURE 18.13** A Gaussian beam entering a slab of material that has a dielectric constant  $\epsilon = n^2 + nn_2 |E|^2$ . The dashed curve gives the beam radius without self-focusing ( $n_2 = 0$ ).

**Self Focusing**

$\chi^{(3)}$

### 4.5.3 Self-Focusing

If the intensity of a transmitted light beam is sufficiently high almost every material, gases, liquids or solids, will show a nonlinear interaction. Thus the refractive index will be changed as given in (4.99) by remarkable values if the nonlinear range of the electric field or intensity is reached. This refractive index change will modify the light propagation not only with respect to the polarization but in its geometrical properties too. In particular, if light beams with a transverse intensity profile, as, e.g. Gaussian beams, are applied this refractive index change will be different over the cross-section of the beams and depend on the transversally local intensity of the beam. Thus the refractive index becomes a function of the distance from the center of the incident light beam. This is equivalent to an optical lens effect.

Assuming a Gaussian transversal beam profile of the incident light the intensity across the beam radius  $r$  perpendicular to the beam propagation direction is given as a function of the total power  $P_{\text{tot}}$  by:

$$I_{\text{inc},r}(r, t) = \frac{2}{\pi w_{\text{beam}}^2} e^{-2r^2/w_{\text{beam}}^2} P_{\text{tot}}(t). \quad (4.106)$$

The refractive index  $n(r, I)$  in the material will be modified across the beam diameter based on (4.99) to:

$$n(r, I) = n_0 + \Delta n_{\text{nl},r} = n_0 + \frac{3}{8} \frac{e_{11}}{n_0} E_{\text{inc},r}^2 = n_0 + \frac{3}{4} \frac{e_{11}}{n_0^2 c_0 \epsilon_0} I_{\text{inc},r}$$

$$\Delta n_x = \frac{e_{11}}{2n_0} \left( \frac{3}{4} E_{\text{inc},x}^2 + E_{\text{ext}}^2 \right)$$

$$= n_0 + \gamma(\nu_{\text{inc}}) E_{\text{inc},r}^2$$

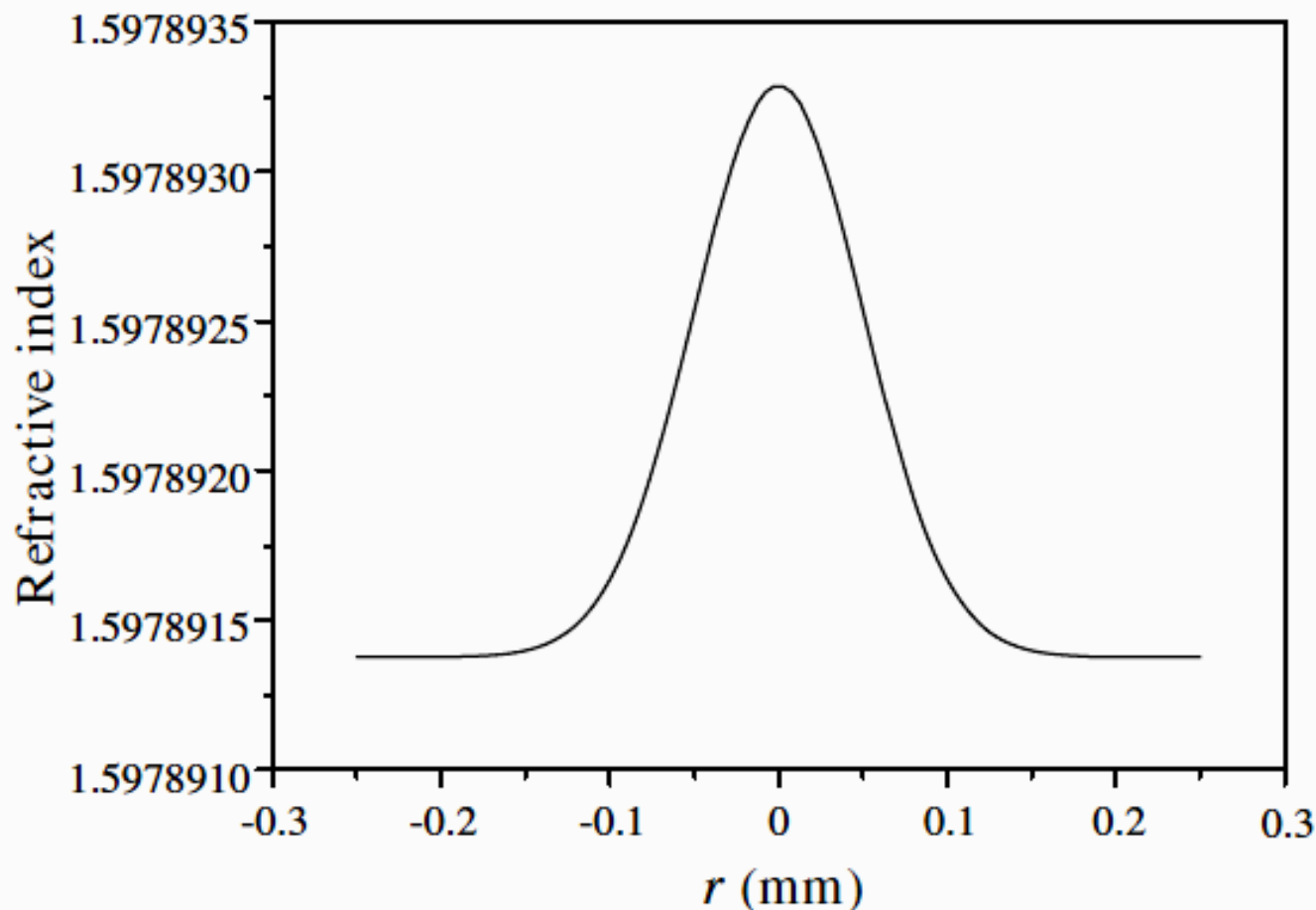
$$= n_0 + \gamma_I(\nu_{\text{inc}}) I_{\text{inc},r}$$

$$(4.107)$$

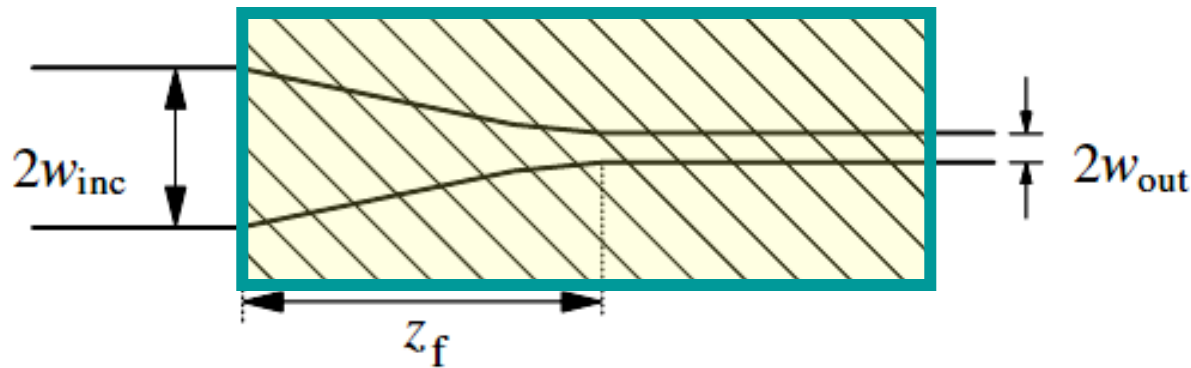
with the coefficients  $\gamma$  and  $\gamma_I$  as given in Table 4.7 (p. 211) valid for linearly polarized light. In the case of nonpolarized or circularly polarized light the effective coefficients have to be reduced to 2/3 of the given values:

$$\gamma(\nu_{\text{inc}}) = \frac{3}{8} \frac{e_{11}}{n_0} \quad \gamma(\nu_{\text{inc}})_{\text{circ}} = \frac{1}{4} \frac{e_{11}}{n_0}. \quad (4.108)$$

This Gaussian refractive index profile is shown in Fig. 4.25 resulting from a Gaussian shaped incident beam. This refractive index profile acts as a lens analogous to the quadratic refractive index profile in Table 2.6 (p. 37) and will focus the beam as shown in Fig. 4.26



**Fig. 4.25.** Refractive index modification of  $\text{CS}_2$  ( $n = 1.598$ ) by third-order non-linear interaction of a Gaussian beam of wavelength  $1.06 \mu\text{m}$  with a beam radius  $w_{\text{beam}} = 0.1 \text{ mm}$  and a power of  $10 \text{ kW}$  leading to self-focusing. For the calculation  $\gamma_I = 3 \cdot 10^{-18} \text{ m}^2 \text{ W}^{-1}$  was used



**Fig. 4.26.** Self-focusing of a beam in a third-order nonlinear interaction with matter. The refractive index profile will focus the beam and thus increase its intensity. Finally the divergence of the beam and the focusing compensate each other and wave guiding with a constant beam diameter takes place after propagating the length  $z_f$  which is the *focal length* of this self focusing

This focusing increases the intensity and thus the refractive index will be changed even more. The positive feedback for more and more focusing is limited by the increased divergence of the Gaussian beam with smaller diameter. After a certain length  $z_f$  an equilibrium between focusing and divergence is reached and the beam will propagate as in a waveguide with constant diameter  $d_{sf}$ .



As a hint for the order of magnitude of the self-focusing effect the critical power  $P_{\text{cr}}$  was estimated under different assumptions, e.g. of aberration-free focusing, to give

$$\text{critical power for self-focusing} \quad P_{\text{cr}} = \frac{\varepsilon_0 c_0 \lambda_{\text{inc}}^2}{4\pi\gamma}. \quad (4.109)$$

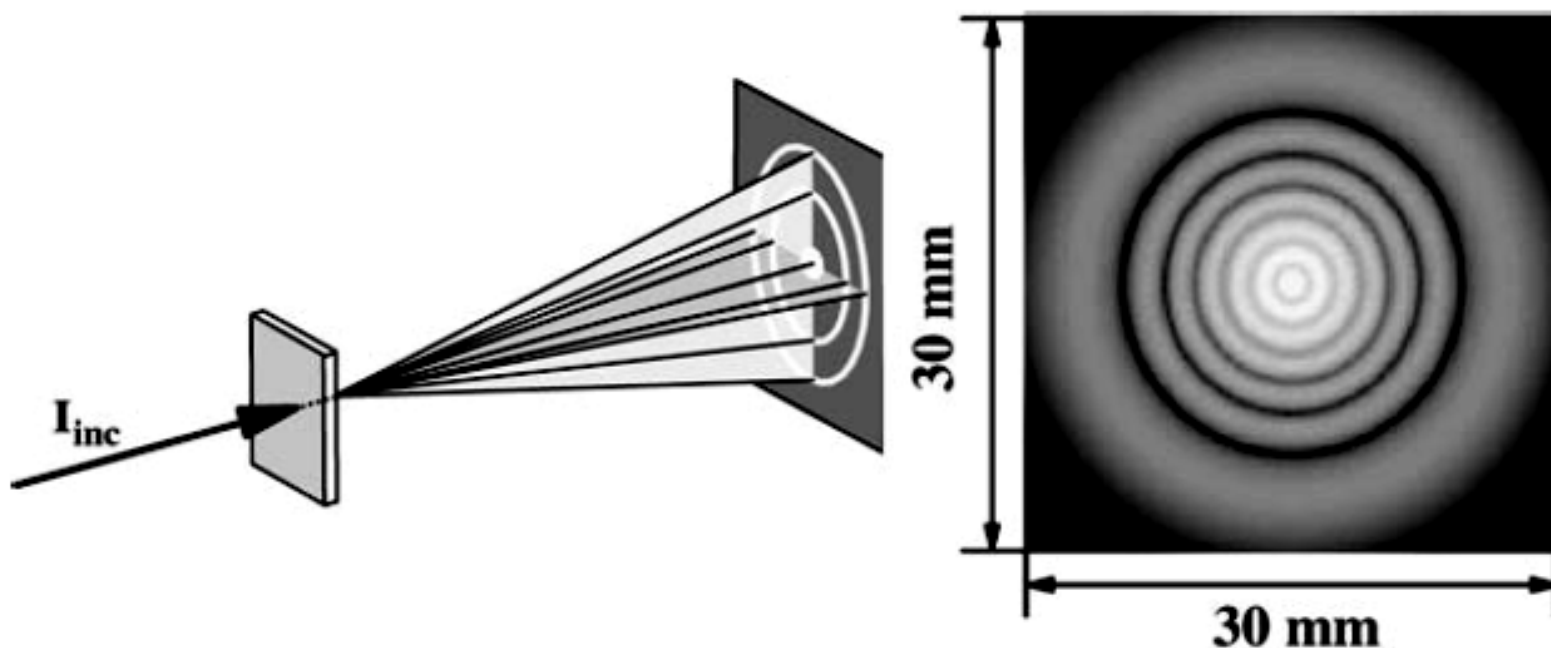
At this critical power an incident plane wave front will stay planar or in other words the incident beam will be waveguided with unchanged diameter. Thus this power gives *self-trapping* of the beam. The compensation of smaller divergence with larger beam diameter balances out the intensity effect and thus the power is the relevant quantity. At higher powers the incident beam will be focused with a focal length  $z_f$  which was estimated in the same way as  $P_{\text{cr}}$  to give:

$$\text{self-focusing focus length} \quad z_f = \frac{\pi w_{\text{inc}}^2}{\lambda_{\text{inc}} \sqrt{\frac{P}{P_{\text{cr}}} - 1}} \quad \text{for } P \geq P_{\text{cr}}. \quad (4.110)$$

This formula is derived for the beam waist positioned at the front surface of the material. For powers smaller than  $P_{\text{cr}}$  the focusing leads to a more or less negligible decrease of the divergence of the beam.

## 4.5.5 Self-Diffraction

If the refractive index change is produced in a thin slice of matter by a tightly focused laser beam a lens-like index profile will occur. At this induced refractive index profile the laser beam will be diffracted as shown in Fig. 4.28.



**Fig. 4.28.** Self-diffraction of a laser beam in a thin slice of matter resulting in a diffraction pattern behind the sample. The number of diffraction rings can be counted and thus the refractive index change determined. The schematic is on left and the calculated pattern at a distance of 0.5 m is on right. For details see text.

The resulting nonlinear change of the refractive index  $\Delta n_{\text{nl}}$  can be assumed to be proportional to the incident intensity. If the incident beam has a transverse Gaussian beam profile the resulting index change will show a transverse Gauss function, too.

The resulting far-field diffraction pattern can be calculated using Fraunhofer's integral. The phase of the emission pattern of the light behind the matter results from the local delay in the sample as a function of the local refractive index depending on the intensity profile of the laser beam. In this simple approach each phase shift  $\pi$  causes destructive interference and produces a dark ring. It turns out that the number of diffraction rings  $N_{\text{rings}}$  can be used to determine the maximum change of the refractive index in the matter  $\Delta n_{\text{nl,max}}$  at the center of the Gaussian beam for normal incidence as:

$$\text{maximum index change } \Delta n_{\text{nl,max}} = \frac{\lambda_{\text{beam}}}{L_{\text{mat}}} N_{\text{rings}} \quad (4.115)$$

with the wavelength of the light  $\lambda_{\text{beam}}$  and the thickness of the sample  $L_{\text{mat}}$ .  $N$  counts the number of  $2\pi$  phase shifts for the transmitted light from the center to the wings. It is of course a function of the intensity of the incident light.

Liquid crystals are especially favorable as a nonlinear material for these experiments because of their large nonlinear refractive index change

This huge optical nonlinearity in, e.g. nematic liquid crystals relies on reorientation of the molecules in the optical field. This process requires a certain electric field strength. Therefore the laser intensity has to exceed a certain threshold-like value in order to obtain self-diffraction rings.

The observed nonlinearity of liquid crystals is strong enough to obtain a diffraction pattern as shown in Fig. 4.28 (p. 217) at the right side with cw laser radiation in the 10 W range